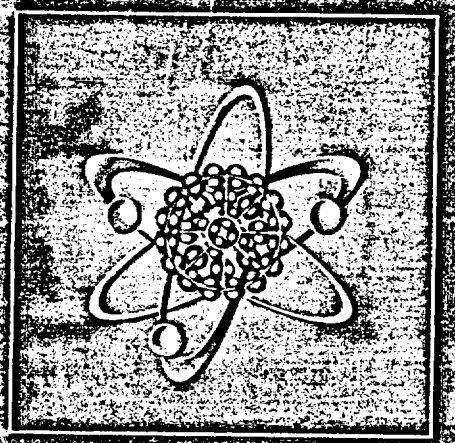
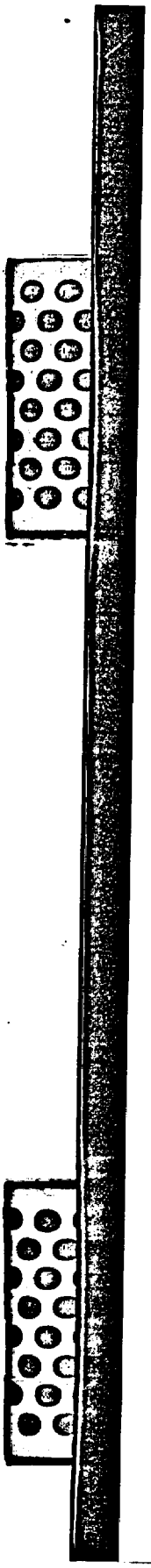


Canada Enters the Nuclear Age

A TECHNICAL HISTORY OF ATOMIC ENERGY OF CANADA LIMITED AS SEEN FROM ITS RESEARCH LABORATORIES



Canada Enters the Nuclear Age

A Technical History of Atomic Energy of Canada Limited as Seen from Its Research Laboratories

Written by sixteen of Canada's pioneering nuclear scientists, *Canada Enters the Nuclear Age* focuses on Canada's nuclear program at AECL's laboratories at Chalk River, Ontario, and Whiteshell, Manitoba, between the years 1943 and 1985. Topics include the organization and operations of AECL's laboratories, nuclear safety and radiation protection, radioisotopes, basic research, development of the CANDU reactor, and management of radioactive wastes.

Not only a valuable historical perspective on Canadian science, *Canada Enters the Nuclear Age* also provides useful guidance for innovative scientific development in the future, a future that will depend on developing and nurturing technically sophisticated industry.

AECL

R.A. (Bob) Speranzini
Ph.D.
General Manager
CANDU Technology Development

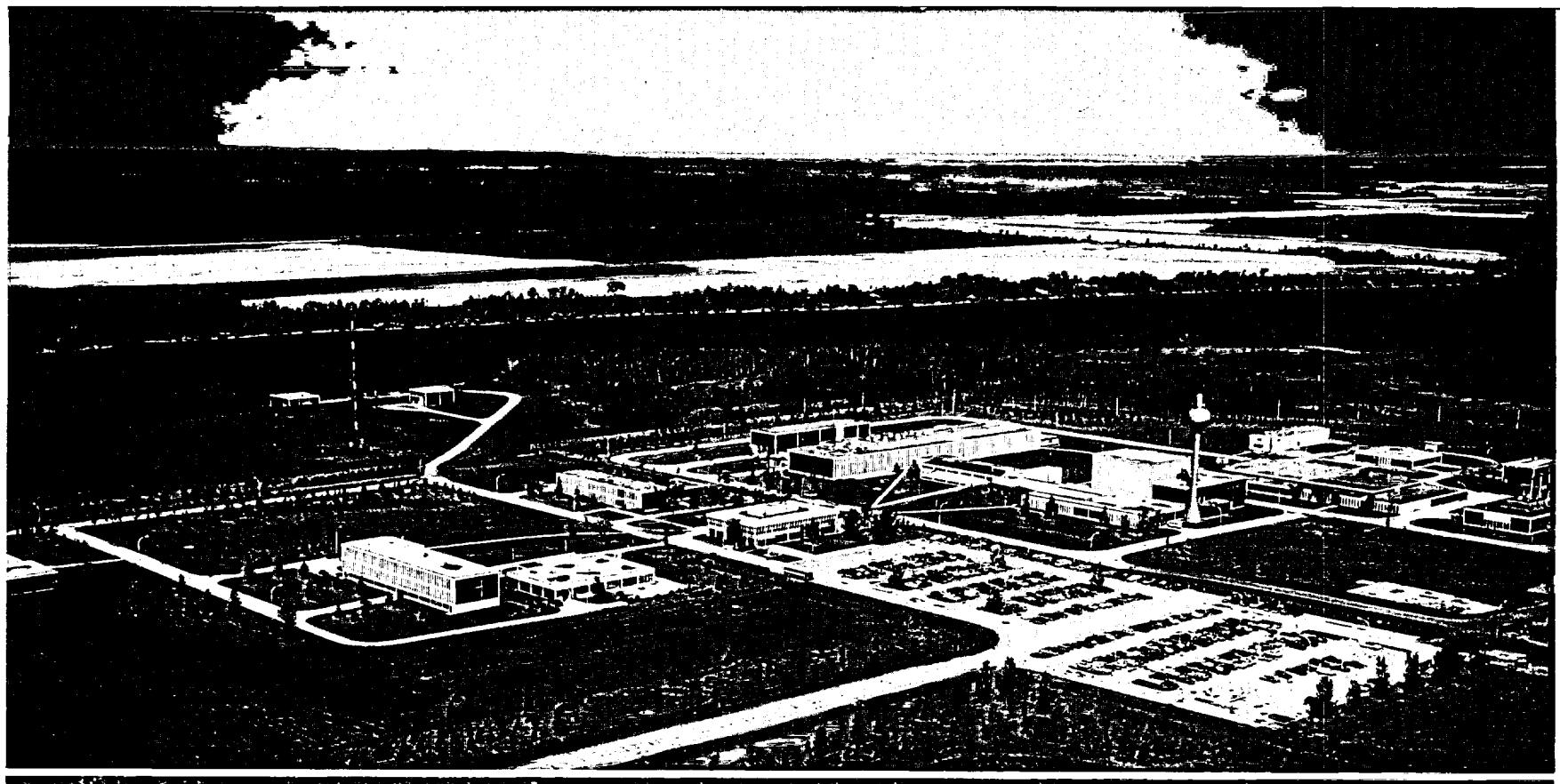
Chalk River Laboratories
Chalk River Ontario
Canada K0J 1J0
(613) 584-8811 + 3215
(613) 584-3311
Fax (613) 584-9569
speranzinir@aecl.ca
www.aecl.ca

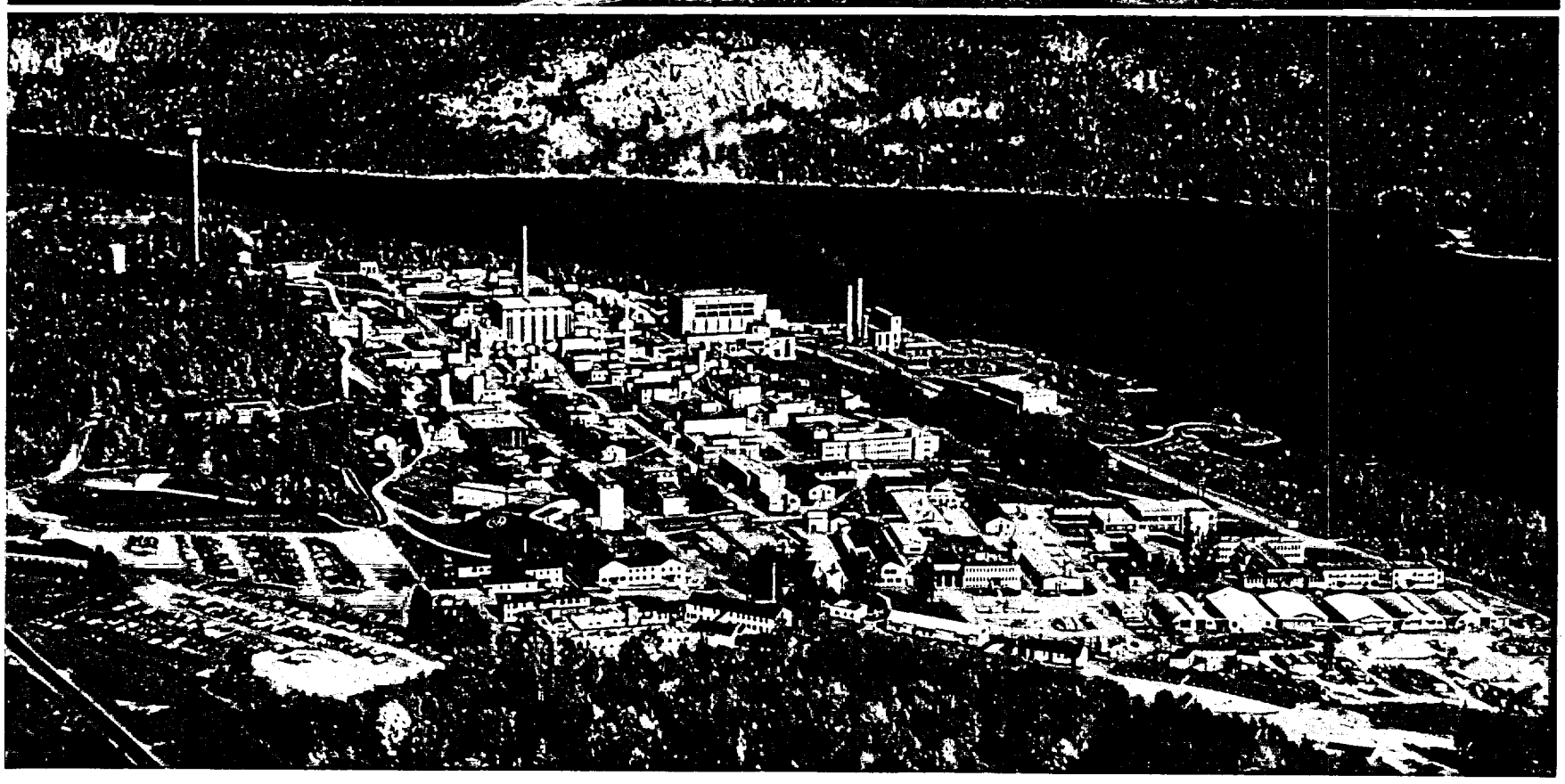
Canada

Atomic Energy of Canada Limited

Canada Enters the Nuclear Age

Overleaf: Whiteshell Laboratories
Chalk River Laboratories





Canada Enters the Nuclear Age

A Technical History of Atomic Energy of Canada Limited

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Contents

Foreword

As president of AECL in 1989, I was delighted when a number of retirees and elder statesmen came forward with a proposal for a technical history of the company. The time was opportune, they said, to produce a record of the research and development that had set the stage for Canada's notable contributions to the peaceful applications of nuclear energy. Not only was their enthusiasm inspiring, but as some of Canada's nuclear pioneers, their experience and knowledge made them the ideal authors of this chronicle. They have done an outstanding job in portraying the challenges of a unique and exciting quest to develop a complex and demanding technology.

The dawn of Canada's eminent achievements in nuclear development was almost a century ago, when New Zealand physicist Ernest Rutherford came to McGill University in 1898. He had just discovered that the radiation emitted by uranium and its compounds were of two types which he called alpha and beta rays. (A third type, gamma rays, was discovered by Paul Villard in Paris two years later.) These terms are still used. In 1901 Rutherford was joined at McGill by British chemist Frederick Soddy. Their brief partnership produced nine important publications which showed that radioactivity involves the transformation of one element into another.

Canada's more formal entry into the nuclear age came forty years later, in September 1942, when it was agreed that the heavy water and uranium oxide research that was being done at the Cavendish Laboratory in Cambridge, England, would be moved to Canada. And so began an era of intensive research and development that ultimately made a demanding technology into a safe, reliable and beneficial one.

The many facets of nuclear research and development are meticulously described in this book. Despite the modesty of the authors one can discern the impressive creativity, the tenacity, the intellectual capacity, and the dignity that underlay an achievement that would be marked with such accolades as a Nobel Prize and a place among Canada's top ten engineering accomplishments.

Many of the attainments of AECL's laboratories are significant milestones in the history of Canadian science. Not only that, they gave Canadian research global stature and provided world-leading products and services. It's a story that had to be written while the footprint of pioneering research and development is still conspicuous. It's the story of a venture that sprang from a vision, that persisted with a sustained sense of purpose, and that succeeded through dedication and talent. As a historical perspective it provides useful guidance for innovative scientific development in the future. The story deserved to be written and it deserves to be read.

Stanley R. Hatcher

Preface

The appearance of Robert Bothwell's book, *Nucleus: The History of Atomic Energy of Canada Limited*, which was largely a corporate and political history dealing with applied and industrial achievements, triggered interest in the preparation of a complementary technical history. In early 1989, Don Hurst, a former AECL employee and former president of the Atomic Energy Control Board (AECB), wrote a letter to AECL formally suggesting that such a technical history be prepared. The importance of timing was recognized since the main participants from the early days, whose recollections would be of great value, were disappearing from the scene.

AECL approved and sponsored the project, assigning the task of co-ordinating the preparation of the history to an employee, Gene Critoph (who retired in 1992 but maintained a coordinating function).

A Technical History of AECL Committee (THAC) was established to oversee the project. Over its lifetime THAC has had the following membership:

Members:

AECL retirees: A.S. Bain, J.H. Collins, E. Critoph (chair), M.F. Duret, T.A. Eastwood, C.E. Ells, R.E. Green, G.C. Hanna, R.G. Hart, A.M. Marko, J.C.D. Milton, D.K. Myers, H.K. Rae, J.A.L. Robertson, B. Ullyett.

AECB retirees: F.C. Boyd, D.G. Hurst.

Corresponding members:

G.L. Brooks (AECL retiree), D. Evans (Nordion), M. Tomlinson (AECL retiree), N.H. Sagert (AECL).

THAC held the first of more than a score of meetings in 1989 November.

THAC created targets for the end product of the project, including i) that the book be suitable for an audience of educated laypersons with special interests in science, and ii) that it give a balanced treatment of the various technical activities within its scope. THAC established an outline for the book that included a detailed list of suggested topics for each section. Each member of THAC then undertook responsibility for the preparation of part of the book, and provided valuable criticism to other authors on their contributions. THAC, as a whole, reviewed successive drafts of each submission. Over the period 1991-92 a consultant, D. Peirce of P.J. Spratt & Associates, was engaged to assist any author requesting help in improving comprehensibility, readability and interest for their contribution. THAC made numerous decisions regarding the style and structure of the book. Examples of the more controversial decisions were: i) to limit to very few the names that would appear in the text, in the interest of fairness to all, and ii) to keep formal references to an absolute minimum to improve readability.

In general this history extends from the late forties to 1985. The year 1985 seemed appropriate as a nominal closing date in view of the major restructuring that was implemented in 1986. Where significant events have occurred in the ensuing nine years, these are treated briefly.

The point of view adopted for this book is a research-and-development (R&D) one, with the main focus on the role of AECL's laboratories at Chalk River and Whiteshell in the Canadian nuclear program. Therefore the non-R&D aspects of AECL's role are not fully treated, and other parts of the nuclear industry are mentioned only inasmuch as they affected AECL.

The relative geographical isolation of the AECL research laboratory sites and the nature of the work being done required both a high degree of self-sufficiency for AECL Research and its operation as an integrated team. AECL's success rests on contributions from thousands of individuals from numerous disciplines – designers, fabricators, constructors, operators, researchers, technologists, regulators, development engineers and many others.

It was recognized that not all the R&D that was done could be recorded in a book of practical size. The authors of the individual chapters accept responsibility for content, but have, in general, sought the views of former colleagues. While it is hoped that readers will go through the entire book, many may wish to concentrate on particular sections. Nevertheless, everyone is urged to read chapter one, which provides an overview and a context for the more detailed chapters that follow, and also chapter twenty-one, the retrospective on learning from history.

Many people have contributed to the preparation and production of this volume. In addition to the specific contributions to the actual writing of the book that are acknowledged by the authors within their own sections, we collectively acknowledge the support freely given by the many AECL staff members who were approached for help. In particular we would like to acknowledge our debt to the staff in Technical Information Services, the Scientific Document Distribution Office, Central Records, Business Services and Corporate Relations. We would also like to specifically acknowledge the great contributions of Natalie Bellavance and Cathy Bennett in the difficult task of typing many drafts and the final manuscript, and McGill-Queen's University Press for bringing this book to fruition.

J.L. Gray

W.B. Lewis

Authors

ALASTAIR S. BAIN, B.A.Sc. (British Columbia)
Al Bain joined AECL in 1951 and spent the next twenty-eight years at the Chalk River Laboratories. His work included development of fuel rods for the NRX reactor, pyrometallurgical reprocessing of metallic fuel rods, and set-up of research hot cells. As head of the Fuel Development branch he was responsible for experimental work on CANDU fuel components and development of CANDU fuel bundles. He is author or co-author of about 200 documents on nuclear fuel. In 1979 Mr. Bain moved to Ottawa to work with a group newly formed in AECL to transfer technology under license to Canadian and foreign organizations. He retired in 1991.

FREDERICK C. BOYD, B.A.Sc. (Toronto)
After graduating in 1949 Fred Boyd worked on radioisotope applications with Eldorado Mining and Refining and then on the first CANDU plant with Canadian General Electric. In 1959 he joined the Atomic Energy Control Board. As head of Nuclear Facility Licensing he developed the Canadian Siting Guide, a foundation document in nuclear plant licensing. From 1972 to 1978 he was nuclear energy advisor in Energy, Mines and Resources. After a period as an International Atomic Energy Agency advisor to Korea's Ministry of Science and Technology, he rejoined the AECB as Orientation Centre director. He was admitted as a fellow of the Canadian Nuclear Society in 1994.

EUGENE CRITOPH, B.A.Sc., M.A.Sc. (British Columbia)
Eugene Critoph joined AECL in 1953 and worked in the field of reactor physics until 1967. Subsequently he held a number of increasingly responsible scientific and management portfolios within the laboratories. His senior positions included director of Fuels and Materials, director of Advanced Projects and Reactor Physics, and vice-president and general manager of the Chalk River Laboratories. In 1986 he was appointed vice-president of Strategic Technology Management for AECL Research. Also in 1986, in recognition of his significant contributions to nuclear research, he was awarded the Canadian Nuclear Association's W.B. Lewis Medal. It was under Mr. Critoph's leadership that this volume was developed.

MAURICE F. DURET, B.Sc., M.Sc. (Queen's), Ph.D. (Toronto)
Maurice Duret joined the Reactor Physics branch of AECL at Chalk River in 1952. When this branch was later split into two parts, he was appointed head of the part that became the Applied Mathematics branch. In that position he was responsible for developing analytical methods including theoretical aspects

of reactor physics and applied mathematics problems in areas such as fuel element modeling to predict cladding strains and stress analysis. His collaboration with international organizations included membership of the OECD Nuclear Energy Agency Committee on Reactor Physics and a major study for the World Energy Conference on the contribution of nuclear power to energy supplies.

T. ALEXANDER EASTWOOD, B.A., M.A., (Western Ontario), Ph.D (McGill), D.Phil. (Oxon)
Alex Eastwood joined the pioneers working on methods for the production of radioisotopes at Chalk River in 1947. This was his introduction to nuclear chemistry, the subject of his subsequent research career. His particular interests were the study of the decay properties of radionuclides and of unusual nuclear reactions in the reactors, and the determination of neutron capture cross-sections. He served as head of AECL's Research Chemistry branch from 1965 to 1967, assistant director of the Chemistry and Materials division from 1967 to 1970, and director of the division until his retirement in 1985. Dr. Eastwood is a Fellow of the Chemical Institute of Canada and an Emeritus Member of the American Chemical Society.

CHARLES E. ELLS, B.A.Sc., M.A. (Toronto), Ph.D. (Birmingham)
Charles Ells spent four years of World War 2 with the RCAF, obtaining the Burma Star from attachment to the RAF during the campaign in the Imphal region of Assam. Prior to obtaining his Ph.D. he worked on nuclear metallurgy at the Bureau of Mines in Ottawa and at Chalk River. After a period with Canadian Westinghouse he returned to Chalk River in 1957. Here his work was associated mainly with the development of CANDU fuel channel components. After retirement from full-time employment with AECL in 1990, he was appointed as Researcher Emeritus at Chalk River for a further four years.

RALPH E. GREEN, B.Sc., M.Sc. (Dalhousie), Ph.D. (McGill)
Ralph Green joined AECL at the Chalk River Laboratories in 1956, working first in reactor physics at Canada's first research reactor ZEEP, then in accelerator physics, and later as head of the Reactor Control branch for eight years. In 1979 he transferred to AECL's head office in Ottawa as a senior advisor to the executive vice-president of AECL Research Company. In 1981 Dr. Green was appointed acting general manager of the Whiteshell Laboratories. The following year he became vice-president and general manager of the Whiteshell site: In 1986 he was appointed vice-president of Reactor Development, responsible for all reactor-related R&D in AECL Research.

GEOFFREY C. HANNA, M.A. (Cantab.), F.R.S.C.
After graduation Geoff Hanna joined the British Ministry of Supply as a physicist working on radar development. He came to Canada in 1945 as part of the United Kingdom mission first in Montreal and later at Chalk River. His early research included the pioneering use of high-gain proportional counters to study low-energy radiations, investigations of the properties of transuranic nuclides, and measurements of nuclear data for reactors. He was head of the Nuclear Physics branch, director of the Physics division, and director of Research. He has an honorary D.Sc. from McGill University, and was president of the Canadian Association of Physicists in 1984-85.

ROBERT G. HART, B.Sc. (Toronto)
Bob Hart came to Chalk River Laboratories in 1948 and embarked on a notable career. After working on various projects including purification of heavy water in a reactor system, reprocessing of nuclear fuels and studying the physical properties of these fuels, he was transferred to Whiteshell Laboratories in 1965 as head of the Reactor Core Technology branch. He was appointed director of the Applied Science division in 1969, managing director of the Whiteshell site in 1973 and a vice-president of AECL in 1974. In 1978 he became executive vice-president in charge of AECL research. Mr. Hart was awarded the W.B. Lewis medal by the Canadian Nuclear Association in 1981.

DONALD G. HURST, B.Sc., M. Sc., Ph.D. (McGill), F.R.S.C.
Donald Hurst's career in nuclear energy began with post-doctoral work in the United States and in England. In 1939 he joined the National Research Council in Ottawa. He moved in 1945 to Chalk River, where he subsequently became director of the Reactor Research and Development division and director of Applied Research and Development. In 1970 he became president of the Atomic Energy Control Board. After retirement his activities included a period as chairman of the Senior Advisory Group of the International Atomic Energy Agency. Dr. Hurst received the Canadian Nuclear Association's W.B. Lewis Medal and Outstanding Contribution Award, and the American Nuclear Society's Tommy Thompson Award for outstanding contributions to nuclear safety.

ARTHUR M. MARKO, B.A. (Saskatchewan), M.D., Ph.D. (Toronto)
Following postgraduate research at the National Institute for Medical Research in London, England, Art Marko became a researcher and later professor of chemistry and paediatrics at the University of Saskatchewan. He came to Chalk River in 1961 as assistant director of AECL's Biology and Health Physics division, and became

division director in 1965. Dr. Marko has made outstanding contributions to molecular biology, radiation protection, and effects of radiation on living matter. He served on the executive council of the International Radiation Protection Association and has been a medical advisor to the Atomic Energy Control Board, as well as chairman of its Advisory Committee on Radiological Protection.

J.C. DOUGLAS MILTON, B.Sc. (Manitoba), M.A., Ph.D. (Princeton), F.R.S.C.
Doug Milton joined AECL in 1951 and began a notable career as a nuclear physicist, including a leading role in the Tandem Accelerator Superconducting Cyclotron project. He was appointed head of the Nuclear Physics branch in 1967, director of the Physics division in 1983, and vice-president of Physics and Health Sciences in 1986. Upon his retirement he was named AECL's first researcher emeritus. A fellow of the Royal Society of Canada and of the American Physical Society, Dr. Milton has served on many external committees and has been a visiting scientist at several institutions including the Lawrence Berkeley Laboratories and the University of Strasbourg. He was president of the Canadian Association of Physicists in 1992-93.

DAVID K. MYERS, B.A., M.A. (Toronto), Ph.D. (Amsterdam)
After post-doctoral studies in pharmacology and biochemistry at the University of Amsterdam, David Myers joined the Defence Research Board at Suffield, Alberta, in 1956. Two years later he joined AECL's Radiation Biology branch at Chalk River and was appointed head of that branch in 1976. In 1973-74 he was on attachment to the Institute of Medicine, Kernforschungsanlage (KFA), West Germany. Dr. Myers has been a prolific author in the fields of biological effects of ionizing radiation and the assessment of radiation hazards to humans. His professional affiliations have included the Radiation Research Society, the New York Academy of Sciences and the Canadian Radiation Protection Association.

HOWARD K. RAE, B. Eng. (McGill), Ph.D. (Princeton)
Howard Rae joined Chalk River Laboratories in 1950 and pursued chemical engineering research for twenty-two years, making major contributions in the areas of irradiated fuel processing, heavy water process development, and reactor chemistry. His management positions included head of the Chemical Engineering branch, director of the Fuels and Materials division, director of Applied Research and Development, and vice-president of Radiation Applications and Isotopes. He was chairman of the Advisory Committee to the Canadian Fusion Fuel Technology Progam. Dr. Rae was president of the Canadian Society for Chemical Engineering

in 1984-85 and received the society's major honour, the R.S. Jane Memorial Lecture Award, in 1978. He also received the Canadian Nuclear Association's Outstanding Contribution Award.

J.A.L. (ARCHIE) ROBERTSON, M.A. (Cantab.), F.R.S.C.
Archie Robertson served to captain in the Royal Engineers (1943-47). Following graduation he spent seven years on nuclear-fuel research at the United Kingdom Atomic Energy Authority's Harwell Laboratory. In 1957 he came to AECL, where he held positions that included director of the Fuel and Materials division, assistant to the Chalk River site head and director of Program Planning. Since retiring in 1985 he has been a consultant to the Ontario Nuclear Safety Review, the Atomic Energy Control Board, Ontario Hydro and the Auditor General of Canada, among others. He has been awarded the Canadian Nuclear Association's W.B. Lewis Medal and the Kroll Zirconium Medal.

BERNARD ULLYETT, B.A.Sc. (Toronto)
After a three-year period in Quality Control with Bell Telephone Bernard Ullyett joined Eldorado's radium operations at Port Hope. In 1952 he became supervisor of Radioisotope Operations at Chalk River and moved to the new Commercial Products Division (CPD) in Ottawa in 1955 as head of Radioisotope Operations. He subsequently established a product quality control program and became divisional manager for Operations Coordination. In 1975 he was posted to France as liaison engineer when CPD contracted with CGR.MeV (France) to produce medical linear accelerators. He was appointed project engineer for the new radioisotope processing operations at Kanata in 1978. Later he became a consulting engineer.

INTRODUCTION AND OVERVIEW

Chapter One

Overview of Nuclear Research and Development

D.G. HURST

INTRODUCTION

Canada is one of the very few nations that have successfully developed economical nuclear power plants, an achievement by no means to be expected, considering the small technological base of Canada's pre-war industry.

A fragile chain of events beginning in 1939 led to the establishment of a major nuclear laboratory in Canada. Under good leadership the laboratory acquired world recognition in science and initiated the CANDU®[1] heavy-water nuclear power system which by 1988 was generating 15 percent of the electrical power used in Canada. Canada currently produces a large fraction of the world's commercial trade in bulk radioisotopes.

This book records the scientific and technical advances that underlie Canada's nuclear achievement, discusses the associated planning and strategies, and considers some of the lessons learned. The story is being told by some of the scientists themselves, and the book covers the years 1939 up to 1985.

Background

Nuclear science was not entirely new to Canada. At the beginning of the century, while in Canada as a professor at McGill University in Montreal, Ernest Rutherford, the outstanding nuclear physicist of his time, made several of his famous discoveries. He showed, for example, that radioactivity is in many cases a manifestation of the long-sought transmutation of elements. After Rutherford returned to England, nuclear research was continued in Canada mainly at Queen's

1 CANada Deuterium Uranium; registered trademark.

A NUCLEAR PRIMER

All substances consist of *atoms*. Atoms are the smallest amounts of a substance that enter into chemical reactions.

A substance in which all atoms are practically indistinguishable by ordinary chemical means is called an *element*. Common examples are carbon, oxygen and iron.

An atom consists of a *nucleus*, which carries a positive electric charge, surrounded by *electrons*, each of which carries a negative electric charge.

Atoms have diameters in the order of 5/100,000,000 cm and nuclei about 1/50,000 times smaller. A trillion atoms can make only a microscopic speck of dust. Except for a fraction less than 1/1800 contributed by the electrons, the mass of an atom resides in its nucleus.

A nucleus consists of *protons* which are particles having a positive charge, and *neutrons* which are uncharged particles having about the same mass as a proton; since the nucleus of ordinary hydrogen is a proton, (the exceptional case of a nucleus without neutrons), all three – proton, neutron, and hydrogen atom – have very nearly the same mass.

The charges on electrons and protons are of equal magnitude but opposite signs. The magnitude is the fundamental unit of charge. An atom is electrically neutral, therefore the number of electrons is equal to the number of protons and this number is called the *atomic number*. The total number of protons and neutrons in an atom is called the *atomic mass number*.

All the atoms of an element have the same atomic number but may differ in atomic mass numbers because of differing numbers of neutrons. For example, the element lithium is atomic number 3 but has atoms of mass numbers 6 and 7. These two kinds of lithium are called *isotopes* of lithium and occupy the same place in the Periodic Table of the elements. An isotope is usually designated by its mass number and the abbreviation for the element. An isotope is nowadays called a *nuclide* when the relation to its sister isotopes is not involved.

Differences in the neutron content of isotopes have such slight effects on chemical properties that isotopes are almost indistinguishable in ordinary chemical reactions. However, small chemical and physical differences may be exploited by appropriate but expensive means to separate isotopes. Isotopes differ greatly in their nuclear properties.

The binding of the electrons to the nucleus is weak compared to the binding of the protons and neutrons in the nucleus; because of this, energies associated with nuclear changes are generally many times the energies associated with changes in the arrangement of the electrons. Consequently, our everyday experiences with materials result from interactions of electrons, not from direct nuclear effects.

Most of the nuclei that exist in nature are stable. With minor exceptions, the few unstable nuclei are either survivors from the primeval genesis of the solar system or are unstable descendants of such nuclei; examples of nuclides having unstable nuclei are uranium-238 and a descendant – radon. Nuclei with proton/neutron combinations that differ from the normal can be produced artificially, for example by bombardment with very energetic particles, but they are unstable.

Unstable nuclei transform to stable nuclei by what is called radioactive decay, giving off high energy radiations in the process – beta particles (fast moving electrons), alpha particles (fast moving helium nuclei), or gamma rays (high energy photons). Radioactive decay occurs randomly in time but in such a manner that the nuclide has a well-defined half-life. A free neutron is unstable and decays to a proton and a beta particle.

A different kind of nuclear instability occurs when an atom like uranium-235 captures a neutron. The resulting nucleus has a great excess of energy and may instantaneously break up into two parts of comparable size and some neutrons i.e., it *fissions*.

The first indication of the existence of radioactive decay was in 1896 when radiation from naturally occurring unstable elements was detected. Fission was first announced at the beginning of 1939.

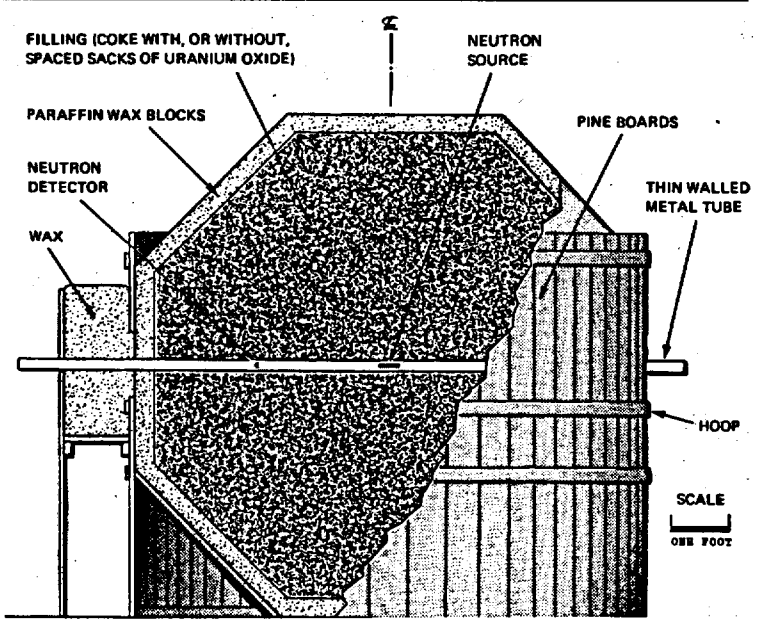
While the Pétain government was taking control of France, the heavy water was removed to Cambridge, England, in the custody of Halban and Kowarski. There they made measurements which showed that a mixture of heavy water and uranium could sustain a chain reaction. Wartime conditions severely restricted the support that could be diverted to further the work at Cambridge. The American nuclear project was advancing rapidly and the main British nuclear-weapons team would soon be moved to the United States. However, proposals to transfer the heavy-water group to the United States withered in the climate of tight security and growing American exclusivity.

The dilemma was solved by Canada's agreement to supply a laboratory for a joint Canada/United Kingdom/United States project. Major factors contributing to the solution were: pioneer work at the National Research Council of Canada (NRC), in Ottawa, by G.C. Laurence, who was the first person in the world to study neutron multiplication in a large assemblage of carbon and uranium (see figure 1.1); Halban's interest in being near the United States; the ready acceptance and promotion of the project by C.J. Mackenzie, NRC president, who foresaw that Canada would thereby have a head start in a field with major peacetime applications, including a new source of energy; and, perhaps, Canada's position as a uranium supplier.

In 1942, the heavy water and many members of the Cambridge group were sent to Montreal, where they were joined by several Canadian scientists in a laboratory that was initially set up in a large house, but which soon moved to the University of Montreal. Halban was director. For more than a year the Montreal Laboratory's activities included measurements and theoretical analyses of nuclear properties of heavy water, of mixtures of heavy water and uranium, and of assemblies of graphite and uranium. Radiochemistry, reactor design and instrument development were also actively pursued. There was, however, no clearly defined mission. This lack of a mission, dissatisfaction with Halban as director, and uncertainties about the laboratory's future as British/American cooperation waxed and waned undermined morale.

Early in 1944, as a result of decisions made in August 1943 by Prime Minister Churchill and President Roosevelt at the Quebec Conference, the Montreal Laboratory was assigned the task of developing the heavy-water reactor system beyond the stage expected to be reached by a 300 kW heavy-water reactor nearing completion at the Argonne Laboratory in the United States. (The Americans were arranging to produce heavy water; in particular, they had arranged for production of approximately half a tonne of heavy water a month from electrolytic hydrogen produced by the mining firm, Cominco, at Trail, British Columbia.) Specifically, the project was to design, build and operate a 10 MW heavy-water-moderated, natural-uranium-fueled reactor, to be named NRX. The reactor would provide neutrons to be used in research and produce plutonium-239, a fissile isotope of a new element plutonium, by capture of neutrons in the uranium-238 of the fuel. An auxiliary purpose was to produce, by neutron irradiation of thorium, the fissile isotope uranium-233, which does not occur in nature. These fissile products of reactors were initially sought as material for nuclear weapons.

FIGURE 1.1
Cutaway drawing of Ottawa sub-critical assembly, 1941–42



The heavy water would be held in a vertical cylindrical tank 8.75 feet (2.67 m) in diameter and 10.5 feet (3.2 m) high with 198 tubes passing through it. A tank of this type is called a calandria (see figure 2.14). The uranium would be metal in rod form, sheathed in aluminum and cooled by ordinary water. Each sheathed rod would be inside a calandria tube. The calandria tubes would be vertical and arranged in a hexagonal array. In reactor jargon, a regular arrangement of fuel and moderator is called a lattice.

The Americans were successfully pursuing the graphite reactor route for plutonium production on a schedule that promised availability before the expected end of the war. The impossibility of large-scale production of heavy water within that time made heavy water a backup or a long-term alternative to graphite for plutonium production. The heavy-water system could usefully be developed in Canada without significantly affecting the main thrust of the American effort, and it gave promise of peacetime uses.

At the end of April 1944, Halban was replaced as director by J.D. Cockcroft, under whose able direction the work of the laboratory and of associated organizations involved in the major expansion of the program progressed well. By mid-August a site was chosen on the Ottawa River near the Village of Chalk River, Ontario, and construction began of what is today called the Chalk River Laboratories. A site was also chosen for the Town of Deep River to provide housing for the staff.[3] The work was centred on the NRX reactor, which had excellent built-in facilities for research and which would become extremely successful. Originally specified for operation at 10 MW with a design maximum power of 20 MW, NRX was operating at 40 MW by 1954 with a corresponding increase in neutron intensity.

Soon after his arrival, Cockcroft decided to build a low-power heavy-water reactor as a preliminary to NRX. A detailed proposal for this reactor, ZEEP (Zero Energy Experimental Pile), was put forward in August 1944 and Kowarski, who had recently arrived from England, was put in charge of its design and construction. The operating power chosen was one watt. ZEEP, which went critical on 5 September 1945, was the first reactor to operate outside the United States. It provided information on such matters as transient behaviour of heavy-water reactors in advance of NRX. Later, it served for many years as a testbed for measurements on nuclear properties of materials and on heavy-water/uranium lattices proposed for future reactors.

While NRX was still under construction Cockcroft was appointed head of the atomic energy establishment being built at Harwell, England. For nearly a year he shuttled back and forth between England and Canada, then in September 1946, W.B. Lewis was appointed director at Chalk River (formally the Atomic Energy Division of NRC). Later the tripartite partnership was dissolved, but cooperation was maintained with both the United Kingdom and the United States at levels that fluctuated considerably.

On 22 July 1947, the NRX reactor was brought into operation, completing a chain of events stretching from the Paris group's decision to obtain the Norwegian heavy water, through the hazardous travels of the material and the experiments with it, to the appointment of Lewis as director. Failure of any of these links would almost certainly have kept Canada from assuming a significant nuclear role, as there was no Canadian policy to enter into the nuclear-weapons race as there was in France, the United Kingdom, and the United States. Laurence's innovative work on a uranium-carbon assembly was not likely to have led to the Canadian government's setting up a major project, but it is to the government's credit that having had the heavy-water baby deposited on its doorstep, it took it in and nurtured it handsomely.

The NRX reactor was for several years the world's best research reactor. With NRX as its centrepiece, the Chalk River laboratory embarked on an expanding program of research and development, including the production of radioisotopes. These activities brought international recognition and gave Canada high status in nuclear matters; one outstanding success has been the development of the CANDU nuclear-power system.

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[3] There was no scientific basis for the local story that the Chalk River site was chosen because heavy water collects in the deep portions of the Ottawa River from which Deep River gets its name.

Organization of the Canadian Atomic Energy Project

The tripartite Montreal Laboratory was part of the NRC organization. Canadian members of the staff were NRC employees; other members of the staff were on the United Kingdom team, which included many nationalities.[4] The United States took little part in the work of the laboratory, but it made available special materials, for example heavy water, and it provided much information on a need-to-know basis; a liaison officer was stationed at the laboratory.

When the decision was made to build a reactor, Defence Industries Limited (DIL), the wartime branch of Canadian Industries Limited, was brought in to handle the detailed design, operation and services of the project and the planned residential village; construction was by the Fraser Brace Company. On 1 February 1947 DIL withdrew and NRC took over the whole project, except that DIL continued to oversee the current construction. Fortunately, continuity was maintained because most of the DIL employees transferred to NRC. The Atomic Energy Control Act (1946) established the Atomic Energy Control Board[5] (hereafter called "Control Board"), with responsibility for atomic-energy matters in Canada. Instead of itself becoming directly involved in the Chalk River project, the Control Board asked NRC to continue to operate the project on behalf of the Control Board.

Preparation for Harwell, the United Kingdom establishment corresponding to Chalk River, became an important interest for the United Kingdom staff. Some Chalk River facilities were made available, and there was collaboration on chemical and chemical engineering aspects of plutonium separation. The move of personnel to Harwell began in the latter part of 1945, and by the end of 1947 most of the United Kingdom staff had returned to the United Kingdom or joined the Canadian team. For many years a few United Kingdom staff members were stationed at Chalk River for specific tasks and there were occasional postings of Chalk River staff to Harwell.

By the early fifties, a semi-industrial nuclear power program was emerging that was considered to be inappropriate for the research-oriented NRC. Accordingly, a crown company, Atomic Energy of Canada Limited (AECL), was incorporated and on 1 April 1952 it replaced NRC as the administrative body. There were then two boards in the line to the minister – AECL's board of directors and the Control Board. The revision of the Atomic Energy Control Act in 1954 cancelled this cumbersome arrangement and put AECL directly under the designated minister.

The members of the AECL board of directors were chosen because their backgrounds were relevant to the mission of the new company. For example, some were utility representatives, Ontario Hydro's being its chief engineer. The replacement of the NRC as the administrative body for the project by a company board of directors weighted towards industry was an important factor in promoting the practical utilization of nuclear power in Canada. However, the change from NRC to AECL had no immediate impact at the scientific and technical levels.

Soon after its formation, AECL acquired, by transfer from Eldorado Mining and Refining, a division located in Ottawa that was engaged in the commercial preparation and distribution of radioisotopes, and that was developing radiation-therapy devices of its own design, for which there was a worldwide market. In 1958, responsibility for the design and engineering of nuclear power plants was moved from Chalk River to a division newly established in Toronto for that purpose. Construction of a research and development laboratory at Whiteshell, Manitoba, was authorized in 1960. AECL became responsible for the marketing of CANDU reactors abroad, and when problems arose with heavy-water production plants it took over the operation of two such plants in the early seventies. Chapter two discusses AECL's organization, management style and operations, with a particular focus on the research side.

In 1978, four semi-autonomous companies were formed from the original AECL: a research company, an engineering company, a chemical company, and a radiochemical company, all

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[4] In August 1945, when the existence of the laboratory and its nuclear mission were made public, a Montreal newspaper reported that there were several "unclear" (sic) physicists on staff.

[5] In accordance with the proposed Nuclear Safety and Control Act, at the time of print Bill C23 (1996), the name will be changed to the Canadian Nuclear Safety Commission.

being branch companies under a corporate AECL management company. In 1979, an international company was added. In 1982, the engineering, chemical and international companies were consolidated into a new organization: CANDU Operations.

Over the more than four decades covered by this book, the principal organizations have had several changes of name. For simplicity the following informal names that have been in common use will generally be used.

- Montreal Laboratory: Established in late 1942 under NRC. Closed in 1946 with the transfer to Chalk River.
- Chalk River: At various times called Division of Atomic Energy of NRC, Atomic Energy of Canada Chalk River Project, Chalk River Nuclear Laboratories (CRNL) and, most recently, Chalk River Laboratories (CRL).
- Commercial Products Division (CPD) or Radiochemical Company: The Ottawa-based AECL organization responsible for the preparation, sale and distribution of radioisotopes and the design and manufacture of associated equipment – now separately incorporated firms Nordion and Theratronics International.
- Power Projects (later CANDU Operations, now AECL CANDU): The AECL organization based in the Toronto and Montreal areas, responsible for the implementation of AECL's nuclear power program, originally called the Nuclear Power Plant Division (NPPD).
- Whiteshell: the AECL Whiteshell Nuclear Research Establishment (WNRE) in Manitoba, now called Whiteshell Laboratories (WL).
- Head Office: In Ottawa. Now called the Ottawa office.

OVERVIEW

The tripartite decision to proceed with the development of the heavy-water reactor system as the principal mission of the Montreal Laboratory gave a great impetus to the laboratory's work early in 1944. The size of the staff was increased, effort was directed towards a specific reactor design, questions of reactor safety became realistic, and a site had to be chosen and developed.

Immediately after the plant site (Chalk River) was chosen, heat transfer and fouling tests with the river water were begun and were continued during the 1944-45 winter by engineers who were housed in summer cottages modified for winter habitation.

By the summer of 1945, construction at the plant site was well advanced and houses in the townsite (Deep River) were being occupied by staff members from Montreal and DIL. Their moves were made in step with plant and house construction, and by the middle of 1946 the Montreal Laboratory was closed.

Legacy of the Montreal Laboratory

Although the early work of the Montreal Laboratory had not been directed towards a specific mission, it provided experience and equipment that enabled the Montreal staff to begin work related to NRX without delay. Measurements of the nuclear properties of arrangements of uranium rods in heavy water were very soon underway to determine the parameters of the NRX lattice; the associated calculational methods had been the subject of much development in the earlier days. Measurements were done on heat transfer. The electronics group began work on electronic instrumentation for the new reactor and for radiation monitoring.[6] Ion chambers to be installed in the reactor as sensors of neutrons were developed; a related project was the production of quartz-fibre electroscopes suitable for personal radiation monitoring and other radiometric purposes. From this technology there came quartz-fibre microbalances for chemical studies of transuranic elements. A solvent-extraction process

[6] A hand-held monitor was called Pluto after the cartoon dog that went about sniffing. When General Groves, military head of the American project, learned of this, he demanded that the name be changed for security reasons - Pluto sounded too much like plutonium. A staff member proposed the name Lili. That name has been applied for many years to such instruments. The staff member had just been to the Gayety, a burlesque theatre, and seen Lili St. Cyr, a famous stripper. Use of the name Pluto for such instruments continued in the United States, presumably not having reached General Groves.

for plutonium separation was selected by the chemists after trials with hundreds of organic solvents.

Planning and design were undertaken for a charged particle accelerator of the Van de Graaff type, the first of a sequence of accelerators installed at Chalk River.

There was also research into fundamental properties of matter, some of which would have application in the design and use of nuclear reactors; the chemistry of uranium and plutonium (the latter limited by availability) was studied, as were the nuclear and fissioning properties of these elements. An important result was a measurement of the yields of several radionuclides produced by uranium fission; the resulting mass-yield curves for fission of uranium-235 were published in 1946. A new type of spectrometer for measuring beta-ray energies had been developed by a staff member in his graduate years. Spectrometers of this type were later used at Chalk River and in many Canadian university physics departments for much physics research. Radiolytic chemistry of water was important from the early days of the project. Chapter six describes AECL's commitment to basic research, and chapters seven and eight deal with chemistry and physics, respectively.

The medical use of radium and X-ray equipment in the first few decades of the century had resulted in severe exposures and therefore severe health effects to some users and patients. By the mid-forties, there was some quantitative knowledge about such matters. An important component of planning for NRX was preparation to limit exposures to radiation by providing adequate shielding, appropriate work procedures, and monitoring of the exposures.

The monitoring of radiation doses to workers and the public, the determination of dose rates in the environment and in the workplace, and the surveillance of waste management repositories are all essential aspects of radiation protection. Chapter three deals at greater length with this subject.

ZEEP[7] proved to be a very useful low-power reactor, enabling low-power measurements to be made for many purposes. These included the measurements of lattice parameters, transient effects and effects of various materials on reactivity (reactivity is a measure of the ability of the reactor to sustain a chain reaction); graphite intended for the first reactor at Harwell, England, was tested in ZEEP to ensure that it met specifications of low neutron absorption.

NRX Design and Safety

The design work by DIL was carried out in Montreal under the direction of the engineering group of the laboratory. Intense radioactivity of the reactor core during operation, and the residual radioactivity that persists after shutdown, introduced novel constraints on the engineering. To protect the workers the core was surrounded by thick shields of iron and concrete. Ensuring safe operation and making provision for handling accidents involving radioactive releases were priority aspects of the design; these added to the problems arising from inaccessibility of much of the reactor once it had operated at high power. Of special concern was the "boiling disease": loss of light water from the cooling channels by boiling would increase the reactivity and this would increase the rate of fissioning; the result would be more boiling, culminating in a power runaway if not stopped by rapid detection and counteraction of the increasing reactivity. Fast-acting shutoff rods were developed. These would be brought into action by evidence of reactor overpower, a rapid power increase, or reduction of cooling flow through a fuel channel. The design included so many shutoff activators that it was often stated that the reactor would never operate because there were hundreds of ways for it to shut itself off, but only one way to start it up.

Immediately surrounding the cylindrical wall of the calandria there is an annulus of graphite approximately three feet (90 cm) thick. Its purpose is to reflect back into the calandria neutrons

[7] The mechanical design of ZEEP was done at NRC in Ottawa under extreme secrecy, which forbade any public mention of the nuclear interest of the project. A security-minded visitor to the design office was appalled when a stranger looked in and asked, "Have you had your neutrons yet?" The visitor was somewhat mollified when he learned that there was a general staff NRC newsletter called "The Neutron," so named because, like neutrons, it was free of charge.

that escape through the side wall. Without this reflector the core of the reactor would have to be larger, to compensate for the loss of neutrons. The reflector and inner shield are air-cooled. The air is exhausted through a tall chimney, because some of the argon in the air is made radioactive by absorption of neutrons. In 1945, tests were run in which smoke discharged from the chimney under various wind and weather conditions was observed, to forecast the dispersion of the very weakly radioactive plume of effluent air.

NRX Research Facilities

Conceived in wartime but planned with an eye to peacetime uses, NRX incorporated many features intended to facilitate research. The advent of nuclear reactors increased the availability of neutrons by factors of many thousands over the most intense previous sources (i.e., accelerators). Work with those sources had, however, provided experience which, combined with lessons learned from one or two American reactors, enabled an excellent design for NRX to emerge. Horizontal holes through the graphite reflector and the shielding provide beams of neutrons to research equipment at the main-floor level. Access to the maximum neutron flux is available in a large vertical tube through the centre of the core (the so-called central thimble), or in the vertical tubes normally used for fuel. (The flux is sometimes said to be the number of neutrons crossing a square centimetre in one second; more precisely, it is the number of neutrons traversing a sphere of cross-sectional area 1 cm^2 in one second.) Two horizontal columns of graphite roughly two metres square extend from the graphite reflector through the iron and concrete shields to the outer perimeter, where they are closed with thick, shielding doors of lead. They are called thermal columns, because the neutrons at the outer ends are in thermal equilibrium with the graphite; that is, the energy distribution of the neutrons is similar to that of the molecules of a gas at the temperature of the graphite. The remaining area of the side wall of the reactor has special devices for irradiating many small samples, to produce radioisotopes. A ring of ninety vertical holes through the top shield enables tubes containing material for irradiation to be inserted into an annulus in the graphite reflector near the inner surface. These positions were originally intended for the irradiation of thorium to produce uranium-233, but some were soon being used to produce other isotopes; e.g., carbon-14, an isotope having many uses in biological research.

Chemical Extraction Plants

Uranium-233 and plutonium-239 were two products of NRX of major interest as fissile nuclides. Two heavily shielded chemical extraction plants were built near NRX, one to separate uranium-233 from the thorium carbonate that was irradiated in the annulus in the reflector, and the other to separate plutonium from the uranium and fission products in irradiated fuel rods. The chemical processes were new and the feed materials intensely radioactive; consequently, startup and operation were difficult. Nevertheless, both plants eventually delivered satisfactory output, but were later shut down as being not immediately relevant to the project's developing mission.

Waste Management

Operation of a nuclear reactor produces unwanted radioactive material, the safe management or disposal of which poses problems. The Chalk River site is in a sandy, rocky area not valuable for agriculture. Immediately adjacent to the plant site and within the restricted area is a sand-filled rock basin with a single water outlet, a creek flowing from a small lake called Perch Lake. From the beginning, Perch Lake basin has been used for radioactive waste management; the outlet water can be monitored for escape of the waste. Radioactive scrap from the repair of NRX in 1953, waste from the processing plants, and other radioactive wastes have been deposited in this area, usually in sealed containers. Research into the movement of radioactive material through the soil and other aspects of disposal of radioactive material have been pursued intensively. The movement through soil is very slow. Some underground tank storage was

provided near the chemical-extraction plants for storage of radioactive liquids.

Early Status of the Chalk River Project

By the time of NRX startup (July 1947), the tripartite organization had been disbanded and Canada had assumed full responsibility for the project, but with good connections to the United Kingdom and the United States (within the limits imposed by their security regulations). Excluding construction workers, there were approximately 500 staff members (including 140 scientists) and 500 hourly rate workers; there were also forty research and technical United Kingdom staff. During 1947, the Canadian total increased by about 10 percent and the United Kingdom total decreased by about 20 percent. The new town of Deep River, built to house many of the staff, was well established on the shore of the Ottawa River, eleven kilometres upstream from Chalk River.

NRX

The permitted power level of NRX was raised in steps until it reached 30 MW by mid-1950. The reactor was operated to produce uranium-233 and plutonium, to provide radioisotopes, and to provide beams of neutrons for physics experiments. A less obvious reason was to develop experience in a new, promising technology; for example, the intense ionizing radiation posed novel operating problems. There was as yet no clearly defined mission, but the underlying hope for a source of energy was an incentive. In 1948, the Control Board stated that the general policy was to operate the reactor in the best interests of fundamental research and not for the specific purpose of producing plutonium.

Early Research based on NRX

Gamma rays

When an atomic nucleus captures a neutron, the result is a new nucleus in a state with considerable excess energy, a so-called excited state. In most cases, the new nucleus very quickly gets rid of excess energy by emitting gamma rays. Measurement of the gamma-ray energies provides information about the energy levels and mass of the newly formed nucleus. Research on these neutron-capture gamma rays was a major component of physics research at NRX. Two different types of equipment were installed: one measured high-energy gamma rays, the other lower-energy gamma rays. Many important results were produced; for example, within about a year, it was shown that the accepted mass of a neutron was in error and a much more accurate value was obtained.

Radioisotopes[8]

The nuclei resulting from the capture of neutrons are often radioactive and therefore not usually found in nature. Reactors are prime sources of these nuclei, and NRX has been a prominent supplier. Within a few months, there was a steadily increasing stream of requests from both project staff and outsiders for radioisotopes. The radioisotopes were themselves objects of research or were useful as tracers in other research. Over the years, medical uses for research or clinical applications predominated. In 1946 an isotope-production branch was established to develop, produce and ship radioisotopes for outside users.

Neutron diffraction

For many purposes, the neutron is treated as a material particle, but, like all other matter, it also has a wave character. Slow neutrons (i.e., those having energies like the energies of molecules in a gas) exhibit wavelengths similar to the spacings of atoms in solids and liquids. Just as X rays having similar wavelengths are diffracted by crystals, so are slow neutrons. By such diffraction, neutrons of specific energies may be selected or the structure of the crystal determined. A neutron spectrometer was installed at

[8] The modern term is radioactive nuclide or radionuclide, unless the intention is to relate it to nuclides belonging to the same element. However, universal usage dictates the older term, radioisotope, for general commercial use.

the experimental face of NRX, where a neutron beam could be extracted. This instrument provided mono-energetic beams of neutrons for research on structures of gases, liquids and solids, nuclear properties of deuterium, scattering of neutrons, and fission cross-sections as a function of energy.

Decay of the neutron

The mass of a neutron is greater than the sum of the masses of a proton and electron. From the well-known relation due to Einstein, $E=mc^2$ (E is energy, m is mass and c is the velocity of light), it follows that the decay of a neutron into a proton and an electron is energetically possible, and indeed it had been predicted but not observed. Equipment was set up at an NRX beam hole to detect and study the decay. This was first observed elsewhere, but the NRX experiment yielded a value of 12.8 ± 2.5 minutes for the half-life. The identification and measurement of the protons and electrons in the presence of neutrons and other radiations required experimental design and analysis of a high order.

Fission

From the beginning, there was wide scope for basic research into such matters as details of the fission process, the properties of the fission products, their relative yields, and differences in yields between the various fissile nuclides. Neutrons, initially from a thermal column but later from a beam hole, were used in detailed studies of the energies of the fission fragments and of the emission of light nuclei in the fission process; another experiment showed that the fission neutrons are emitted from the moving fission fragments.

Transuranic elements

For some years the high neutron flux of NRX gave it a superior position for synthesizing the newly discovered transuranic elements through the neutron irradiation of uranium and the sequentially produced elements. The chemical and physical properties of these new elements were investigated at Chalk River, and, at a later stage, in cooperative experiments with other laboratories.

Reactor characteristics

Many experiments on arrangements of uranium and moderators had been done in the United States, in the Montreal Laboratory and in ZEEP, but they were with fresh uranium at very low power and at room temperature. Calculations of temperature coefficients of reactivity and other properties of operating reactors were based on measured or estimated basic properties of the constituents, but supported by few direct measurements. It was therefore important to determine experimentally such properties of NRX.

The first major measurements were done during 1948, when the power was being raised to 20 MW. Temperature and power coefficients of reactivity and other parameters were measured, a special case being xenon poison. The radionuclide xenon-135 has an enormous cross-section for the capture of slow neutrons, of the order of a million times the average cross-section. (The cross-section is the target area the nucleus presents for interaction with the neutron flux. There are several kinds of cross-sections, for example, capture, scattering or fission cross-sections. Each of these has its own form of dependence on the neutron energy. The rate of interaction per nucleus is the product of the cross-section multiplied by the flux.) Xenon-135 is the result of radioactive decay of iodine-135, which is a product of fission. The interplay of the rate of destruction of the xenon-135 by neutron absorption and the decay periods of the two radionuclides leads to complex variations of reactivity in a high-flux reactor. Shutdown of a power reactor may be followed by a day or two of forced inaction, until the xenon poison dies out by radioactive decay.

Interest in power reactors made knowledge of the effect of irradiation on the reactivity of fuel of prime importance. Experiments to determine this effect were done in October 1951, March 1952 and December 1952, with NRX as the measuring instrument. The last set of experiments was terminated abruptly by the 1952 NRX accident (which will be discussed later). The reactor was restored by February 1954 and the first 1,000 megawatt-days' operation (roughly six weeks) was done under conditions specified for a major experiment on reactor

characteristics. During this time, the reactor power was raised in steps to a new limit of 40 MW.

One of the long-irradiated rods measured in October 1951 was made the subject of a detailed examination that continued for many years, yielding results important for use in calculating the reactivity of power-reactor fuel. Eighteen thin discs were cut from the rod for metallurgical studies, chemical analysis, and measurement of radial distribution of various nuclides. Slugs were sent to an American laboratory for measurement of reactivity.

Future Systems Group

In December 1944, Cockcroft had formed a Future Systems Group, with a membership predominantly from the United Kingdom staff. Initially, the group considered reactors for the United Kingdom, with emphasis on reactors for plutonium production; production of uranium-233 and energy were secondary goals. Uranium was thought to be a rare element, with an ultimate availability of less than ten thousand tons. Only 0.7 percent of uranium, the uranium-235, is readily fissile. The remaining 99.3 percent, uranium-238, is practically inert insofar as fission is concerned; however, it can be converted to fissile plutonium by absorbing neutrons. Consequently, breeder reactors, which produce more fissile material than they consume, had high priority. Fast-neutron chain reactions with plutonium as fuel were known to be possible breeder systems, and there were indications that thermal-neutron reactors using uranium-233 as the fissile material and thorium to capture the excess neutrons and convert to uranium-233 might also breed. Fissioning of the precious uranium-235 would be necessary for the initial production of the plutonium or uranium-233 needed for the breeders.[9] (Breeding has recently been achieved in a power reactor but even yet it is not an accepted alternative to other power-reactor systems.)

The group's early discussions concentrated on fast-neutron plutonium-breeder reactors and gas-cooled thermal-neutron reactors moderated by graphite or beryllium oxide. Beryllium has low absorption for neutrons and, because it has lighter nuclei than carbon, seemed to offer superior moderation. The oxide behaves well at high temperatures, but otherwise little was known about its suitability. Gases considered for cooling included hydrogen, helium and carbon dioxide. Heavy water was not stressed as a possible moderator.

Information was sought on nuclear parameters of fuels and on such engineering properties as erosion of the moderators by hot gas, diffusion of fission products through them, and effects of intense irradiation. Action was taken to procure massive blocks of beryllium oxide.

In 1946, a "water boiler" was added to the list of possible reactors as a source of neutrons for experiments. The core would be a light-water solution of a plutonium compound contained in a stainless-steel vessel. Pulsing of the power would provide momentary neutron fluxes a hundred times the flux expected in NRX. The plutonium would have to be supplied by another reactor of a different type.

NRU, a Replacement for NRX

As a result of transfer of staff to the United Kingdom and the change in directorship from Cockcroft to Lewis, the Future Systems Group did not meet for nearly a year and a half. In November 1947, Lewis convened his first meeting of the group with a revised and enlarged membership to consider a replacement for NRX. The corrosion-limited life of NRX had been estimated as about five years and soon after NRX was in operation the Control Board had approved consideration of a second reactor to provide continuity in the event that NRX had to cease operation.

The reactor types initially chosen for study by the new group were very similar to those considered earlier. A reactor cooled and moderated by lithium-7 was added in spite of the formidable problem of separating lithium-6 from the lithium-7. In retrospect it is clear that none of the reactor types would have

[9] A remark that illustrates the importance given to the supposed shortage of uranium is recorded in the minutes of a 1946 Future Systems Group meeting: "Large-scale production of power is not likely to be economically interesting until breeding has been made effective."

been a good choice. In April 1948 a suggestion was made that consideration be given to an improved high-power heavy-water reactor. At the next meeting six weeks later, a heavy-water reactor with a larger neutron flux than NRX, good experimental facilities, good economy of neutrons and possibly some use made of the power, was said to be the tentative choice of the group. Lewis christened this reactor NRU, with the "U" standing for universal. A subcommittee was established to prepare technical outlines and rough cost estimates for four versions of a heavy-water reactor. In the spring of 1949 the version corresponding to NRU was selected for further study. A Reactor Development Panel was appointed to supervise the next stage of conceptual design of NRU. For twelve months the design concept was developed while the question of approval was moved closer to government. The cost of the reactor was a stumbling block, but the plutonium that NRU would produce was thought to be a valuable commodity that could be extracted and sold to the British or Americans. There was considerable negotiation, which culminated in the sale of the used fuel to the United States for its plutonium content.[10] This sale of intact fuel removed the need for a costly new chemical extraction plant at Chalk River. The American involvement not only made the project financially acceptable, but also ensured the availability of fuel and heavy water.

The NRU reactor, as proposed by the Reactor Development Panel, was to be moderated and cooled by heavy water, fueled by natural uranium, have fuel-changing under power, provision for experiments with neutrons (beam holes, through tubes) and several positions for fuel tests. The operating power was to be 200 MW; the idea of generating useful energy had been dropped. Government approval to proceed was given on 13 December 1950. The final design was done by a consulting engineering firm, C.D. Howe Co., with guidance from Chalk River.

Nuclear Power

The prospect of generating useful energy from the heat produced by nuclear fission helped get the Canadian project on the road and to keep it moving. Inevitably, some dead ends were encountered along the way. The Future Systems Group in its early consideration of plutonium-production reactors for the United Kingdom proposed converting some of the heat to electric energy. This anticipated the first British power reactors, which had a dual purpose: to produce plutonium for military purposes as well as electric power for civilian use.

When the revised group sought a replacement for NRX, it had studies done on proposals for useful energy generation. This was not pursued, but a steam engine project was begun to demonstrate that coolant from a loop in NRX at a temperature of 200°C could generate power "at a not insignificant efficiency". A reciprocating engine to run on steam was ordered in mid-1952, but a serious accident in NRX delayed the project to a time when it would have little value, as a demonstration of this kind had been made in the United States; also, power reactors were being designed. The project was cancelled.

Apart from keeping interest alive in nuclear power, the Future Systems Group contributed little to the birth of the Canadian nuclear power program. This program was conceived outside the activities of the group and began to take shape in 1951. Nevertheless, in July 1952, Lewis suggested that the group maintain action on a liquid-metal-cooled thermal neutron reactor and breeder reactors. About this time the idea arose to produce fissile material by the absorption in uranium or thorium of neutrons resulting from the spallation (breaking into smaller pieces) of heavy elements bombarded with high-energy protons; this idea was to be developed further over the next fifteen years.

By 1947 the United States navy had a program under Admiral Rickover to develop nuclear-powered submarines. Light water[11] and liquid metal were considered as coolants. Eventually the admiral vigorously promoted, as the heat source, a light-water-cooled and -moderated reactor fueled with enriched uranium, all contained in a pressure vessel. To test fuel for these reactors,

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[10] Bothwell, *Nucleus*, 139-43.

[11] The term light water is commonly used for ordinary water in contrast to heavy water, since 99.986 percent of the hydrogen in ordinary water is light (i.e., ordinary) hydrogen.

hydraulic circuits known as loops containing fuel elements and cooling water at temperatures and pressures suitable for power generation were installed in some American reactors. NRX offered superior conditions (higher neutron fluxes and larger in-reactor volumes), and in 1950 negotiations led to the use of positions in NRX for American loops. Many details of the fuel elements under test were American secrets, but what information was revealed was of great value to Canadian power planning, as was the experience with the technology of loops. Cooperation and competition between the United States and Canada on fuel technology increased over the years and proved extremely profitable for both parties.

The breakthrough in Canadian thinking on nuclear power came in 1951 when Lewis combined knowledge of the United States loop experiments with other facts. Proposing an elementary design of a heavy-water power reactor, he estimated the cost of production of electric power to be in the commercial range, thus for the first time indicating the economic viability of nuclear power.

In summary, the facts were:

1 NRX fuel had survived the production of 3,000 megawatt days per tonne and had retained much of its reactivity.

2 In loops, fuel was being cooled by water at temperatures and pressures like those in thermal power plants. Zirconium was a possible sheathing material in these conditions.

3 Fuel changing under power was planned for NRU.

The proposal fell on fertile ground and blossomed in the next few years. The engineering community had a general professional interest in nuclear power, but Ontario Hydro had a specific interest, because it was approaching the time when all suitable hydroelectric sites in Ontario would have been developed and new plants would be thermal; i.e., based on the burning of fuel. (Ontario has no indigenous practical sources of fossil fuel despite the fact that the first oil well on the continent was in Ontario.) At the beginning of 1952, there were discussions with Ontario Hydro about nuclear power and by the end of 1952, AECL and Ontario Hydro had moved towards cooperating on a nuclear power plant.

The NRX Accident, 12 December 1952

The program of the newly established AECL was progressing well in 1952. Research was producing important results, radioisotope production was increasing, NRU construction was underway, and nuclear-power planning was moving into an era of optimism. Then on 12 December, the NRX reactor was seriously damaged in an accident, a costly interruption to the work of the project.

Experiments had been in progress to measure the effect of irradiation of fuel on its reactivity. The reactor was being operated according to procedures that differed from those for normal operation. Somewhat similar procedures had been used successfully in a previous set of measurements. An operator error was the first of a sequence of events, a difference in any one of which might have prevented the serious consequences. There was a power runaway, enhanced by deliberately reduced cooling arrangements for certain rods. Steam explosions in several channels caused irreparable damage to the calandria. Central portions of several fuel rods disintegrated, releasing gaseous fission products to the atmosphere and becoming oxidized by contact with water and air.[12]

Removal of the heat generated by decay of fission products in the fuel required that a small flow of water be maintained through the core. The water, laden with highly active material, either reached storage tanks or leaked into the basement below the reactor building, where it accumulated to a depth just under a metre. All the active water was eventually piped to the waste-management area.

[12] On the basis of a preliminary telephone call from Chalk River, AECL's president in Ottawa told the press there had been a pinhole leak. When a group of Americans visited Chalk River to discuss the accident, one of them began his remarks by saying that Canadians must measure pinholes as they do gallons because this was certainly an imperial pinhole.

NRX Restoration

The damage to the reactor was so extensive that the feasibility of restoration was initially in doubt. However, after a brief study, a decision was made to proceed with restoration. Radioactive fission products released from damaged fuel rods were spread about in the calandria and the building basement, making disassembly difficult. Workers soon reached their radiation dose limits, which were then 0.3 rem per week (0.003 sievert per week). (A rem is a unit of radiation dosage. It is defined in terms of the energy deposited in tissue by ionizing radiation, with adjustment to take into account the biological effects of the specific radiation. A sievert is the Système International dose unit corresponding to a rem; one sievert = 100 rems.) The Canadian army and the American navy supplied extra workers. The latter had two incentives to participate: the NRX facilities were important in the development of fuel for nuclear submarines; and the United States navy had a group being trained to handle radioactive contamination resulting from an accident or a nuclear-bomb explosion. NRX gave them hands-on experience in a real situation.

A restored and improved NRX was brought to criticality on 16 February 1954, and in April of that year it was operating at 40 MW, a new upper limit made possible by a modification during the restoration.

A remark made soon after the accident, that it might be a blessing in disguise, was not fully justified, but it did have some validity: the restoration itself was an encouragement to the nuclear community; major benefits were (i) the renewal of strong concern for safety among staff members, many of whom became prominent in the nuclear power program, (ii) improvements in safety systems planned for NRU or being developed for power reactors as a result of reviews prompted by the accident, and (iii) the demonstration that a reactor could be repaired after an accident.

Space in the reactor for loops, and accommodation in shielded rooms for auxiliary equipment were in demand by Canadian, American and British researchers. An example of actions taken to satisfy the demand for space for auxiliary equipment was the move of the gasholder for the moderator cover gas, helium, to outside the building, to make available the pit in which the gasholder was originally located.

An increasing diversity of NRX usage required frequent changes in the operational program. In the early days, only a weak link was needed between the operational timetable and the plans of researchers (who, in the main, were using neutron beams), because changes could be made to the research equipment whether or not the reactor was operating. The advent of loops introduced a need for close coordination of reactor and loop timetables.

Chemistry and physics research using neutron beams and radionuclides from NRX had also expanded and was producing first-class results. The value of NRX to Canada was at its peak during this interval and it was to continue at a high level for at least a further twenty years.

NRU

Even with the NRX experience as background, the design and development of NRU presented major problems. The higher flux, 3×10^{14} neutrons/cm^2/s, at 200 MW compared with 6×10^{13} in NRX at 40 MW, meant a greater transfer of heat from the fuel and consequently a different design of fuel element. The flat bar shape selected for the uranium-metal fuel elements made the assurance of contact between sheath and uranium more difficult than with the round NRX fuel. An innovative design concept involving the changing of fuel at power was complicated by the need to avoid any loss of the heavy-water coolant.

Access to high-flux regions was provided in several ways: vertical tubes, horizontal through-tubes and re-entrant tubes called thimbles. The space under the reactor was arranged to facilitate a possible calandria replacement. Progress in design and construction was slower than originally scheduled, but by 3 November 1957 the reactor reached criticality. There were some teething problems with the new fuel. Water entered through cracks in the aluminum sheath, causing it to swell away

from the uranium. Perhaps there were some rods in this condition when a power surge on 24 May 1958 resulted in disruption of several rods. During a difficult rod removal, a piece of uranium fell out of the removal flask into a pit at the top of the reactor and caught fire. The building was severely contaminated and three months passed before the reactor was started again at lower power. By the end of November, NRU was again up to power. This was the most serious incident in NRU and subsequently, with fuel in which the uranium was bonded to the aluminum by nickel, NRU operated in an exemplary manner. It has been, like NRX in its heyday, a pre-eminent research reactor and one of the world's outstanding reactors for engineering tests and radioisotope production.

Nuclear Electric Power, NPD

The ten or twelve years following 1952 were crucial years for the future of nuclear power. NRX and reactors elsewhere had demonstrated the feasibility of producing large amounts of controlled energy from uranium fission, but the energy was available only as heat at relatively low temperature and its production had been without serious cost constraints. Each branch of science and technology faced new restrictions in its application to reactors. For example, material used to construct the cores must have low absorption for neutrons, and coolant chemistry had to take into account the ionizing properties of radiation and the possible transport of radioactive material by the coolant. Such restrictions complicate the cost estimates required in nuclear-plant planning.

The United States arrived at civilian nuclear electric power in 1957 by way of power plants for submarines – small cores containing enriched uranium with ordinary water as moderator and enclosed in pressure vessels. For its first power plants (1956), the United Kingdom modified its designs of gas-cooled, graphite-moderated, natural-uranium-fueled, plutonium-producing reactors to provide dual-purpose plants that would produce plutonium for weapons and electric power for civilian use. In neither case was there strong economic motivation to avoid wasting neutrons.

The situation in Canada was different. A nuclear power program would be a significant item in the national economy, with no military nuclear program to dwarf the expenditures on civilian energy. Ontario Hydro was the only Canadian utility large enough to accommodate a nuclear power plant that was also in need of a new source of energy; but the cost of the power it supplied to its customers was low, setting a difficult target for the nuclear option.

Lewis, whose proposal of 1951 set the Canadian power program in motion, continued to expand and refine his considerations of cost, putting emphasis on the value of neutrons and insisting that they must not be wasted. Some neutrons produced in a reactor disappear without causing fission by leakage out of the core, by unproductive capture, or by capture in uranium-238 to yield plutonium-239, a fissile nuclide.[13] For the power to remain constant, an average of one neutron per fission must result in a further fission. Wastage of neutrons by excessive leakage or unproductive capture reduces the useful life of the fuel.

Because plutonium is fissile, its production prolongs the useful life of the fuel. An important aspect of costing is choosing the balance between the gain due to an increase in plutonium and the added costs to achieve it. Estimating the cost of nuclear power became a growth industry. The Canadian plan to use heavy water as moderator and natural uranium as fuel had some good data from NRX experiments on which to base predictions of fuel consumption. Americans regarded Canadian cost predictions as unduly optimistic, but by the mid-seventies the success of the Pickering Nuclear Generating Station validated the Canadian estimates. The enriched-uranium, light-water system has a lower capital cost but a higher operating cost than the CANDU system; the former is competitive where there are high capital charge rates, as for private utilities in the United States.

[13] The survival time of a neutron in the core is so short compared to its lifetime for natural decay to a proton and an electron, about 1 to 10^6, that the effect of natural neutron decay on reactivity may be neglected.

In 1954, a Nuclear Power Group consisting of engineers from Canadian industries and utilities was assembled at Chalk River to design a prototype nuclear power plant. They soon had a design concept for a 10 MWe plant, later 20 MWe, in which the reactor was moderated and cooled by heavy water at high temperature and high pressure, and was fueled with natural uranium metal contained in zirconium-alloy tubing. The reactor core was enclosed in a pressure vessel.

In spite of its moderately large neutron absorption, stainless steel was being considered by the United States for fuel sheathing in its power-reactor cores, but this was feasible only with enriched uranium. Very early, zirconium had been considered as undesirable because of an apparently significantly large absorption cross-section. However, in 1947, researchers in the United States discovered that much of this cross-section was due to an impurity, hafnium, making purified zirconium an attractive possibility as a fuel-sheathing alloy, especially for natural uranium.

A partnership was formed between AECL, Ontario Hydro and Canadian General Electric, each being given distinct responsibilities. Some engineers transferred from the nuclear power group to work on the prototype nuclear power plant, called the Nuclear Power Demonstration (NPD). A site was chosen for NPD near the hydroelectric generating station 25 km up the Ottawa River from the Chalk River laboratory. Excavation and construction were soon under way.

The high temperature of the cooling water made the proposed use of uranium metal fuel questionable. Uranium dioxide (UO_2) was shown, largely in NRX loops, to have physical and chemical properties more suitable for in-reactor use than uranium metal, but its lower density made it less desirable from a nuclear standpoint. There were strong opinions on both sides, but eventually a zirconium alloy developed in the United States and UO_2 were chosen.

The Nuclear Power Group began work on the design of a 200 MWe nuclear power plant based on the concept of zirconium-alloy pressure tubes instead of a pressure vessel, uranium dioxide instead of uranium metal, and an alloy of zirconium for sheathing. NPD, designed as a reactor contained in a pressure vessel, was now headed into a dead end. In March 1957, construction was halted for preparation of a new design based on pressure tubes and uranium dioxide fuel.

Problems had been envisaged with the pressure-vessel NPD. As a prototype, its success would not have assured scaling up to high-power versions, since the pressure vessels required would be very large, much larger than pressure vessels for corresponding light-water reactors. (Because its slowing-down distance is longer, heavy water is a less effective medium for slowing down neutrons than is light water, and so it requires a larger volume of moderator between fuel rods.) In addition, the safety of the pressure vessel was being questioned.

The change to pressure tubes and uranium dioxide was welcomed and put Canada firmly on the road to CANDU, a Canadian-developed class of reactors moderated by heavy water, with fuel in zirconium-alloy pressure tubes and fuel-changing at power. The current representatives have horizontal pressure tubes and are cooled by heavy water; the fuel is uranium dioxide in the form of short cylindrical pellets sheathed in zirconium-alloy tubes, which are assembled into bundles 50 cm long, with spaces between the tubes for flow of coolant (see the illustration in chapter thirteen). The fuel bundles are fed through the reactor in opposite directions in alternate pressure tubes, resulting in a symmetrical distribution of power longitudinally. Normally, the bundles passing through any one pressure tube emerge with a similar degree of irradiation, but the intensity of irradiation varies across the reactor from pressure tube to pressure tube. By appropriately varying the length of time the fuel spends in a tube, the distribution of power may be flattened across the core. Fueling is normally done with the reactor operating at power, a major advance that avoids long non-producing intervals while fuel is being changed. Fueling at power, longitudinal symmetry, and flattening of the power distribution all enhance the efficient use of fuel. The fuel has been natural uranium, but the design is also suitable for enriched uranium, fuels containing plutonium or a fuel cycle using thorium.

Within about six months the new design of NPD, modeled on the emerging CANDU concept, was sufficiently developed for construction to be resumed. The complex discussions and

negotiations among the three corporate partners and the government have been described by Robert Bothwell in Nucleus. The cognate scientific and technological developments are described later in this book. Of special importance were the behaviour of uranium-oxide fuel and the response of zirconium alloys to their environment, especially the deterioration that could result from the absorption of hydrogen and its subsequent precipitation as zirconium hydride.

After the necessary commissioning, NPD was brought to criticality on 11 April 1962, and power was first fed into the electricity grid on 4 June 1962. With its refueling at power and low loss of heavy water, NPD successfully demonstrated the feasibility of the CANDU concept. Chapter ten gives a more detailed overview of the CANDU nuclear power system.

Douglas Point

Even before NPD went critical, construction of the 200 MWe plant had begun at Douglas Point on Lake Huron. At that time, 200 MWe was regarded as full-scale, but before the plant was completed it had been outclassed by the start on the Pickering station, where there would soon be four 500 MWe plants. Douglas Point is now usually referred to as a prototype.

In 1958, the Nuclear Power Group was disbanded and a new AECL division, the Nuclear Power Plant Division, was set up in Toronto, to be responsible for the design and development of the Douglas Point plant and to oversee the work on NPD.

Whiteshell Organic Cooled Reactor (WR-1)

Water, either ordinary or heavy, has disadvantages as a coolant in a power reactor. At the temperatures needed for efficient generation of power, the water exerts a high pressure, which increases rapidly with temperature. In pressure-tube reactors, a balance has to be struck between the coolant temperature and the thickness of the pressure tube. Increasing the temperature increases the pressure and thus a thicker pressure tube is needed. The higher temperature permits a higher thermodynamic efficiency, but the thicker tube captures more neutrons. The balance results in a lower efficiency than that obtained in coal- or oil-burning plants.

In the fifties, terphenyls, which are organic liquids used for heat transport, were studied as possible alternatives to water as reactor coolants. Canadian studies indicated that a heavy-water-moderated reactor cooled with an organic liquid might produce power more cheaply than a water-cooled CANDU. The organic coolant could be run at a higher temperature with thinner pressure tubes than the water.

A proposal to build a test reactor cooled by organic liquid as the centrepiece of a nuclear research establishment to be established in Manitoba was approved in 1960. The establishment was named the Whiteshell Nuclear Research Establishment. Construction of the reactor was begun in 1963 and criticality was achieved in 1965. The reactor was called WR-1.

Safety

Hazards associated with fission reactors – e.g., the production of dangerous radioactive material and the potential for rapid overheating (but not for a bomblike explosion) – have produced a special concern for safety that has been evident ever since the early days of NRX design. Safety aspects were routinely considered and, on occasion, were referred to the Pile (reactor) Operating Committee. In 1958 a ZED-2 Safety Review Committee was formed, very soon to be replaced (1959) by the Review Committee on Reactor Safety, with a mandate initially to review the safety of ZED-2 and, eventually, all AECL reactors. In 1967 it was replaced by the Nuclear Safety Advisory Committee (NSAC).

The presence in power reactors of large volumes of water at high pressure and high temperature (about 300°C) introduced major increases in hazards over those of research reactors such as ZEEP or NRX. In the event of a destructive power runaway, steam would greatly enhance the escape of fission products from the disrupted reactor and their dispersion in the environment. Even without a runaway, the pressure of the hot water may cause catastrophic rupture of the pressure-retaining components. Much applied research and development has been

done by AECL to enhance the safety of power reactors, to analyze the course of potential accidents, and to ensure containment of radioactive material in the event of a failure. This work is described in chapter fifteen.

Because the moderator in a pressure-tube reactor is cool, there is much less water at high temperature than there would be in a pressure-vessel reactor. This in itself enhances safety, but in addition the cool moderator can act as an energy sink rather than promote dispersion. The change from metal to oxide fuel was another safety improvement, because if the hot metal came into contact with water it would have reacted chemically to release hydrogen, which forms an explosive mixture with air.

Before NPD, the Control Board had let AECL handle reactor safety internally. Unlike the existing reactors, which were owned and operated by a federal agency (AECL), the NPD reactor would be operated by Ontario Hydro and owned federally. For this and perhaps other reasons, the Control Board decided to require the licensing of reactors not fully owned and operated federally. A safety assessment was required for the licence, and in 1956 the Control Board established the Reactor Safety Advisory Committee (RSAC) to do the assessment. Under the guidance of its chairman, G.C. Laurence, who had earlier been involved with the subject, the RSAC developed a distinct Canadian approach to reactor safety. General principles are described in chapter four.

International Aspects

The Canadian project was formed with tripartite international participation. The tight security of the early days hindered but did not prevent broader contacts; once the nuclear character of the project had been revealed, in 1945, most of the basic research could be freely published and discussed; exceptions were, for example, properties of plutonium and certain parameters of the fission process.

Canada earned a high status in nuclear matters at the United Nations. It had membership on the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and on the United Nations Scientific Advisory Committee (UNSAC). The first of four United Nations Conferences on Peaceful Uses of Atomic Energy was convened in Geneva in 1955, the organization of the conference being a responsibility of UNSAC. Canada helped organize the conference and presented thirteen technical papers. Before the conference, a vast amount of information was declassified by agreement between the United States, the United Kingdom, and Canada. This declassification broadened the scope for publication and left only a small fraction of the work at Chalk River secret. However, commercial secrecy appeared as the development of nuclear power plants produced proprietary designs and processes.

In addition to convening the 1955 conference, the United Nations promoted the establishment of the International Atomic Energy Agency (IAEA). The purposes of the IAEA were to provide for the control of fissile material and to assist its member states, particularly the developing countries, in peaceful uses of atomic energy. The problem of control was a difficult one, which has resulted in a system of safeguards involving the inspection of nuclear facilities to detect and hence deter military applications.

Canada was made a permanent member of the IAEA's board of governors and many Canadians have served the IAEA in a variety of positions. Particularly noteworthy was Lewis's twenty-year-long chairmanship of its Scientific Advisory Committee.

Values of nuclear properties such as cross-sections for neutrons are important for designing reactors and in research. The three countries – Canada, the United Kingdom, and the United States – shared information on nuclear data, with due regard to individual national restrictions. To facilitate the transfer, a Tripartite Nuclear Cross-Section Committee was established in 1956. The committee was very active, not only in the transfer of data, but also in such matters as standardization of notation, exchange of samples for measurements, and arranging for joint measurements; it was dissolved in 1963. Canada was a member of the European-American Nuclear Data Committee formed a few years earlier, which soon became the Nuclear Energy Agency Nuclear Data Committee. In 1966, an International Nuclear Data Committee was set up by the IAEA, Canadians having played a part in its formation.

Early in 1955 Canada and India began discussions which culminated in an agreement that Canada would supply an NRX-type reactor to India under the Colombo Plan; the Canadian-supplied reactor is now called CIRUS. This was AECL's first large international project and indeed also established a new magnitude for international nuclear aid. Cooperation on the technical aspects was good, but India was loath to accept Canadian inspection to ensure that plutonium was not diverted to non-peaceful uses. There were no international standards to guide the discussion. However, Canada received the right to inspect Canadian-supplied fuel and India gave assurance of peaceful uses only.

In December 1963 an agreement was signed between Canada and India for the construction of a Douglas Point-type CANDU reactor at Rajasthan, India, to be called RAPP. Three years later an agreement for a second reactor was signed. The first reactor, RAPP 1, was in operation in 1973. Then in 1974, India detonated a nuclear explosive device using plutonium from CIRUS, and claimed that it was a peaceful explosion. This ended Canada-India nuclear relations. RAPP 2 was completed by India alone in 1981.

The potential for a large international market for nuclear power plants was recognized early. By 1958, international marketing of nuclear power plants had begun; Canada was expounding the prospect of cheap power from Douglas Point but was not yet in the marketplace. By 1962, Canada was also looking for sales. Except for its impact on the technical work of the laboratories, the marketing story is not germane to the purpose of this book and has already been told (Bothwell, see Bibliography). Suffice it to say that Canada has maintained a sizeable foothold in a field where the competition includes intense sales efforts by the U.S.A., France and Germany.

A 132 MWe CANDU nuclear plant was built in Pakistan at Karachi by Canadian General Electric. Named KANUPP, it began operation in 1972. This was the only non-AECL Canadian sale of a CANDU.

The international marketing of radioactive isotopes is an unqualified Canadian success story. By the mid-fifties, an important share of the worldwide demand was supplied by AECL's commercial products division. Its cobalt teletherapy machines for the treatment of cancer won wide acceptance, and continue to today. Demands for the radioactive cobalt isotope used in these machines taxed the capacity of NRX until NRU was in operation; many other isotopes were also produced. Medical applications continue to predominate, but there are also many industrial uses. Chapter five describes the activities of the commercial products division in more detail.

Power Reactor Development Program Evaluation Committee

A few weeks after AECL's tenth anniversary (1 April 1962), NPD began to feed electric power into Ontario Hydro's grid. AECL had come a long way. It was an opportune time for AECL to reassess the paths it was taking, and to seek ideas for major new research facilities.

To assess and guide the power reactor program, the president of AECL set up the Power Reactor Development Program Evaluation Committee (PRDPEC), with Lewis as chairman. It held its first meeting in March 1962. Its initial task was to compare four nuclear power plants based on heavy-water-moderated, natural-uranium-fueled reactors. Each plant comprised a steam turbine, an electricity generator, the necessary auxiliary equipment, and a reactor. The reactors differed in their primary coolants:

1 Pressurized heavy water (PHW). PHW had the best technological base because NPD was in operation and Douglas Point was under construction.

2 Organic liquid (terphenyl). Considered promising as a reactor coolant by several countries, organic liquid contains only about three-quarters as much light hydrogen as water does; the outlet coolant in the comparison reactor was 130°C above that of the D_2O in PHW, and at one-sixth of the pressure, both advantageous possibilities. Organic coolant was to be used in the organic test reactor (WR-1) being planned for Whiteshell; in fact, the initial name for WR-1 was OTR.

3 Light-water fog. Fog cooling, in which the coolant enters the fuel channels as low-quality steam (i.e., steam loaded with water droplets) and leaves as higher-quality steam (i.e., with fewer droplets), was of interest, because it would reduce the amount of light water in the reactor core compared to pressurized or boiling light water. Fog cooling was, however, at an early stage of development and problems were foreseen with dryout of the fuel-sheath surface, and with the means to introduce fog into the channels; a direct cycle was considered, with steam going directly to the turbine.

4 Boiling light water (BLW). BLW offered several advantages over PHW, one being direct-cycle operation, another being the avoidance of losses of heavy water from the coolant system, but BLW would be unacceptable unless the amount of light water in the core could be kept low.

During the first year, the committee concentrated on analyzing the four types to arrive at comparative costs of power. It arranged for a report to be prepared on each of the plant types. For this to be done required company-wide involvement, including sufficient design effort to permit credible costing of the power produced. Procedures were established to ensure that the comparisons were equitable.

The choice of the electrical output of the plants was the subject of much discussion. Douglas Point would be 200 MWe, an output that was considered, at the time of commitment, to be full-scale, but as a result of increasing turbine sizes and forecasts of demand growth, the plant was already being looked upon as a prototype. Many sizes were considered, ranging up to 750 MWe; the size chosen for the final comparison was 450 MWe.

The committee reported on the comparison in March 1963. The spread in estimated costs of power was about 0.04 cents/kWh (roughly 8 percent of the cost). Organic had the lowest cost, and PHW and fog were close together at the high end. In spite of this spread the committee recommended, "In view of the competitive economic position and advanced stage of design and development, the D_2O-cooled reactor deserves priority for extension to higher power ratings and general improvement ... The economic attraction of the organic-cooled design justifies its continued development ... Development of the fog- and boiling-water-cooled channels may bring rapid rewards and should be pressed vigorously."[14]

PRDPEC remained active for many years, watching over the power program and giving advice on the full-scale plants planned by Ontario Hydro.

Power Reactors

Nuclear Power Demonstration (NPD)

The success of NPD was an important advance in the Canadian approach to nuclear power. It proved the feasibility of the pressure-tube system for heavy-water cooling, of fuel changing at power, of zircaloy-sheathed uranium dioxide as fuel, and of the CANDU concept in general. Modifications of components and procedures were tested; for example, various designs of fuel, and boiling in some channels. The NPD station became the training centre for Ontario Hydro's nuclear operating staff; it was kept in useful operation for twenty-three years in spite of its mere 20 MWe contribution to the Ontario grid.

Douglas Point

The Douglas Point reactor began operation in 1966. Early problems with heavy-water leakage and other matters prevented steady operation for some time. During operation, radionuclides were transported out of the core by the coolant and deposited on portions of the coolant system outside the shielding, resulting in high radiation fields in working areas. These problems were eventually overcome and Douglas Point provided much electricity to the grid. In later years it served as a source of steam for a heavy-water extraction plant. The lessons of Douglas Point enabled designers to provide a more successful station at Pickering, Ontario.

Pickering

Even before Douglas Point was completed, the growing demand for electricity hastened the move by Ontario Hydro to acquire

[14] AECL, 1730 *Power Reactor Development Evaluation*, 21-22.

nuclear stations of "commercial" size. In line with PRDPEC's conclusion that the PHW reactor was the advisable choice for higher power, the design of a 500 MWe reactor was undertaken during 1963, and the project developed into a four-reactor station at Pickering, just east of Toronto. The same site had been considered for the reactor that became the Douglas Point reactor, but, because of its proximity to Toronto, it had been rejected in favour of a site on Lake Huron. The development of vacuum containment (i.e., a large vacuum building to act as a sink to suck in steam, radioactive gases and aerosols in the event of a serious reactor accident) provided an economic solution for a four-unit station that was judged to make the Pickering site acceptable. The first of four 500 MWe plants was operational in 1971. By 1973 these four plants constituted the Pickering A station. Four more plants were added to the site by 1985 to form the Pickering B station. AECL was the designer of the nuclear parts of Pickering A. In its early years the Pickering station was very successful; recently, however, pressure-tube replacement has been necessary somewhat sooner than expected.

Gentilly BLW (Boiling Light Water)

Soon after the PRDPEC recommendations, Hydro-Quebec, the Quebec provincial electricity utility, expressed interest in nuclear power. This, combined with the recommendation of PRDPEC for a strong follow-up on light-water cooling, led in 1965 to an agreement for the construction of a heavy-water-moderated, BLW-cooled power reactor at Gentilly, Quebec. The quality (i.e., the weight percent of steam) of the outlet coolant was 16.5 percent. The channels were vertical, to permit going later to higher-exit quality. The plant reached its design output of 250 MWe in May 1972. The combination of BLW and natural uranium imposed heavy demands on the control system, adding to other operational problems. Although controllable at 250 MWe, the system did not really lend itself to larger sizes. As the 250 MWe plant was not economically competitive with the cost of power in Quebec, the plant was taken out of service in June 1977. Chapter sixteen describes the CANDU-BLW in more detail.

RAPP (India) and KANUPP (Pakistan)

(See International Aspects)

Bruce

By the late sixties Ontario Hydro foresaw the need for more power, and began to plan for a four-plant station in the same area as Douglas Point. The new station was an Ontario Hydro project, with much of the nuclear engineering and development being done by AECL. The plants were each of 750 MWe capacity, with extra heat output to supply steam to a heavy-water plant. The station, called Bruce A, came on line from 1976 to 1979. Another station, Bruce B, with four plants, each of which was of 860 MWe capacity by 1991, was completed from 1985 to 1987. Initially the operation of both stations was excellent, but problems have developed in Bruce A.

CANDU 6

By the early seventies AECL, having decided that there was a potential market for plants of moderate size, designed a 600 MWe plant that incorporated many of the lessons learned over the years. The result was CANDU 6 (originally CANDU 600), and plants have been installed at:

- Gentilly-2, Quebec — 1982 criticality
- Wolsong-1, Korea — 1982 criticality
- Point Lepreau, New Brunswick — 1983 criticality
- Embalse, Argentina — 1983 criticality

All of these units have operated at high capacity, Point Lepreau being a world leader.

The following figures, from a general proposal for CANDU 6, indicate the huge size of a nuclear power plant; values for specific plants may differ. The calandria is a horizontal stepped cylindrical tank made of stainless steel, 6 metres long and, in the main, 7.6 metres in diameter. It accommodates 380 pressure tubes, each containing 12 fuel bundles. The total uranium content is 84 tonnes (Mg). The calandria contains 240 tonnes of 99.75 percent heavy water as moderator; the moderator system contains a further 23 tonnes. The primary heat transport system contains 200 tonnes. The total fission power is 2,180 MW, from which 685 MWe are generated, 638 being sent out on the grid.

The CANDU nuclear power system is very flexible regarding its fuel. The stations presently in use are in essence fueled with natural uranium. A variety of fueling schemes has been considered, including the use of plutonium, enriched uranium and thorium. Chapter eighteen describes the relevant fuel cycles.

Research Reactors

ZEEP

Although ZEEP was too small to be useful for all measurements on fuel assemblies for power reactors, because of their large sizes and spacings, it continued in use for many years as a facility for measuring specific parameters. For example, in the early sixties a mockup of a power-reactor fuel channel was installed in ZEEP and measurements were made with it to determine the neutron energy spectrum (the energy distribution of neutrons) in various regions of the fuel assembly. In 1980 ZEEP was permanently decommissioned.

NRX

The NRX reactor continued to be a major facility for research and isotope production. With the confidence gained from the 1952-54 restoration, a spare calandria was kept on hand and modified from time to time to provide better facilities. It was installed in 1970. Natural-uranium metal, the only possible choice for the original NRX fuel, was also suitable for chemical extraction of plutonium. When Canadian interest in plutonium waned, the spent NRX rods were sold to the United States as part of the agreement for sale of NRU rods. In anticipation of the expiry of the agreement, studies were made of several less costly fueling schemes for both NRX and NRU. In these reactors the property of interest is the flux, not the power; consequently, enriched fuel proved to be preferable. In NRX an increase of about 50 percent in flux, with no change in power, was obtained with a fuel loading of natural uranium dioxide and rods of an alloy of enriched uranium and aluminum. Both types of fuel were sheathed in aluminum. The change was completed over an interval of six months in 1962, the flux being kept constant by reducing the power until the new fuel was in place; the power was then raised to the old value to increase the flux. This procedure eased the transition for experiments such as loop irradiations. Later the fuel was converted entirely to enriched alloy.

PTR (Pool Test Reactor)

The primary purpose of PTR (described later in this chapter) was to enable measurements of reactivity to be made with irradiated fuel, an intensely radioactive material. The information provided data for accurate predictions of the lifetime of fuel in power reactors - in other words, the amount of energy that could be extracted from the fuel before the destruction of fissile material and the accumulation of fission products lowered the fuel's contribution to an unacceptable level. Measurements continued over the years, some as parts of international programs.

NRU

Like NRX, the NRU reactor was initially fueled with natural-uranium metal. In 1963, with the expiry of the plutonium sales contract with the United States, the fueling was changed to rods of an alloy of highly-enriched uranium and aluminum. There was a reduction in power to maintain the same flux, which was of a magnitude appropriate for tests of power-reactor fuel. The needs of these tests outweighed the attractiveness of higher flux for other research, but in small regions of the reactor, provision could be made for special flux requirements. There was a capability for increased radioisotope production.

Corrosion and other deterioration of various components of the NRU calandria and its attachments resulted in a decision to change the calandria, which was done in 1973-74. The opportunity was taken to improve the in-core facilities for loops.

NRU is a major source of radioisotopes, including cobalt-60 of high specific activity and molybdenum-99, which decays to technetium-99m, a radioisotope in great demand for medical diagnoses.

ZED-2

ZED-2, a larger and more elaborate version of ZEEP, was built for measurements on power-reactor lattices. It has been an essential

tool in the expansion and refinement of the power program, permitting diverse measurements on a variety of lattices.

Organic Test Reactor (OTR) WR-1

The PRDPEC recommendations on organic cooling meant that the reactor approved for Whiteshell was in the main stream of AECL development. This ensured its construction, which began in 1963; reactor startup was in 1965. Developmental tests of organic liquid discovered small changes that greatly improved its performance, such as the removal of chlorine, and the addition of a trace of water.

By 1970, experience with WR-1 led to chemical conditions of the coolant that gave excellent results; in particular, there was hardly any transport of radioactive materials by the coolant. However, by that time large commitments had been made to heavy-water coolant for the Ontario Hydro system and money for the development of an organic-cooled power plant was not forthcoming. With hindsight, it might have been better to have followed the organic route for the Gentilly-1 reactor, rather than the light-water route. The experience with organic coolant in WR-1 and the concurrent development of a CANDU-organic cooled reactor design concept are described in chapter sixteen.

Taiwan Research Reactor (TRR)

AECL sold an updated version of NRX to Taiwan late in the sixties. A diplomatic break with Taiwan occurred before the construction was complete, but it did not interfere with the project.

Small Reactors

During the very early days of nuclear energy there was uninformed talk about such things as nuclear-powered cars. What did appear was a number of very large fixed installations for plutonium production and, later, for electric power generation. There were also reactors such as ZEEP and NRX, and as time went on, research reactors with enriched fuel operating in "swimming pools". Small power reactors were built in the United States and the USSR, especially for military uses such as submarine propulsion.

In the mid-sixties, AECL began to consider reactors to produce up to 30 MW of heat for housing in remote areas, or for industrial use. The designers considered them safe enough for unattended operation. Although no commercial reactors were produced, the idea of safe, unattended operation was followed up, as explained in the next paragraph. A 2 MW demonstration reactor was built at Whiteshell in the mid-eighties, but in the absence of a promising market, the project was not developed further.

SLOWPOKE

A low-power research reactor capable of safe, unattended operation was developed and given the name SLOWPOKE (Safe Low Power Critical Experiment). A prototype SLOWPOKE, after tests at Chalk River, was installed at the University of Toronto, where for many years it or its replacement, a production model, provided an adequate neutron flux for trace-element analysis, tracer experiments and radioisotope research. SLOWPOKEs have also been installed in several other Canadian institutions, and one has been installed in Jamaica.

MAPLE

After extensive consideration of Chalk River needs, and an assessment of research reactors elsewhere, particularly in the United States, AECL designed a research reactor called MAPLE (Multipurpose Applied Physics Lattice Experiment), which would use enriched fuel. Chapter twenty describes AECL's studies on small reactors, and the realization of the SLOWPOKE and MAPLE reactor concepts.

Heavy Water

Until about 1970, the Canadian project was entirely dependent on the United States for heavy water. Because of time constraints, the Americans had not chosen heavy water as a moderator for their wartime production reactors, but they maintained an interest in it for many years, and had chosen it for production reactors committed in 1950. The heavy-water extraction plants built to supply those reactors provided the heavy water for

NRU, NPD, Douglas Point and the first unit at Pickering. By 1964 a need was felt for a Canadian supply. The political, financial and technical maneuverings were complicated (Bothwell pp. 320-331). In outline, the construction of a plant at Glace Bay, Nova Scotia, to produce 200 tonnes of heavy water per year using sea water for cooling, was a joint undertaking of the provincial government and a private company. This was a technical and financial disaster; by 1976, the plant was brought into production under AECL management following a major reconstruction and conversion to fresh water. A second plant, established by Canadian General Electric in Port Hawkesbury, Nova Scotia, was more successful and first produced heavy water by 1970. The failure of the Glace Bay plant put the startup of subsequent units at Pickering in jeopardy. AECL had to scrounge heavy water from all over the world as well as borrow the heavy water from ZEEP, NRU, Douglas Point and Gentilly-1.

Because Ontario Hydro felt the need for a heavy-water plant under closer control, AECL built a plant on the Bruce site, which began operation in 1972. By previous agreement the plant was sold to Ontario Hydro in 1973.

As AECL became involved in heavy-water production, laboratory staff undertook research that solved many technical problems arising in the plants. They developed an alternative process of separation to the pilot-plant stage for possible future use. Laboratory staff also developed several instruments for measuring the deuterium content of water. Chapter seventeen gives further details on heavy-water production.

Research and Development, 1952-1985

In the early years of the project, when there was considerable ignorance of the properties of new materials and devices, progress was made possible by large design allowances. Good fortune, combined with those allowances, usually resulted in success. A noteworthy example is the design of NRX for 10 MW operation and its subsequent operation at 40 MW. With time, and particularly with the move towards commercial power plants, came pressure to reduce some of the large allowances by emphasis on research to improve design data. Also, it was essential to understand the exact behaviour of materials, both customary and new, in the novel environments of reactors. Applied research in chemistry and materials science became vital, as did basic research into the underlying phenomena; engineering development was a major user of the results.

In all fields, technology was moving from an empirical base to a scientific base as the knowledge and application of chemistry, physics and materials science increased. In the nuclear field much of the technology was new, in many cases because special requirements brought in previously little known materials and novel environments; decisions putting at risk millions of dollars sometimes had to be based on rather immature technical information. Intense, directed research was required. Underlying research was still necessary to provide a background of knowledge for the applied research. Clarification of terminology used to classify research is given at the beginning of chapter six.

This section provides some examples of research done by AECL from 1952 to 1985; a few other examples appear in the preceding text, notably on heavy-water extraction, organic coolant and safety. Subsequent chapters discuss the examples in greater detail.

Fission

The program of fission studies that was started soon after NRX began operating continued for many years with increasing sophistication of techniques and instrumentation (see chapter eight). This was mainly basic research, because the needs of the reactor program for such data had been largely satisfied. Time-of-flight measurements gave more accurate values of masses and energies. Fission caused by accelerated deuterons provided further information about the fission process.

Radiation chemistry

Ionizing radiation in a reactor or from radionuclides consists mainly of gamma rays, fast electrons or neutrons; a result of the ionization is the production of chemical changes in irradiated material. An important example that required study very early

in the project is decomposition of the heavy-water moderator into deuterium and oxygen gases. Studies of such chemical changes constitute radiation chemistry, a subject that has been a significant part of chemistry research at AECL (see chapter seven). When organic cooling was adopted, the effects of radiation on the coolant were carefully researched. The processes that result from the ionization in a material are complex and their interpretation has required development of considerable theory.

Radiotracers

Radioisotopes of an element provide a means of locating the element and following it through a series of movements or reactions. Many research problems have been solved by the use of such radiotracers. An example of practical importance was the unravelling of the mechanism of oxide growth on zirconium alloys. Important uses of SLOWPOKE reactors include radionuclide production and irradiation of specimens with neutrons. In the latter case, the resulting radioactivities serve to identify chemical elements in the specimens.

Channeling

During a determination of the ranges of accelerated ions in metals, an effect was observed that was later interpreted as relatively unimpeded travel of suitably directed ions along the spaces between rows of atoms in the crystals of the metal. This channeling, as it is called, is a phenomenon that has had many applications in basic research and in such applied research as development of semiconductor electronic devices.

Gamma rays of neutron capture

The neutron-capture gamma-ray measurements developed into a program of worldwide interest. The gamma-ray energies became a basis for international tabulations and provided important data for theories of nuclear structure, as they represent energy differences between energy levels of the nucleus that results from the capture. They are characteristic of that nucleus and can be used as an analytical tool for its identification (see chapter eight).

Physics with accelerators

AECL's major program of basic research has been the study of nuclear structure by the use of accelerated ions. After some interesting results were obtained using relatively low-voltage accelerators, an impetus was given to the program by the coming into operation, about 1952, of the three-million volt Van de Graaff accelerator, planned in the Montreal Laboratory. This was followed in 1958 by the first tandem accelerator, a great advance in accelerator design that gave not only higher energy but also a much wider choice of the atomic number of the accelerated ion. This tandem was replaced by an MP Tandem, which, after many years, had a superconducting cyclotron coupled to it. This series of machines has enabled Chalk River physicists to spearhead much important research on the structure and behaviour of nuclei (see chapter eight). Development of detectors and auxiliary equipment has been an important factor in the success of the program.

Life Sciences

Research into the life sciences has been directed towards furthering knowledge on the biological effects of radiation, the behaviour of radionuclides in the environment, the use of radionuclides for biological research, and developing instruments for radiation protection. Mathematical models have been developed to describe the metabolic behaviour of radionuclides and the resulting doses from them to organs and tissues in the human body. The effects of radiation have been studied not only on complete organisms but also at the cellular and molecular levels. In particular, important information has been obtained on the association between variations in radiosensitivity, DNA repair deficiency and cancer proneness in humans; an understanding of the various levels of radiosensitivity in people with cancers is necessary to establish a sound protocol for their treatment by radiation. Chapter three discusses this subject in greater detail.

Neutron diffraction and scattering

The original diffraction spectrometer and later models provided monoenergetic beams of neutrons, used mainly for measurements

of the intensity of neutron scattering by substances as a function of angle. The successful design and operation of a spectrometer that incorporated a second crystal that could be rotated about an axis to measure the energies of the scattered neutrons greatly broadened the scope of the research. It enabled the characteristics of vibrations of crystals to be determined and it helped to resolve a long-standing problem of anomalous behaviour of liquid helium at low temperature. This triple axis spectrometer and achievements with it brought several honours, including a Nobel Prize, to the senior researcher (see chapter eight).

Reactor Physics

Studies of uranium/heavy-water lattices begun in the Montreal Laboratory were continued in ZEEP for some general cases, but specifically for NRU and NPD. To obtain the required precision with lattices containing multi-element fuel assemblies for power reactors, a larger version of ZEEP was installed at Chalk River and operating by 1960. It was called ZED-2, because Z-2 would have been Americanized in pronunciation to ZEE-2. ZEEP and ZED-2 were limited to studies of unirradiated fuel.

A major research program on reactor physics to find the effect of irradiation on reactivity was essential to enable estimates of fueling costs to be made. Early measurements with NRX have been mentioned, as has the extensive program of detailed measurements on samples from a long-irradiated NRX rod. To make more precise and varied reactivity measurements, a small, low-power reactor designed for such measurements was installed at Chalk River and was operating by late 1957. Called the Pool Test Reactor, PTR, it is fueled with uranium enriched in uranium-235 and is moderated by ordinary water contained in a deep "swimming pool". Irradiated samples of fuel are shuttled into and out of the reactor core on a fixed-time cycle. The resulting oscillations of reactor power, when compared with corresponding signals from unirradiated material, yield information about the effects of irradiation on reactivity. Chapter eleven describes the evolution of the physics of the CANDU reactor core.

The program on irradiated material involved much chemistry and mass spectrometry for determining the chemical and isotopic changes in the fuel. Theoretical formulas for the lattice properties were matched to the results of this extensive experimental program as they became available, the matching being done by adjustment of parameters in the formulas. With the advent of computers, the formulas formed the basis for elaborate computer programs that were used in the CANDU engineering design and cost estimates. The quantity of primary interest for estimating the fueling cost is the energy derivable from the fuel before the fuel must be removed because of loss of reactivity.[15]

Spallation; Electro-nuclear breeding

When fuel is irradiated in a reactor the accumulating fission products absorb neutrons, which are thus lost from the chain reaction; moreover, the relative production of neutrons decreases, because the loss of uranium-235 is not fully compensated by the production of plutonium-239. These effects necessitate the replacement of irradiated fuel by fresh fuel. Possible means of delaying replacement are to supply the reactor with plutonium produced elsewhere from uranium-238 by a source of neutrons, i.e., electro-nuclear breeding, or to feed neutrons into the reactor. It was for purposes of this kind that spallation was considered as a means of producing neutrons. In the fifties, little was known about the yield of neutrons in spallation by high-energy protons. Canada had no accelerator capable of accelerating protons to the desired energies (many hundreds of millions of volts), but there are protons of the requisite energies in cosmic rays. An experiment to measure neutron yields from spallation of heavy elements by cosmic-ray protons involved measurements at high-altitude sites; although difficult, it was successfully completed. More accurate measurements were later made at the Brookhaven Cosmotron in cooperation with a group from the Oak Ridge Laboratory (see chapter nine).

ING (Intense Neutron Generator)

The NRX and NRU reactors have been excellent research and engineering test reactors. In the early days their large neutron

[15] On a blackboard in his office Lewis kept a chart of this quantity, updating it as each new result from the program introduced a change. The chart was known as "Dr. Lewis's fever chart".

fluxes had given Chalk River physics research an edge over the rest of the world. By 1960 these fluxes were no longer exceptional, and in other countries facilities with larger fluxes were in operation. From an engineering viewpoint, the NRU fluxes were nearly ideal for testing power-reactor fuels; much higher fluxes would have been an inconvenience. But for some research higher fluxes would be advantageous or even essential.

For Chalk River to regain a position as one of the institutions with the highest neutron fluxes, a major new facility was needed. Careful study led to a proposal to build a high-current, high-energy proton accelerator to produce neutrons by spallation. This proposal for an intense neutron generator (ING) was an outgrowth of the idea behind the cosmic-ray experiment of the fifties. Such a facility, if located at Chalk River, would restore the laboratory's favourable position as regards neutron flux. It would also be a major step towards the development of electro-nuclear breeding. Initially, two accelerator types were considered: a linear accelerator and a separated orbit cyclotron. The former was chosen for development.

Characteristics proposed for ING were in several respects beyond the technical capabilities of the day. Development programs were started on ion sources to yield a proton current of a few tenths of an ampere, on efficient high-power radio-frequency oscillators, on accelerator structures, and on liquid-metal- cooled targets able to cope with the approximately 100 MW of power in the proton beam. As a result of opposition in the scientific and engineering communities and a failure to win government support, the ING project was terminated in 1968. Sufficient progress had been made to justify continued accelerator development, especially of medical and industrial irradiation devices. The ING project and the subsequent accelerators are described in chapter nine, as is AECL's involvement in a Canadian fusion program.

Chemical and metallurgical technology

In the early days, the extraction plants for plutonium and uranium-233 presented many problems in chemical engineering. Later, the analysis of irradiated CANDU fuel posed related problems.

Getting the chemistry of the reactor-cooling water to be compatible with the various metals in the primary heat-transport system was of great importance, as was a similar requirement for the water in the secondary system. Many considerations were involved in the choice of materials for these systems; steam generators are subject to costly failures if there is incompatibility. Canadian experience with steam generators has been among the best, although recently there have been failures. Chapter fourteen describes research that resolved many of the chemical and materials problems of the heat-transport system.

Thermalhydraulics

The performance of a power reactor depends critically on the transfer of heat from the fuel sheaths to the coolant. High rates of heat transfer mean a lower cost of power, but too high a rate can result in fuel damage. The difficult choice of the design rate for the multi-rod CANDU fuel bundle is based on empirical relationships derived from elaborate and costly experiments. These include tests on instrumented models of fuel bundles with electrically heated sheaths and heat removal by circulated hot water. The quality of the empirical data available has been greatly improved by continuous research in this field of thermalhydraulics during some forty years.

Another aspect of thermalhydraulics deals with the behaviour of the coolant under accident conditions. Chapter fifteen describes research on thermalhydraulics.

Loops

The in-reactor loops at Chalk River and Whiteshell, together with associated equipment such as shielded cells equipped for remote handling and examination, constitute equipment of great importance to research and development of fuel for nuclear power. Beginning in 1951, with the installation in NRX of an American loop, the number and sophistication of loops increased progressively. At times there were seven loops in NRX, four in NRU and four in WR-1. The situation was not static; in some cases, a loop would be installed, used for a series of tests, and dismantled to make room for a different loop. The staff acquired considerable expertise in choosing,

planning and executing experiments. Loops and spaces for loops were rented to organizations from other countries, usually with the stipulation that the results be available to AECL. Chapter two describes loops in greater detail.

Fuel

The choice of uranium compounds suitable for CANDU fuel was limited and soon settled mainly on uranium dioxide, but uranium carbide was used successfully in the organic-cooled WR-1. Much research had to be done to achieve consistently good behaviour over long exposure to neutrons.

Fission products include the noble gases xenon and krypton; their release from the uranium dioxide, although only partial, requires the provision of free volume. Free volume is also needed to accommodate thermal expansion. Temperatures inside the fuel pellets are high enough to promote movement of material, particularly gases. Impurities, especially water, can have a profound effect on the interaction of the fuel with its zirconium-alloy sheath. The study of these and other effects was the purpose of many loop irradiations. Chapter thirteen describes the complex behaviour of uranium dioxide in a power reactor and the problems associated with sheathing it.

Pressure tubes and sheaths

For the CANDU reactors with their water-cooled, natural-uranium dioxide fuel, the only metals suitable for pressure tubes and fuel sheaths are zirconium alloys. Two classes of alloy have served the Canadian program: zircaloys, developed in the United States, and zirconium-niobium, pioneered by the USSR. As with the fuel material, much research was needed to find the optimum materials and conditions. Hydrogen dissolves in zirconium and moves down a temperature gradient, concentrating in the coolest region. With increasing concentration, precipitation of zirconium hydride causes localized swelling and can have deleterious effects on the mechanical behaviour. The sheaths and pressure tubes may pick up deuterium, an isotope of hydrogen, from the coolant; the sheaths may also get hydrogen from any water in the fuel. Much information is required on these phenomena, and on the effects of metallurgical condition on hydrogen pickup. Another matter for important studies is the effect of irradiation on pressure-tube deformation. Many of these studies were based on loop experiments. There were, of course, other studies; for example, pressure tubes were burst deliberately to check the likelihood that bursting of one pressure tube would propagate to others. Longevity of fuel channels is of supreme importance in the economics of CANDUs. Chapter twelve discusses the underlying technology of fuel channels.

Advanced Fuel Cycles

Although CANDU reactors have traditionally been fueled with natural uranium, the fueling system is very flexible and lends itself to other fuels: for example, the plutonium that is a by-product of power production could be chemically separated and fed back into the reactors as oxide mixed with the uranium oxide; alternatively, slightly-enriched uranium could be used; irradiation of thorium in reactors with enriched uranium or plutonium-containing fuel would provide uranium-233 for development of a thorium-based fuel cycle. Schemes like these have been considered during much of the Canadian power program.

The primary incentive was to compare the resulting costs of power with costs from the traditional CANDU fuel. More recently, there has been a marketing incentive because certain characteristics of these advanced fuel cycles, particularly the possibility of reduced uranium consumption, appeal to some potential foreign customers. As chapter eighteen makes clear, AECL has maintained an extensive program on advanced fuel cycles.

Waste Management

The Perch Lake basin at CRL provides a suitable area for the management of local wastes; it has also accommodated waste from various nuclear activities outside AECL. It has been the site of many researches into the behaviour and management of wastes. When the Whiteshell laboratory was established, it had similar needs for waste management, but further research was required because of different soil characteristics.

A waste-management problem of much greater magnitude has arisen because the nuclear power program is producing quantities of highly radioactive used fuel and waste of varying levels of radioactivity. The treatment must take into consideration radioactive lifetimes of many thousands of years. There is public concern that significant amounts of radioactive material might enter the environment between now and the next hundred-thousand years or so.

In the mid-seventies, AECL began a program to prove that the wastes could be managed without harm to the public over such long times. The program has continued and has grown to include much research into waste repositories and the movement of water in soil and massive rock structures. An underground research laboratory has been established near Whiteshell. Chapters three and nineteen describe the waste management research and development. Chapter nineteen includes some of the political aspects of the long-term storage of nuclear-power-plant wastes.

Conclusion

By 1985, the closing date for this book, AECL had accomplished much of its primary mission. Twenty CANDU reactors were in service in Canada and elsewhere – Argentina, India, Korea and Pakistan. Several of these had appeared repeatedly among the first ten of the world's power reactors listed in the order of capacity factor, a measure of production reliability. Several more CANDUs were under construction.

In spite of this success, the mission to develop nuclear power plants could not be considered complete. Continued development was essential to keep abreast of the competition and to maintain a capability to deal with problems. To relax would allow the technology to atrophy; applied research and development were maintained at a high level.

The radioisotope business was flourishing. A variety of radioisotopes was being marketed, produced by irradiation in reactors (NRX, NRU and several power reactors) and in cyclotrons in British Columbia.

Pure research was advancing. The advent of the superconducting cyclotron was anticipated with considerable interest, and work on advanced slow-neutron spectrometers was in hand.

From the early days, industrial application of the work of AECL was considered important. This applied not only to the two main items, nuclear power plants and radioisotopes, but also to many aspects of the technology. A specific program for finding commercially viable applications was established. Chapter twenty discusses the successes and failures.

The transformation of AECL from a research organization of moderate size to a large organization capable of designing and supplying nuclear power reactors, as well as having a varied capability in radioisotope production, applied research and development, and pure research, has provided many lessons on how, or sometimes on how not, to proceed. Chapter twenty-one discusses some of these lessons.

Epilogue

A decade has passed since the nominal closing date of the book. It seems appropriate to summarize important events that have since occurred in Canadian nuclear affairs.

All four units of the Darlington Nuclear Station that were under construction on the shore of Lake Ontario, about 65 km east of Toronto, are now in operation, with a net capacity of 3,544 MWe; the station is the largest Ontario Hydro station and adds about 30 percent to the nuclear electric capacity of Canada. Some early problems that caused vibration and damage to fuel have been overcome.

An economic recession and political concerns about nuclear energy have delayed the implementation of Ontario Hydro's plan to proceed with the construction of further nuclear stations. An order prohibiting Ontario Hydro from doing work towards future reactors caused important funding to be withdrawn from AECL's forward-looking program. On the other hand, AECL's expertise and special equipment have been in demand for investigations into difficulties that arise in operating reactors; this demand is one justification for continued federal

support of AECL's technical capability, support that was strongly advocated by the Ontario Nuclear Safety Review, an Ontario commission on reactor safety that reported in 1988.

In spite of its commercial exploitation of its technology, AECL, as a crown company responsible for nuclear research and development, needs considerable core funding from the federal government. Current government retrenchment severely restricts AECL's potential.

Most of AECL's applied research and development is now done for outside organizations, a major change; in particular, the CANDU Owners Group (COG), an organization well described by its name, joins with AECL in choosing and directing, on a cost-shared basis, applied research and development of mutual interest.

The success of the CANDU 6 reactors, especially in Korea, brought an order from that country for three more. Construction of a five-unit CANDU 6 station in Romania resumed after delays caused by political events there.

To be able to respond to an expected demand for units of a modest size, AECL has designed a 450 MWe unit called CANDU 3. The design includes a high level of standardization but is nevertheless adaptable to individual requirements. This standardization is subsequently being applied to a larger 880 MWe unit called CANDU 9.

The radioisotope business flourished to such an extent that the Radiochemical Company was divided into two components, which have been separately incorporated.

NRX has been permanently shut down. Nordion has proposed the building of one or more reactors for radioisotope production when NRU is shut down. AECL has outlined proposals for an Irradiation Research Facility to replace NRU.

The Tandem Accelerator Superconducting Cyclotron combination provides a wide variety of heavy-ion beams. It attracts researchers in many disciplines from across Canada and around the world.

The neutron diffraction and scattering facilities at NRU are also of international status. A new system, comprising two spectrometers of advanced design, was installed in 1991. The facilities are used for basic and applied research on the solid state, and for commercial applications to non-destructive testing.

The goal of controlled fusion energy has been the aim of some very large programs by the major powers. In the late seventies a national fusion program was started in Canada; it has been managed by AECL for the federal government since 1987. The program had a budget of about twenty-five million dollars in 1991 and, with its focus on a few aspects that fit Canadian expertise, is consequently able to make useful contributions to the subject.

Looking to the future of fission energy, AECL sees the CANDU reactor as supplying a significant fraction of the world's needs for nuclear power plants. The only serious competition is from light-water reactors of various types. To support current plants and to prepare for future plants, AECL has a large research and development program, on, *inter alia*, advanced fuels, safety matters, waste management and heavy-water process development; the large investment in heavy water needed for a CANDU plant makes an important contribution to the cost of the power produced, a cost that is in the range of costs of power from light-water reactors. Lowering the cost of heavy water could be a determining factor in the competition.

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INFRASTRUCTURE AND LABORATORIES

Chapter Two

Organization, Management and Operations

E. CRITOPH

While AECL was not established until 1952, there has been continuity in the Canadian, federally sponsored, R&D effort in nuclear energy since 1942. This began with the formation in 1942 of the Montreal Laboratory and continued at Chalk River, first under the National Research Council (NRC), and then, since 1952, under AECL (see chapter one). Over this whole period, various names have been given to this effort, the most recent being "AECL Research".

This chapter describes the milieu, as seen from its laboratories, within which the R&D programs of AECL Research were carried out. The organization, management style and operations of AECL Research (in its widest sense) were vital and ever-present factors in the integrated AECL technical performance.

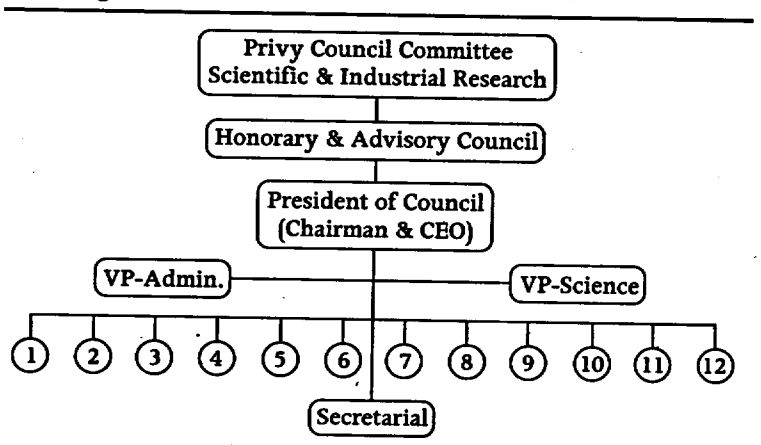
BACKGROUND

The initial organization, management style and operations of AECL Research arose from the historical circumstances under which the Canadian effort in nuclear energy was initiated, the nature of the site on which AECL was originally established, and the nature of the AECL mission, which was to ensure that atomic energy ("nuclear energy" in today's terms) was exploited to the maximum collective benefit of Canadians.

A 1946 NRC organization chart (see figure 2.1) identifies, in boxes, the elements that formed the NRC part of the Chalk River effort. Some support services were supplied by other parts of the NRC organization, and construction and operating support was the responsibility of Defence Industries Limited (DIL), which contracted with the Fraser Brace Company for construction.

The total NRC-directed effort at Chalk River in 1946 consisted of 289 employees, divided among the following branches:

FIGURE 2.1
NRC Organization – 1946



1. Extramural Activities
2. Medical Division
   — Chalk River Branch
3. Biology Labs
4. Prairie Region Labs
5. Chemistry Labs
   — Industrial & Chemical Engineering Branch
6. Mechanical Engineering Labs
7. Electrical Engineering & Radio Labs
8. Physics Labs
9. Atomic Energy Division
10. Patent Corporation
11. Codes & Specifications
12. New & Special Projects

- Engineering (atomic energy division)
- Technical physics (atomic energy division)
- Nuclear physics (atomic energy division)
- Theoretical physics (atomic energy division)
- Chemistry (atomic energy division)
- Research services (atomic energy division)
- Chalk River branch (medical division)
- Industrial & chemical engineering (chemistry labs)

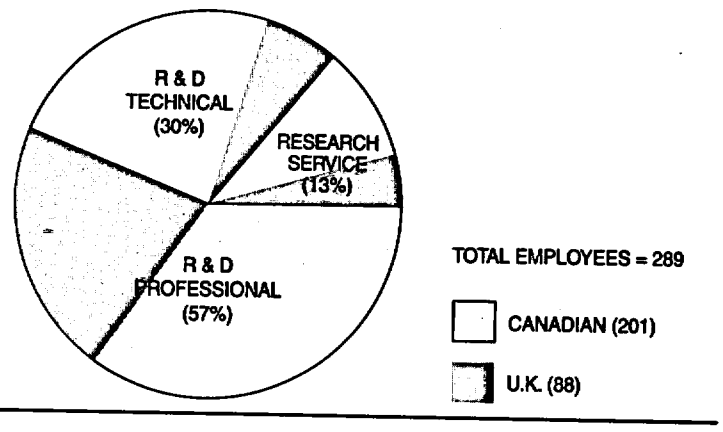
In addition, there were four people in the United States Liaison Office, about 650 workers associated with DIL, and fluctuating numbers of construction workers.

The composition of the NRC-directed staff employed was unusual. For one thing, there was a large contingent of United Kingdom staff, arising from the agreements that led to the formation of the Montreal Laboratory, and there was a high proportion of professional scientists and engineers in a wide range of disciplines. This, in itself, brought new challenges in management and operation. The NRC-directed staff (see figure 2.2) comprised, from Canada and the United Kingdom, R&D professional, R&D technical, and service staff.

Initially, the responsibility for operating the laboratory site fell to DIL. They made an important contribution to the overall program by establishing the good operational practices that provided the basis for the ongoing successful operation of the site. Operational responsibility was transferred to NRC in February 1947. A large part of the DIL effort and personnel was absorbed as part of NRC, at CRL.

One of the significant characteristics of the organization in the forties and fifties was the total involvement of the company in the lives of the staff. Deep River was a company town and hence the company (DIL initially, and then NRC) was landlord to all inhabitants. But the relationship went far beyond this. The company controlled who could live in (and, in the very early days, even who could enter) Deep River and the type of accommodation they could rent. They also took great pains to make life in Deep River as attractive as possible in order to attract and retain top-level staff in spite of the relative isolation. This included the provision of a recreational director and staff, involvement in community affairs through provision of hospital

FIGURE 2.2
CRL – 1946
Distribution of NRC-directed effort



facilities, schools and services, help in planning and constructing recreational facilities, and even a free, weekend bus expedition to Pembroke for shopping. In spite of the rather primitive nature of many of the amenities of life, Deep River was regarded as a desirable place to live by most residents.

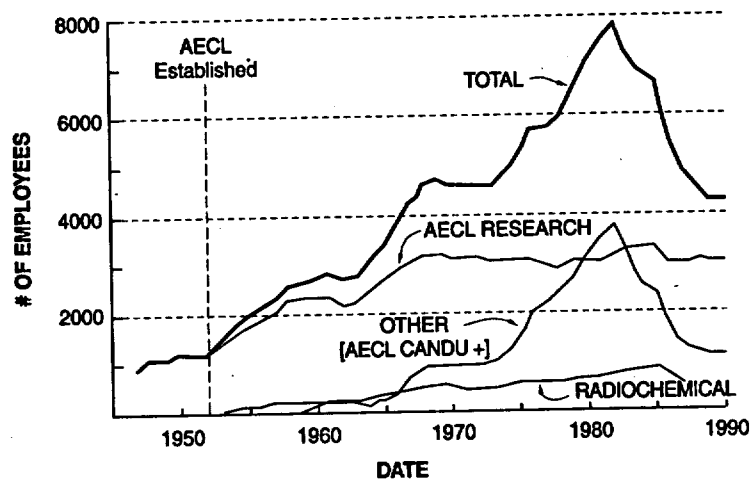
ORGANIZATION

By 1952, when AECL was established as a crown corporation, there were some 1,200 employees at the plant (CRL), about sixty buildings of a variety of sizes, and major scientific facilities comprising NRX, ZEEP and a Van de Graaff accelerator. The proposal to build NRU had been accepted and the reactor was under construction. (By the end of the period covered by this book (1985), the number of buildings had increased to about 150 and there were more major scientific facilities.) Initially, this incarnation of AECL Research was AECL in total. Of course, for most of its history, AECL managed a broader effort than just AECL Research; employees were distributed among various units of AECL over the period from 1952 to 1990 (see figure 2.3).

Within a few months of the establishment of AECL, the commercial products division of the Eldorado Mining & Refinery Company was transferred to it; this division formed the basis of the radiochemical unit, which was centred in Ottawa (see figure 2.3). The unit was dedicated to the packaging, marketing and sale of radioisotopes, and the development, manufacture, marketing and sale of equipment for the application of those radioisotopes (most notably medical therapy units and industrial irradiators). The radiochemical unit depended heavily on AECL Research for the production of the radioisotopes at CRL. Initially this was an exclusive dependence, but starting in the seventies other sources were tapped. In 1988, the unit was separated from AECL as two incorporated companies, Nordion International and Theratronics International (see chapter five).

AECL's main thrust immediately after its establishment was the development and exploitation of a Canadian nuclear power-reactor concept. This effort fell under AECL Research until the

FIGURE 2.3
AECL Employees



formation, in 1958, of the nuclear power plant division (NPPD) in Toronto. This marked the start of the effort labelled "Other [AECL CANDU +]" in figure 2.3. This is the effort that is within the AECL CANDU unit of AECL at the time of writing, plus the Corporate (Ottawa) staff. At various times in the past, it has included units that were named differently; e.g., NPPD, Heavy Water Projects, International Marketing. This effort grew rapidly in the sixties and seventies, reaching a peak in 1981-82 with work on the AECL 600 MWe power reactors for Quebec, New Brunswick, Argentina and Korea (see chapter ten), in addition to work for Ontario Hydro on its reactors.

The AECL Research effort that in 1952 was all at CRL expanded over time to include the Whiteshell Laboratories (WL) near Lac du Bonnet, Manitoba, and head office in Ottawa. Figure 2.4 shows the distribution of AECL Research over the period 1946-90. As might be expected, the total organization grew rapidly from 1952 to about 1971, when the first commercial CANDU came into operation, and has been relatively constant since. The decision to establish WL was made in 1959 on the grounds that CRL was at an ideal size for an applied laboratory of its type and further expansion would be counter-productive. A company townsite was

- bilateral technical exchange agreements with many countries,
- two-way technology transfer with Canadian industry,
- training programs and courses for external groups,
- commercial contract work for industry, both Canadian and international,
- attached staff from foreign laboratories,
- attached staff from Canadian industries and utilities, and
- close collaboration with utilities and, through them and AECL CANDU, with manufacturers.

As a crown corporation, AECL is an instrument of federal-government policies and is committed to following and promoting those policies.

OPERATIONS

General

In general, the industrial operations group at CRL and later the engineering services division at WL were responsible for:

- reliable maintenance and operation of the major facilities needed by the R&D group to achieve their mission,
- provision and maintenance of the buildings on the site, using either in-house or off-site resources,
- provision of some of the conventional services needed by the R&D group to achieve their mission, using either in-house or off-site resources, and
- major production activities, such as chemical extraction and isotope production.

There was also an additional implicit responsibility: that of maintaining a competent staff in those areas required to fulfil the above responsibilities. Since some of the areas were new, and external training unavailable, internal training programs were necessary. An apprentice program was established for the trades that not only served AECL well but also provided significant spin-off to industry in general and the nuclear power program in particular. Technical staff training programs were also set up and, in some cases, expanded to serve the needs of Canadian industry and utilities, as well as international customers and associates (e.g., India and Taiwan). For a few years in the early sixties, a reactor school for international mature students was operated at CRL.

Many of the activities involved in fulfilling these responsibilities were highly technical. However, other important, but less technical, support activities were provided by the operations group as well as other divisions in the laboratory organizations (see table 2.1). Some of these have been shaped by the nuclear mission of AECL Research:

- Of extreme importance to any R&D organization is easy access to a library that is comprehensive in areas relevant to the organization. Libraries established at CRL and WL by AECL Research have become the Canadian repository for nuclear-energy-related material and provide international access to this data. As well as standard library services, special

TABLE 2.1
Support activities – not highly technical

SHAPED BY NUCLEAR MISSION
Library & Technical Information Services
Public Affairs
Medical (Hospital)
Protective Services
Decontamination & Laundry
SPECIAL COMMENT
Workshops
Quality Assurance
Townsite Administration
Transportation
OTHER
Reproduction & Photography
Human Resources Development
Employee Relations
Personnel Services
Compensation
Office Services
Industrial Safety
Finance
Purchasing
Stores

FIGURE 2.4
AECL Research Employees

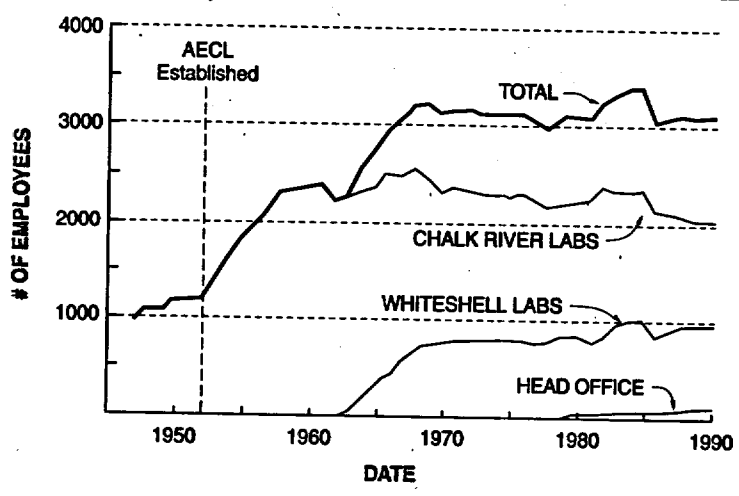
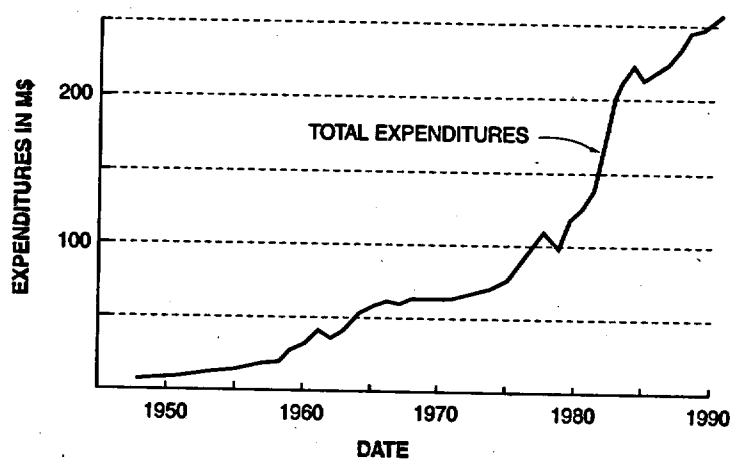


FIGURE 2.5
AECL Research Expenditures ($s of the year)



established at Pinawa, analogous to Deep River for CRL and about the same distance from its plant. The staff started to locate at WL in 1964. Staff shown in figure 2.4 as belonging to WL in 1962 and 1963 were actually located at CRL. WL eventually grew to be about half the size of CRL (in terms of employees and budget).

The major programs in which WL has taken the lead role over the years have been in the development and promotion of the organic-cooled CANDU power-reactor concept (CANDU-OCR) (see chapter sixteen), reprocessing for advanced-CANDU fuel cycles (chapter eighteen), CANDU reactor safety R&D (chapter fifteen), and waste management (chapter nineteen). At the same time, it has contributed to the majority of the other AECL R&D programs.

Figures 2.5 and 2.6 plot AECL Research annual expenditures over the years. AECL has a fiscal year running from April 1 to March 31 and the convention used has been to plot, for example, the expenditures for 1980-81 at 1981. Figure 2.5 shows expenditures in dollars of the year and figure 2.6 shows the same expenditures in constant 1985 dollars; the conversion is made by multiplying the expenditure in dollars of the year by the ratio of consumer price index (CPI) for 1985 to that for the year in question. Figure 2.6 is more meaningful in terms of the real value of the money spent, while figure 2.5 gives the actual amounts spent.

The ratios shown in figure 2.7 aid in an understanding of the implications of Figures 2.5 and 2.6. Figure 2.7 displays the following ratios: Capital/Total Expenditures; Parliamentary Appropriations/Total Expenditures.

As might be expected, the ratio of capital/total expenditures was much higher in the early years, when there were large initial investments in needed facilities and equipment. However, it is now some thirty years since those initial investments were made and higher relative capital expenditures will be required in the future if the viability of AECL Research is to be maintained.

Parliamentary appropriations made up some 90 percent of total AECL Research expenditures over the period up to 1980. (Note that in figure 2.7 no values are given for the period from 1967 to 1978, since it is difficult to determine the exact value of the AECL Research share of the parliamentary appropriations to AECL over that period. In every year of this period except

This division of responsibilities between R&D and operations was a large factor in the success of Chalk River as an effective nuclear laboratory. Each group was responsible for what it was best qualified to do, enabling each to concentrate on its own strengths. This also minimized the risk of conflict of interest in major facilities between the best interests of the experiment and those of the facility, to the long-term benefit of both. There was, of course, always a higher authority to resolve any serious conflicts.

Extensive cooperation and collaboration existed between operations staff and R&D staff, initially on a largely informal basis. There was a great deal of mutual respect between the two groups. Operations staff knew that they could count on R&D staff to help in resolving any complex technical problems that arose, while R&D staff relied on operations staff for aid in meeting unusual research-facility requirements. Projects often were carried out with participants from both the R&D and operations arms and from many subgroups within each. The participants were accountable to the management structure within their side of the organization and had to keep them well informed. However, a great deal of latitude was given to permit a close relationship at all levels so that much of the work could be carried out, and many decisions could be made, without going up one side of the organization and down the other. This system worked well in the early days (perhaps because budgets were not as tight as they later became) and relied greatly on management trust and responsible staff behaviour. Such a system led to difficulties in predicting and assessing the cost and manpower requirements of some projects. More formal procedures began to be adopted in the seventies as government emphasis on accountability increased.

As might be expected, there were differences, as well as similarities, in management style within each of the two arms. On the R&D side the style was akin to that in universities. There was a great deal of flexibility in management of the effort; e.g., work and leave schedules, job assignments, travel, communication. Managers were largely selected from internal candidates on the basis of their technical strengths and achievements, rather than their specific management training or experience. At the lower levels, there were, initially, few routine management chores involved, with administrative assistants being provided for many of these. This led to technical leadership as the main responsibility, and a collegial approach to program content and work assignments. On the operations side the style was more similar to that adopted in industry. While positions were largely filled internally, based on previous experience and performance, there was less flexibility, presumably due to the more clearly defined and shorter-term nature of their duties and objectives. Also, there was more emphasis on the actual management aspects of the managers' jobs. The differences in management style led, on occasion, to some friction, although this never became serious. On both sides of the organization, more emphasis was placed on management, financial and administrative skills and responsibilities as time went on, leaving managers less time for purely technical leadership.

The decisions on whether to use in-house or outside resources for the services required by AECL Research must have seemed fairly straightforward in the early days, given the isolation of the site and the sensitive nature of the work being done. Thus, with the exception of construction services, the decisions were made in favour of "in-house" for the majority of required services. While AECL Research profited from the dedication and commitment of the staff providing the services, there is little doubt that if the same decisions were being made today, a number of them would be reversed on the basis of better communications and transportation, growth in external availability of services, and changed attitudes.

While relatively isolated geographically, the laboratories of AECL Research have never been isolated technically. With management support this has been achieved through:

- close relationships with universities, including: scientific collaboration, shared use of both AECL and university facilities, contracts with universities for work in fields where they have relevant expertise, employment of summer students and visiting professors, authorization of AECL staff as adjunct professors and participation in projects of mutual interest,
- extensive participation in international committees and activities,

FIGURE 2.6
AECL Research Expenditures (constant 1985 $s)

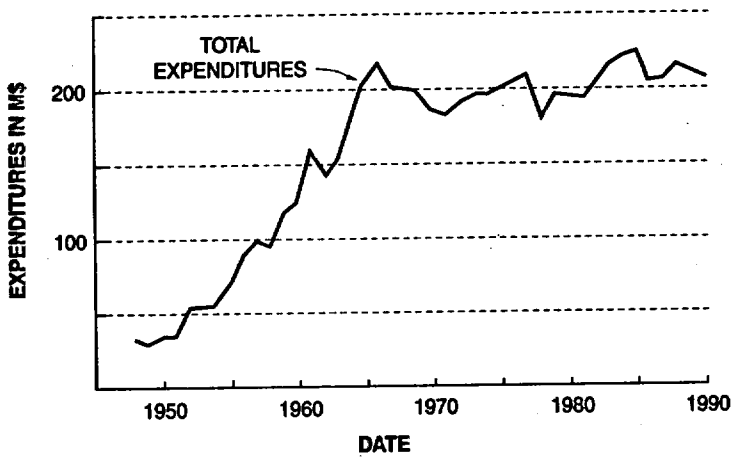
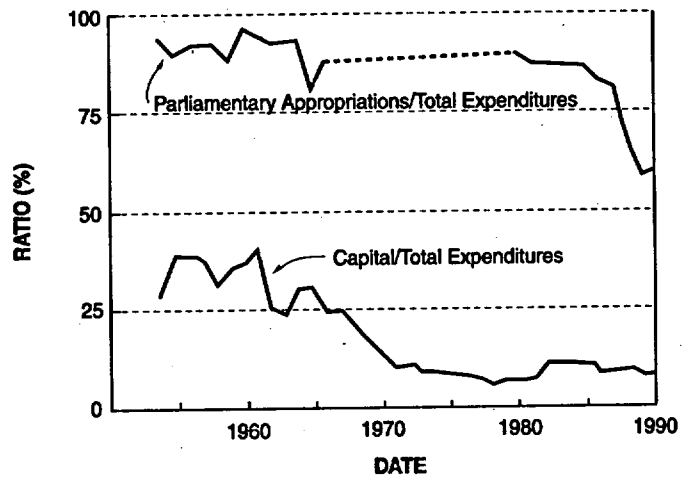


FIGURE 2.7
AECL Research: Informative Ratios



1976-77, when appropriations were 90.5 percent of total expenditures, AECL Research total expenditures were less than the parliamentary appropriation to AECL.) A considerable drop in this ratio occurred in the eighties, particularly after 1985, coincident with a government emphasis on increasing the leverage of government funds by a policy of having those who benefit most contribute to the cost. AECL adopted a more commercial approach to providing know-how and technology. The process was accelerated in 1985 when the federal government, in its budget, projected a 50 percent decrease over five years in its appropriations to AECL. While this decrease was never fully implemented, it initiated important changes in the operation of AECL Research. The biggest of these came from the negotiation of greatly increased support from provincial utilities, most notably Ontario Hydro, and the change in program control that resulted.

Figure 2.8 shows the organization chart just after AECL was established. The basis of the R&D part of the organization was scientific discipline rather than project involvement. Control of budgets was with these organizational units and this system worked well at the time.

The growth of CRL through the fifties and early sixties led to changes in organization, but these were essentially an expansion of the same basic type of organization. By 1959, while the number of employees had doubled from 1952, the number of divisions outside the administration, finance and medical services part of AECL Research had risen from four to seven and the number of associated branches from twenty to thirty. The industrial operations division had been divided into two, creating an operations division and an engineering services division, and two other new divisions had been created: the reactor R&D division and the research engineering division.

The establishment of WL and its growth required that changes in organization be made. In 1967 changes were also made in the internal organization at CRL. The result was an AECL Research organization of the type shown in figure 2.9, which existed in essentially the same form until 1986. Under this organization, CRL and WL were treated as parallel units under the AECL president. CRL, being the larger, had an extra level of management, with the divisions being grouped into research, applied R&D and plant administration and operations, each under a group head. Some other units or functions,

FIGURE 2.11
Distribution of AECL effort at CRL – 1952

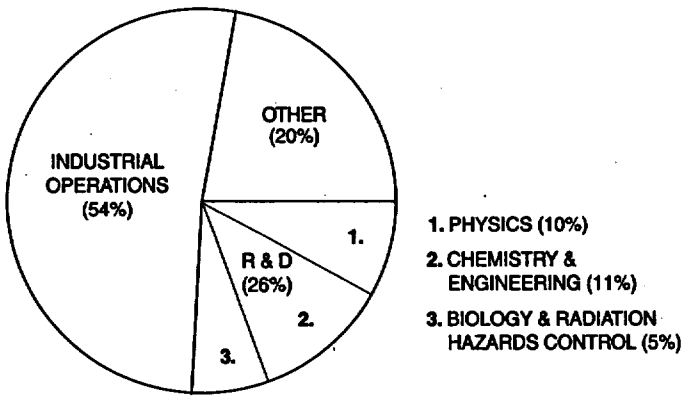
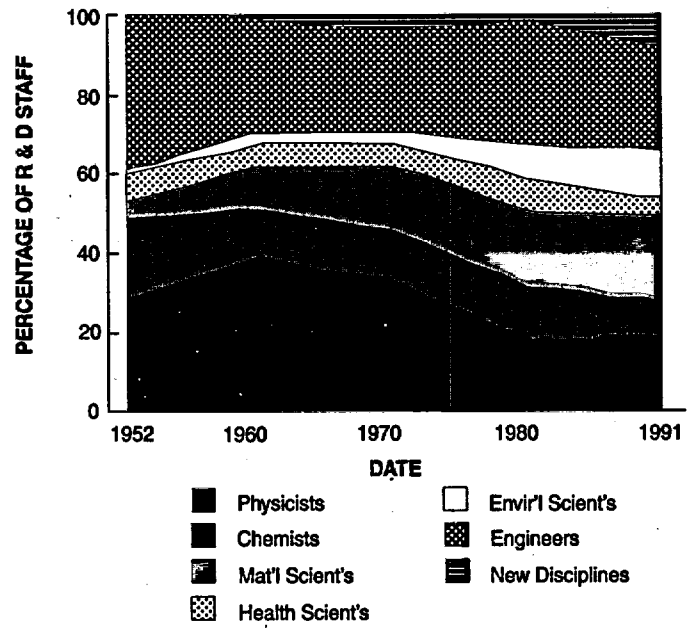


FIGURE 2.12
R&D Staff Disciplines



nature of the site and the need to provide services that in less isolated locations could be obtained externally. Nor was the boundary always clear cut between activities classed as R&D and those classed as Operations. The ratio of direct R&D effort to total effort, in terms of personnel, increased as AECL grew, as would be expected. It reached about 35 percent by the late sixties and fluctuated by a few percent around that value until 1985.

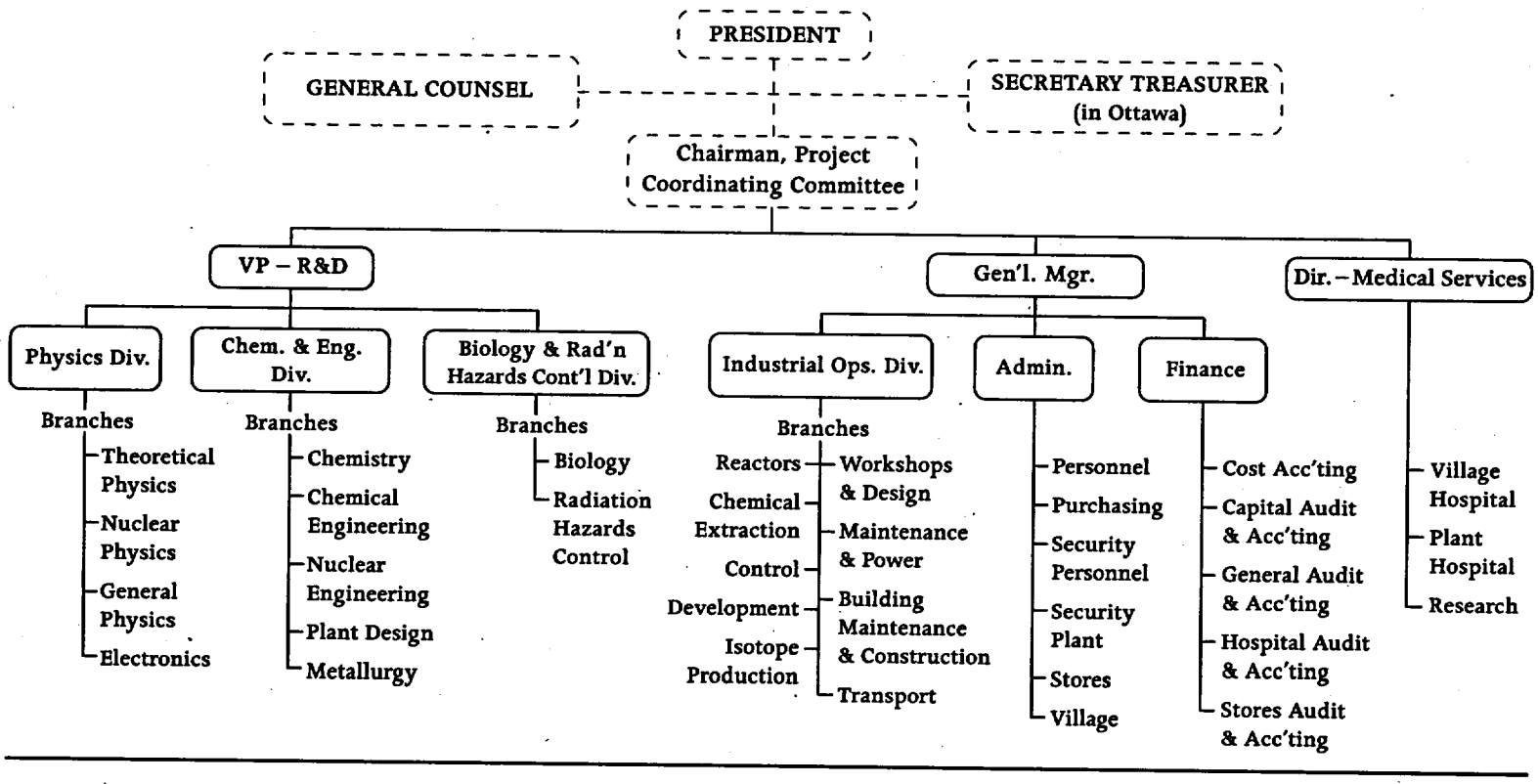
The application of this R&D effort in terms of programs and disciplines varied with time as well. In particular, there was a marked shift from basic to applied R&D (see figure 2.12). The category labelled new disciplines includes computer science, geological science, and human behavioural science.

MANAGEMENT STYLE

In the following discussion of management style, the effects of size should not be forgotten. In 1952 AECL was a relatively small and compact organization where strong personal contacts and relationships were widespread and important. By the seventies staff size had tripled and was geographically dispersed. This inevitably had an influence on management style. What had worked well in the fifties was no longer appropriate to the seventies and beyond for this reason alone, and there were several other factors encouraging change.

In establishing the first major nuclear site in Canada at Chalk River, an approach was taken that resulted in an effective and efficient style of management that served the program well through its infancy and for many years thereafter. There were two distinct arms: "R&D" and "operations" in the broadest sense of those terms. Initially, NRC was the R&D arm and DIL was the operating arm. In February 1947 NRC took over from DIL, but the separation of functions remained. When AECL was formed in 1952 this organizational structure was essentially maintained. As the program expanded, the same principle continued with NRU and other major facilities. Notable exceptions were ZEEP, ZED-2, PTR, the metallographic hot cells, the plutonium fuel fabrication laboratory and accelerators within the chemistry and materials division. These were directly under the R&D management.

FIGURE 2.8
AECL Organization at CRL in 1952

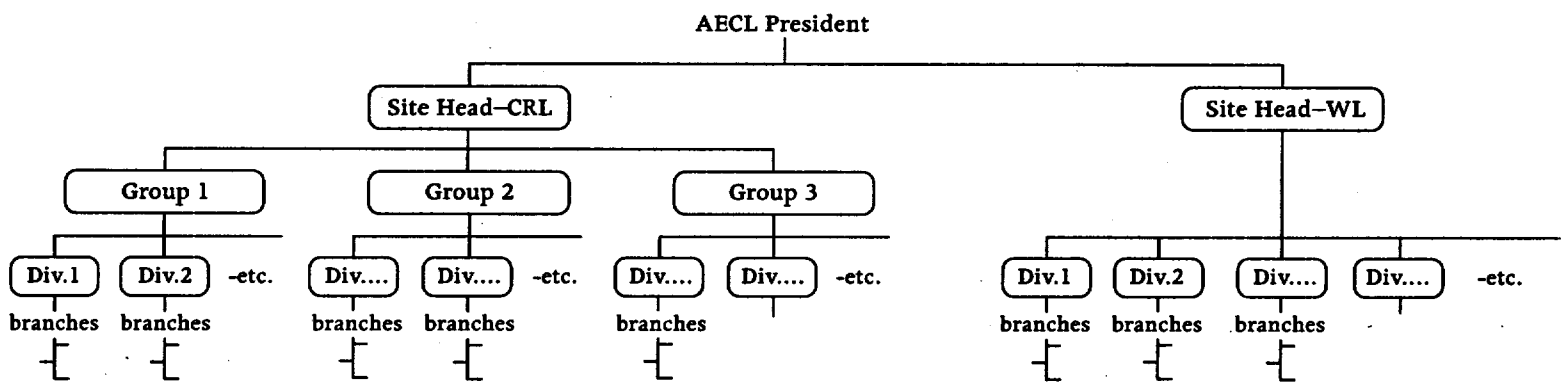


such as finance, quality assurance, and environmental authority, reported directly to the site head. By 1979 the number of employees had increased by fifty percent from 1959 and there were about twenty divisions and eighty branches within the AECL Research organization.

Starting in the seventies, there was an increasing emphasis on accountability and on programs and projects which had their own budgets and management; there was a more careful assignment of support costs to programs and projects. In 1976 an organization consisting of six steering committees – most controlling several working parties – under an R&D program committee was established, to plan and monitor all AECL Research R&D programs. This supplemented the line organization of figure 2.9, particularly in the areas of assigning priorities, monitoring progress and gathering budget information.

Just prior to 1979 the position of executive vice-president, AECL Research, was created, and the site heads reported to this position rather than directly to the AECL president. This was part of a general shift by AECL to a more decentralized structure and made AECL Research somewhat more autonomous in its day-by-day operations, while still being responsible to AECL Corporate for general direction and performance. At the same time, an AECL Research head office was established in Ottawa directly under the executive vice-president, whose role included coordinating functions in finance, planning and business development. Soon after, commercial offices were set up at

FIGURE 2.9
AECL Research Type of Organization 1967 – 1979



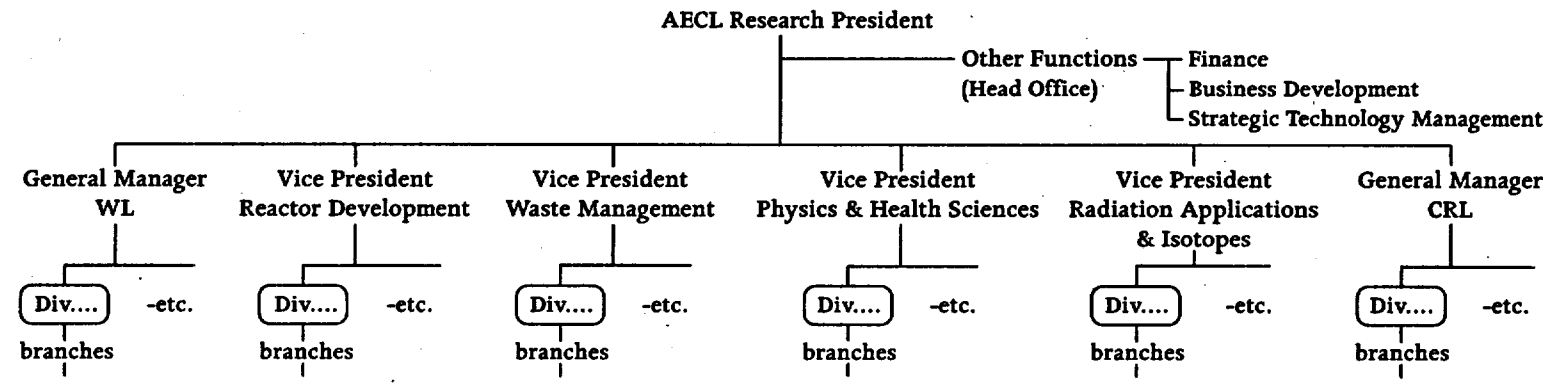
the sites (at CRL the head of this office had the status of a group head.) Otherwise, the organization remained essentially unchanged until 1985.

In 1985, faced with large cuts in federal appropriations over a five-year period, AECL Research took the first large step towards consolidating its site operations. The new organization was defined in 1985 and instituted in early 1986 (see figure 2.10).

Under this organization, although the sites were operated separately, the AECL Research R&D programs were managed across sites. (Site operations were consolidated in July 1990.) In these later years, particularly post-1985, there was an increasing tendency for major projects to control their own budgets, while using staff assigned from a variety of units.

Figure 2.11 shows the relative effort, in terms of personnel, associated with the main units of the organization in 1952; the direct effort associated with R&D was some 25 percent of the total, while that dealing with industrial operations was more than 50 percent. These ratios were influenced by the isolated

FIGURE 2.10
AECL Research Organization 1986



technical information services are provided to aid researchers in identifying and locating references.

- Public affairs specialists at the laboratories ensure that issues unique to the nuclear industry are handled properly and that staff and the public are kept well informed.
- AECL maintains medical and hospital services at the plants for employees.[1] The services ensure immediately available special expertise and treatment in the event of nuclear-related accidents.
- To satisfy national and international obligations, AECL must provide tight security against theft or dispersal of the potentially dangerous nuclear materials in its possession. Thus, AECL has its own protective services in terms of guards and firefighters.
- In many areas radioactive contamination of equipment and protective clothing is inevitable and once-only use would be prohibitively expensive. Decontamination and laundry services deal effectively and efficiently with the clean-up of equipment and clothing.

Comment is warranted on some of the other support services listed in table 2.1:

- There have been two types of workshops at the sites. The general workshops are the largest and include a planning and estimating function. They are under the operations arm and consist mainly of machine shops handling a wide range of jobs, from making experimental devices for researchers to manufacturing special parts for construction and maintenance; they include a small paint shop for site signage and some carpentry. In addition there have been some small machine shops within the R&D arm doing either more highly specialized work on a dedicated basis for the R&D staff, or providing a very fast service on the basis of rough sketches and personal discussions.
- Quality Assurance[2] was established formally as a separate unit within AECL Research in 1980, with the initial aim of introducing formal quality assurance programs to the manufacturing function. The concept was later broadened to one of total quality management; i.e., making customer satisfaction the central focus of the company and all its component parts. This concept was embraced by AECL Research in the late eighties with its Continuous Quality Improvement (CQI) initiative.
- Town administration was important in the early days of both sites, when Deep River and Pinawa were company towns. This importance diminished with the towns' evolution towards independence.
- Chalk River Laboratories operates a unique bus service to and from work for employees who live in Deep River, Chalk River, Petawawa and Pembroke. This service at one time involved a fleet of fifty buses. In the early days, the service was heavily subsidized by AECL. The subsidy was gradually removed and, by the late eighties, the service was fully supported by its users. A service involving transport of materials around the sites and to and from external points is also provided.

The other support services listed in table 2.1, while essential, had responsibilities common to those of similar units in many other organizations. Of course, the nuclear focus of the work did create specific differences as well as problems to be resolved.

For most of the period covered by this book, the operation and use of the large research reactors (NRX, NRU and WR-1) provided the main focus for the work of the laboratories and was central to their mission, strength, *modus operandi* and morale. Figure 2.13 indicates the scope of the effort involved: the central box represents the groups responsible for the actual operation of the reactors; the boxes in the lower half of the figure represent the groups whose support was required for the operation, and the boxes in the top half of the figure represent the groups using the reactor.

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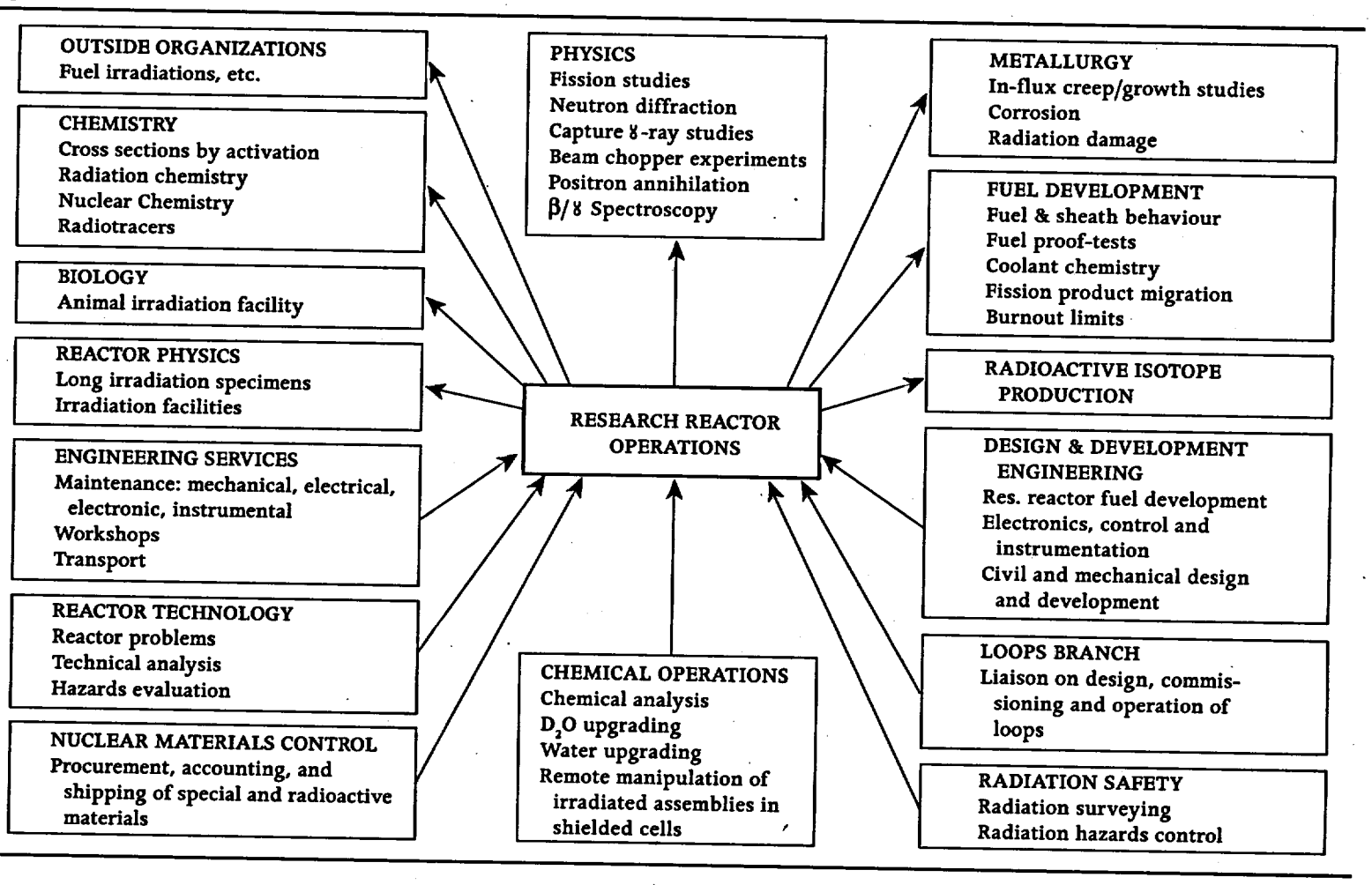
[1] To ensure that medical standards were not compromised for operational efficiency, these services were the responsibility of a senior authority, who was independent of "operations."

[2] Quality Assurance denotes a system of processes designed to ensure that the requirements for a product are well understood by the producer, and that the product meets these requirements. The driving force behind the development of quality assurance as a discipline in Canada came from the aerospace and nuclear power industries, but the discipline has very wide application.

FIGURE 2.13
Operation & Uses of Research Reactors



RESEARCH REACTOR OPERATION

Pre-1952 Canadian Research Reactor Experience

The decision to build NRX, for the dual purpose of providing a source of neutrons for research and for production of plutonium, was made in 1944. Flexibility was one of the key characteristics of NRX, and this paid dividends throughout its life. The flexibility allowed adaptation to changing requirements and allowed applications that were unknown at the time of the design.

Figure 2.14 shows NRX – a heavy-water-moderated, light-water-cooled research reactor. The main features of interest to production[3] and R&D activities are:

[3] The main production activity has been isotope production, but initially there was also plutonium and uranium-233 production.

- fifteen experimental holes, through which beams of neutrons (or other forms of radiation) could be extracted,
- fifteen self-serve mechanisms, through which small samples for irradiation could be easily introduced and removed,
- 198 vertical channels mainly used for fuel rods, but which could be adapted to house control and shut-off rods or various types of irradiation facilities, including loops (which are discussed in a later section),
- the central thimble, which could house a wider variety of types of irradiation facilities than the fuel-rod sites, and these in the highest neutron-flux location,
- two graphite thermal columns, in which there were relatively very few high- energy neutrons, most neutrons being in thermal equilibrium with the graphite, and
- the air-cooled J-rod annulus, with access via a ring of ninety vertical holes through the top shield. The annulus was originally used to produce uranium-233 by irradiation of thorium, but later used to produce other isotopes.

For many years after the startup of NRX, the main characteristics distinguishing it from other research reactors were its availability, for production and R&D, of high neutron-flux levels and relatively large volumes of more or less uniform neutron flux. These characteristics were exploited to the fullest extent. Even after the flux levels were matched and surpassed by NRU and other reactors outside Canada, the combination of flux and available volume kept NRX competitive with foreign reactors in some fields (e.g., fuel element and assembly testing, isotope production) for many more years. In addition, NRX served even longer as a very useful supplement and back-up to NRU.

Some of the key staff who were to be responsible for the operation of the reactor received technical training at the Montreal Laboratory. The "Montreal Lectures" were the basis for this training and the written versions proved to be of great use to new, inexperienced staff for many years afterwards.

Construction of NRX was completed in the late spring of 1947 and everything seemed to be ready for startup. For a startup with fresh fuel in the reactor, specially sensitive neutron detectors were required and had been installed. Because the isotopic purity of the heavy water was to be maintained in the region of 99.8 percent, drying of the system was attempted to remove the last traces of ordinary water. There was evidence that water was entering the system. An intense search traced the in-leakage to porosity of water-cooled castings in a blower intended to circulate the helium cover gas. After the faulty equipment was replaced, water vapour persisted in the system despite the drying. Further searching located a U-shaped pipe that had escaped earlier notice. When that was drained of seven kilograms of water the reactor system was successfully dried. Heavy water was then put into the system and the reactor was brought to criticality at 6:13 a.m. on July 22, 1947.

The power level of NRX was gradually increased from the initial value of a few watts to 250 kW in September, 2 MW by March 1948, 12 MW by May 1948 and 20 MW by September 1948.[4] The power was limited to 20 MW by the capacity of the moderator cooling system. As an interim measure, the river-water cooling for the heavy-water heat exchanger was fed from an available higher-pressure system, enabling operation at power levels as high as 27.5 MW when the river was cool enough.

The operation of a system exposed to intense ionizing radiation presented novel challenges (in addition to more standard ones[5]). It was known that the radiation decomposed water into its constituents, oxygen and hydrogen, and a system had been

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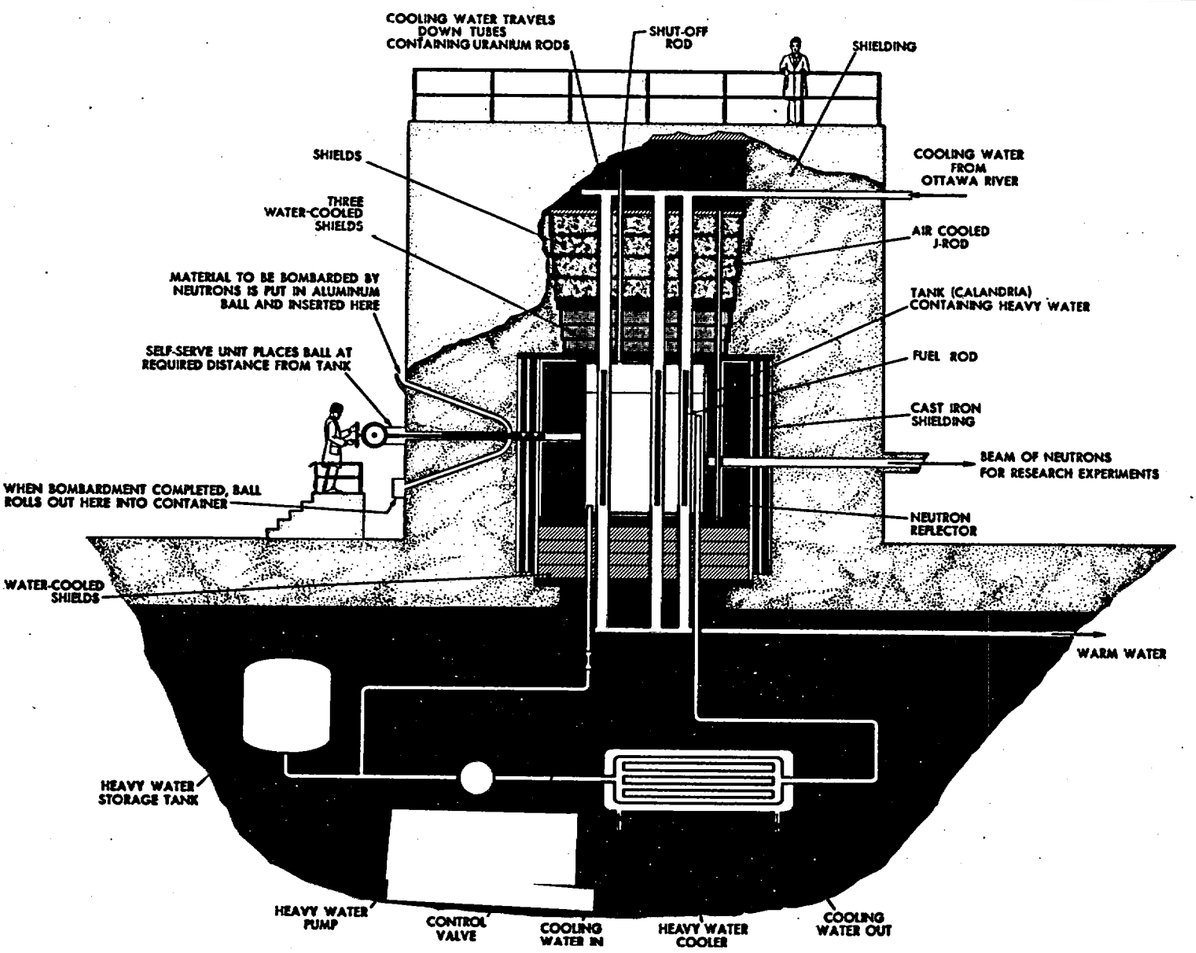
[4] During the initial approach to 100 kW, a jitter of a percent or so appeared on the power meter (which was sensing neutron flux) at about 80 kW. Lowering the power slightly caused the jitter to vanish, but it reappeared at 80 kW. This went on for two or three days; the control room became a meeting place for senior scientific staff who hypothesized about the cause of the "frantic mouse," as the phenomenon was called. Theories ranged from a loose isotope-production capsule bouncing in the cooling air flow to a restructuring of heavy water at a specific temperature. The effect disappeared after a couple of days.

[5] Employee's recollection: "In the winter of 1948 I was given the job of finding a way of preventing algae formation in the inlet end of the NRX fuel rod annuli. I tested a few things in the lab and the problem disappeared as if by magic. It turned out to be due to cold water algae in the river water that disappeared as soon as the water warmed up. I never had an easier problem to solve in my career."

FIGURE 2.14
NRX Research Reactor



provided to circulate the moderator cover gas (helium) through catalysts to recombine the oxygen and heavy hydrogen. The only relevant experience was with a 300 kW heavy-water reactor in the United States, and that indicated high decomposition rates. There were other more subtle effects, which led to acidification of the moderator. Instead of following the American practice of adding deuterated ammonia to neutralize the harmful acids, the NRX operators distilled the heavy water and used newly available ion-exchange resins to maintain purity.[6] Improvements were also made to equipment for removing nitrogen and other impurities from the helium. The result of these measures was such a low rate of decomposition that concern shifted to fear that the low rate was deceptive, and that oxygen and deuterium were accumulating in the heavy water and might be released in a burst of an explosive mixture of the two gases. The fear was found to be groundless, and the moderator system behaved well, an encouraging situation for future power reactors.

Heavy-water leakage control was a major ongoing effort in NRX. To ensure that heavy-water leaks at flanges, valves and other fittings could be detected quickly, heavy-water detectors, called Beetles, were installed wherever the leaks might occur. Any water dripping into the tray of the Beetle collected in a small depression at the bottom and made contact with a probe. An alarm rang in the control room, indicating a water leakage, which was immediately investigated by the operating personnel. (The NRX average net heavy-water operating loss from 1955 to 1960 was 12 kg per month.)

In the early days of NRX operation a great deal of effort went into understanding its behaviour. While considerable experimental information on reactor characteristics had been gathered from American sources, and in the Montreal Laboratory and ZEEP, most of this was at very low power and flux levels, and at room temperature. Characteristics of NRX had been mainly estimated from the basic properties, not all of them well known, of the constituent materials. It was important that these estimates be verified by direct measurement.

The most important characteristics, from the viewpoint of effective operation and safety, were those associated with the effects on reactivity of changes in the composition of the reactor core (through insertions or removals) or of the materials within the core, changes in the temperatures of the core constituents, changes in the relative positions of core constituents, and changes in the neutron flux. To make quantitative measurements of these effects, a nominal geometric reactivity scale was established for NRX, whereby changes in moderator level could be interpreted quantitatively as changes in reactivity (see Box A).

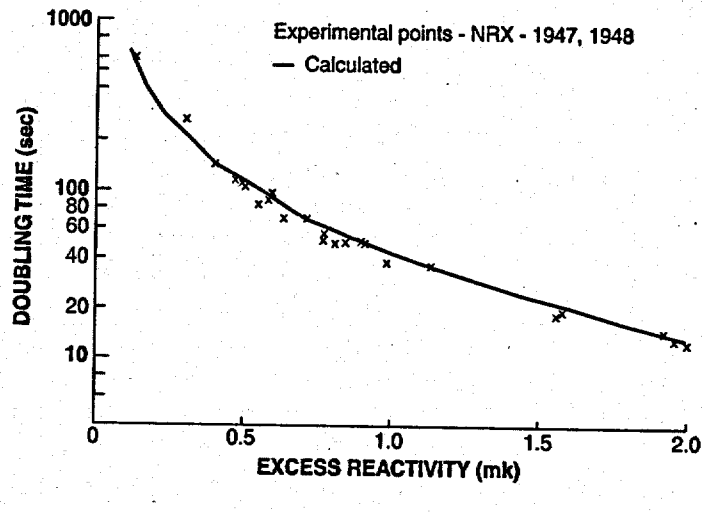
The general method for making the measurements was to start with a critical reactor, make the physical change to be measured, and then restore criticality by changing the moderator level. The change in reactivity due to the physical change was then the negative of the change in nominal geometric reactivity. As mentioned in chapter one, many measurements were made in the period 1947-52. Measurements of particular value to the reactor operation were temperature and power coefficients of reactivity: the ratios of change in reactivity to change in component (moderator, rod assembly, etc.) temperature and to reactor power, respectively; the change in reactivity associated with long irradiation of fuel rods; the differences in reactivity associated with differences in fuel rod design; the reactivity effect of inserting the Hot Loop in the central thimble; the reactivity effect of removing light-water cooling from the in-reactor portion of fuel rods, and xenon

[6] Employee's recollection: "During the first year of operation the NRX moderator became progressively more acidic. Aluminum dissolved from the calandria, then precipitated in the heavy water giving it almost the appearance of skimmed milk. The moderator had to be completely distilled but in six months the problem was as bad as it was before distillation. Fortunately, at about that time, ion-exchange resins first came on the market. I was given the job of seeing if this new product could be used to clean the NRX moderator. The test work was done on the main floor of NRX. I clearly recall the superintendent and assistant superintendent anxiously standing by to watch the first tests, both armed with cotton batting to mop up any drops of heavy water that were spilled. The tests worked, columns were put on a side stream of the moderator circuit, and in seven days the moderator was as clean as when it went in the reactor. Without the timely commercialization of ion-exchange resins, the operation of NRX and its dependent programs would have been much more difficult."

BOX A GEOMETRIC REACTIVITY SCALE FOR NRX

As discussed in chapter eleven, the reactivity of a reactor, k is defined as the ratio of neutron populations in successive generations (with the value of k being =1 for a critical reactor, <1 for a subcritical reactor and >1 for a supercritical reactor). Changes in reactivity are usually expressed in millik (mk) with 1 mk being a change of 0.001 in k. A change in moderator level changes the probability that neutrons will leak from the reactor, and hence changes k. The relationship between moderator level and reactivity can be determined theoretically and, from this geometric reactivity scale, the reactivity change associated with any moderator level change can be determined. The scale can also be determined by measuring the time response of the neutron flux to a change in moderator level from the critical level. Measurements were made of the time for the flux to double after a moderator level change from critical (done at low reactor power to minimize changes in reactivity during the measurements due to temperature changes), allowing enough time to elapse after the addition of the excess reactivity to give an accurate approximation to the asymptotic doubling time. The results from both methods agreed within the uncertainties associated with them (see figure 2.15), giving confidence in the geometric reactivity scale adopted for NRX. To give some feeling for the relationship, at a moderator height of 230 cm an increase in level of 10 cm gives a reactivity increase of 5 mk.

FIGURE 2.15
Asymptotic Reactor Doubling Times vs Excess Reactivity



poison. It was particularly important to the efficient operation of the reactor that xenon poison be understood, since, for example, it was responsible for the relatively short window of opportunity for quickly restarting the reactor after a shutdown from high power. The length of this window depended on the recent reactor operational history, and was typically an hour or less. The usual result of missing the window was a delay of a day or more in getting back to power (see chapter eleven).

About the time NRX began operation, information was received from the United States that, in their experience, when metallic-uranium fuel rods had produced an energy of 300 megawatt days per ton of uranium (MWd/tU),[7] mechanical deterioration such as pimpling and distortion would require the removal of the rods from the reactor. The irradiations of the middle portions of the NRX rods, where the rate of energy production was highest, were kept under review. Occasionally,

[7] There was confusion over what was meant by a ton. Canadian engineers used 2,000 lbs. which is the Canadian ton. The United Kingdom used 2,240 lbs. which is the British ton, and some scientists used a metric ton, which is 1,000 kilograms, (2,205 lbs). Finally, it was decreed that in this matter a ton meant a metric ton (then to be called a tonne, now a megagram (Mg)), and some discrepancies of 10 percent or so in people's numbers vanished. The preferred energy output unit is now megawatt hours per kilogram of uranium (MWh/kgU). {300 MWd/MgU=7.2 MWh/kgU}

rods were removed and examined for pimpling. No serious trouble developed at 7.2 MWh/kgU and irradiations continued, with the middle portions of some rods eventually reaching 72 MWh/kgU. The attainment of such long irradiations was an important factor in the first quantitative Canadian proposal for a nuclear power plant.

Some rods did fail because the uranium expanded and split its aluminum sheath. These failures were detected by instruments that monitored the effluent cooling water for radioactivity. The position of a failed rod was approximately indicated by installed equipment, after which the specific rod was located by a manual radiation survey. The rod was then removed.

Even intact rods could not be left in the reactor indefinitely, because irradiation burns up the fissile material and so reduces a rod's contribution to reactivity. Fresh uranium was needed to keep the reactor operating. In October 1949, a planned refueling at one rod site could not be completed, because the fuel rod was stuck in place. A quick survey showed that nine rods were either immovable or movable only by a few feet. Shortening of the uranium under irradiation had caused wrinkles, like those around a loose sock, to develop in the outer aluminum tube. Some of the wrinkles caught on the surrounding calandria tubes. After considerable ingenuity and effort, all the rods were successfully removed.[8] The stuck rods were from the second batch of rods, in which the rolling conditions had produced a metallurgical texture conducive to shrinkage. There was no satisfactory record of the rolling conditions that produced the original batch, and much effort was required by metallurgists to discover a procedure that would not lead to undue shrinkage. There is no doubt that the project would have been in jeopardy if the original rods had jammed themselves into the core. Several incidents in the intervening two years had given the operating and design staff experience and confidence in dealing with unusual events in a complex radioactive environment.[9]

By late 1948, a simple electrical circuit had been added to keep the reactor power at any chosen level automatically. This was the beginning of a Canadian tradition of automatic control, with the result that Canadian power-reactor designers have pioneered the use of digital computers for reactor control. For many years, operators of research reactors in the United States were required to have a human manipulating a control knob to maintain the power level. Canadian and American views differed over the question of whether it is preferable to give the operator freedom to move about, checking instruments in the control room, or to have him sit for hours doing the boring task of keeping a spot on a galvanometer scale at a fixed point.

In 1949 a pneumatic carrier tube was constructed to carry irradiation samples from the research chemistry building directly to a central fuel-rod position in NRX (some 900 feet).[10] Samples could be installed from the top of the reactor or from the chemistry building into either a central core position or into a rod in the J-rod annulus. The irradiated samples could then be received either at the top of the reactor or in one of four

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[8] The removal of these rods provides a good example of the highly imaginative procedures and equipment, coupled with great skill, that were frequently required to successfully deal with problems that arose. The fuel rod assembly was about 9.4 m long, with the mid 3 m containing the uranium. The solid uranium-metal fuel was contained in an aluminum sheath about 3.5 cm in diameter that had 3 fins to centre it in the 4.3 cm diameter flow tube. The normal air gap between the calandria tube and flow tube was less than 2.5 mm. The decision was made to saw through the wrinkle. A tubular saw was built to close tolerances. It had to slide down through the 2.5 mm annulus. Before this could be used, the upper stainless-steel shielding portion of the fuel rod assembly had to be removed because it was greater in diameter than the fuel section. Using extensive mock-up tests that were an exact physical duplication of the situation, including the wrinkles, a method was developed to disjoint the upper portion of the rod assembly and remove the wrinkles.

[9] Employee's recollection: "This was one of the earliest incidents that established the overall procedural pattern that has been diligently and successfully followed up to the present. You establish a team and restrict the time they spend on the job in spite of their enthusiasm – in this case no more than seventy hours per week. You do extensive and precise mock-up testing of all operations. This incident also pointed out the importance to the operation of what we used to call the gadgeteer."

[10] This facility was nicknamed the rabbit, in recognition of its speed and rapid starts and stops.

receiving stations in different laboratories in the chemistry building. Compressed air was used to drive the capsule (at up to eighty kilometres per hour) and also to cushion the capsule as it entered the rod position and stopped at the elevation of maximum flux. The capsules were about 1.5 inches (38 millimetres) long and 9/16 inches (14 millimetres) in diameter. Although the maximum sample size allowable was about one gram, owing to heat-removal limitations, this was more than adequate in most experiments, thanks to the high neutron flux in NRX.

As mentioned previously, the power of NRX was limited by the capacity of the moderator cooling system. An additional heavy-water heat exchanger, two additional heavy-water circulating pumps and an additional cooling water pump were added to the system. By mid-1950 the reactor power limit was 30 MW.

The first two loops (see later section) were installed in the central thimble of NRX in 1951. These were re-entrant loops (i.e., the coolant both entered and left from the top) and were referred to as the Cold Loop and the Hot Loop. They were used by the Americans in support of Admiral Rickover's nuclear submarine program. This marked the beginning of a long relationship (lasting until 1981) in which the Americans made extensive use of loops in NRX for various tests.[11] The use of loops in research reactors became an important element in the Canadian nuclear power program.

By 1952, fuel rods with a cooling-water annulus 0.071 inches (1.8 millimetres) thick came into service, replacing rods with 0.100 inch (2.54 millimetres) annuli. This reduced the amount of ordinary water in the core, thereby increasing the reactivity.

RESEARCH REACTOR OPERATIONS HIGHLIGHTS

NRX Highlights

The NRX accident in December 1952 was a severe but temporary setback to research reactor operation at CRL. Prompt and effective action in the period immediately following the accident and for the next few days (i.e., reactor shutdown, personnel evacuation, temporary storage and subsequent disposal of highly contaminated coolant[12]) limited its effect on personnel and the environment. As reported in chapter three, a study almost forty years later revealed no excess cancers or leukemias – above those expected for the general population – among personnel involved in the cleanup. It has been estimated that the maximum dose a member of the public could have received due to the accident was less than 1 millirem (0.01 mSv).

The core of NRX was rendered useless by the accident, and there was no experience at that time from which to derive firm confidence that it could be replaced. Nevertheless, a decision to repair NRX was quickly made and it was back in operation in fourteen months.

The restoration of NRX proceeded in parallel with actions to contain and remove the radioactive materials deposited in the building by the accident. Most of the fuel rods could be removed in the normal manner, but removal of the damaged rods was more difficult. Some fuel residues were lying on the bottom of the calandria. Removal of these residues and the decontamination of the basement involved most of the staff at CRL and resulted in some workers receiving radiation doses around the regulatory limit then in force. Extra workers were supplied by the Canadian army and the American navy.

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[11] Employee's recollection: "Early on the USAEC recognized the value of NRX. In the fall of 1948 a special irradiation of uranium oxide in conjunction with neptunium separation studies was done for Argonne. The shipments were made using a special United States Army aircraft. The irradiation was done in NRX because there was no reactor elsewhere with sufficient capability to do the investigation. This was the first formal cooperative experiment where Canada supplied the reactor facilities in exchange for the results of the experiment – something that Dr. Lewis was always very keen on."

[12] About 4,500 cubic metres of cooling water, containing 10 kilocuries of fission products, flooded the NRX basement. An 1,830-metre, 10-centimetre diameter insulated pipeline was laid within nine working days and the cooling water was pumped to the waste management area.

Removal of the calandria on May 22, 1953 was a difficult and important preliminary step towards reconstruction.[13] During the reconstruction, there were improvements over the original NRX. Changes were made to many of the control and shutdown systems to reduce the probability of another incident.[14] New inner shields above and below the core removed the concern that at higher powers differential heating through the original shields would cause undue buckling, and this permitted the power limit to be raised to 40 MW.

The reactor was back in operation on February 16, 1954, and by early April it was at a new operating power of 40 MW.

Following the restoration of NRX, increased user demands for more tray rods, fast-neutron rods, loops and other irradiation facilities put space at a premium, both within the reactor core and within the NRX building. It also meant that more reactivity was needed than could be provided by the standard natural-uranium fuel. Special rods called booster rods were designed, made and used to provide increased reactivity. These booster rods were fueled, in general, with enriched uranium (in the form of a uranium-aluminum alloy), but a few were enriched with plutonium (in the form of a plutonium-aluminum alloy) that had been separated from spent fuel and made into booster-rod fuel at Chalk River.

The fast-neutron rods were used to irradiate samples in a higher fast-neutron flux than was normally available. Over the years the design varied, but it consisted basically of a sample irradiation space within a fuel annulus situated in a normal fuel-rod site. By the early sixties, six fast-neutron rods were installed in NRX.

Tray rods were used for isotope production. These were air-cooled assemblies placed in normal fuel-rod sites. They comprised an open-sided tray assembly with clips to hold thirty sample capsules at various elevations in a rod. The tray rods could be removed from the reactor and the samples changed without shutting down the reactor. By the early sixties there were fifteen tray rods in the reactor core and one in the J-rod annulus. In addition, many miscellaneous experimental rods were installed at various times.

In 1955 one of the plutonium-enriched booster rods in NRX failed, puncturing the calandria tube in which it was located. An estimated 100 grams of alloy containing 20 grams of plutonium, plus some of the light-water coolant, was lost to the heavy-water moderator. It was determined that failure had been caused by the aluminum sheath that surrounds the fuel alloy bulging away from it, causing the fuel temperature to rise enough to cause the failure. All fuel of this type was removed from the reactor core. The NRX heavy water was downgraded to about 97 percent deuterium oxide, but all the heavy water was recovered and reconcentrated. The calandria vessel was cleaned[15] and the damaged calandria tube replaced *in situ* in seven weeks.

A facility called the Hydraulic Rabbit (to distinguish it from the Pneumatic Rabbit already described) was installed in NRX

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[13] About a year after the event, *Popular Science* published an article about the removal of the calandria under the title "An Atomic Dragon's Eerie Funeral". The following is excerpted from this article. "This is the story of a funeral – the strangest funeral in the history of man. The coffin was a huge canvas bag, the grave a great hole and the mourners atomic scientists. The corpse, was the [NRX] calandria – the largest and most dangerous radioactive object that has ever been handled. A long whistle blast warned that the funeral procession was underway. In buildings along the way pens of radiation monitoring instruments went off the charts. The [burial] ground will not go unvisited – but so formidable is this atomic dragon, even in death, that it will be perilous to approach for some time to come."

[14] In the years following the start-up in 1954, the control and shutdown systems were more or less completely changed - the location of the control console was changed, automatic power control was introduced, the weir box for heavy-water-level control was removed, a new heavy-water-level control was introduced, and a new dump system was installed for shutdown.

[15] Employee's recollection: "After the plutonium-aluminum fuel-rod failure there were a lot of flakes of plutonium-aluminum lying on the bottom of the calandria that could not be removed by flushing. If they remained they would have caused hot spots on the inside of the calandria with resultant damage. Bill Stevens had the idea of putting soda water in the calandria to float the flakes so they could be flushed out. It worked like a charm and possibly avoided the need for a second calandria replacement. As it turned out only the calandria tube had to be replaced. I think they got the soda water from a local bottling works."

in 1958, to carry out a large number of short-term irradiations of experimental fuel. Each sample was loaded in the reactor hall basement, and propelled by cooling water through a 7/8 inches (22 millimetres) stainless-steel tube into the highest flux position in a fuel site, where it was held by a retractable stop. When the sample had been irradiated for the required time, the stop was opened and the sample was pushed by the cooling water to the top thermal shields. After a period of decay it was removed to a shielded flask.

Studies of alternative fueling schemes for NRX had been initiated in 1957 as a result of indications from the United States that their agreement to purchase spent fuel (with its plutonium content) from NRX and NRU would be terminated within a few years. Without a market for the spent fuel, the economics for operation of the research reactors would change significantly. Net neutron production from the fuel, rather than power, was the primary characteristic of value for R&D applications. In 1959 the decision was made to convert NRX to use a combination of two types of fuel: i) natural uranium-dioxide and ii) highly-enriched uranium-aluminum-alloy rods. The actual conversion started in the spring of 1962 and the main part was over in about six months, with no break in the normal operation of NRX, and with no structural changes.

The rationale for the choice was fourfold:

- the reactor would have the capability of accepting higher production and experimental loadings,
- a higher flux, by approximately 50 percent, could be achieved in production and experimental sites without changing the reactor power,
- the life of uranium-dioxide fuel was expected to be considerably longer than that of the uranium-metal rods, and
- the experience to be gained in the production and irradiation of uranium dioxide on a fairly large scale would be useful in the power-reactor development program.

A reference loading was chosen that consisted of three roughly defined fuel zones: an outer zone of solid uranium-dioxide rods, an intermediate zone of annular uranium-dioxide rods plus enriched uranium-aluminum-alloy rods, and a central zone of the enriched uranium-aluminum-alloy rods. The experimental and isotope-production facilities were in the intermediate and central zones.

The procedure adopted was to shift over to the reference loading, as occasion warranted, keeping the flux constant by lowering the reactor operating power. On a date announced six months in advance, the reactor was returned to normal power, with a resulting abrupt 27 percent increase in neutron flux. This procedure allowed adequate preparation of test fuel and other irradiation facilities to accept the flux increase. Further smaller increases were made as the reference loading was completed, with the integrated increase of about 50 percent. Some years later, when it had served its purpose, the uranium-dioxide fuel was gradually eliminated and fueling was entirely with enriched uranium-aluminum alloy. This conversion had the effect of multiplying the capabilities of NRX several-fold, thereby extending considerably its productive life.

In the summer of 1963 the first corrosion failure of a calandria tube, under normal usage, occurred in NRX. Sealing devices, consisting of removable plugs with expanding metal washers, were used to seal the ends of the tube. This permitted reactor operation without leakage, but the lattice site was not available for use. Tubes continued to fail for the same reason over a number of years. For some time these were plugged in the same manner, leading to more unusable sites. A tube-replacement program was then initiated, which required a large amount of ingenuity and technical expertise in remote work, but retained the usability of the site where a tube was replaced.

A spare NRX calandria was on hand on the main floor of NRX, which included modifications to allow for three large loops and four large fast-neutron irradiation facilities, as well as other improvements. It was finally decided to replace the NRX calandria. Taking advantage of the experience gained in 1953, this was done in a relatively short time period in 1970.

In the eighties, calandria tubes again began to fail in NRX. The continued operation of the reactor was possible only because the efficient and effective work of the operators, coupled with the expertise of the designers, made replacement of calandria tubes routine.

Taken as a whole, NRX operated extremely well and reliably, and enjoyed a long and productive life.[16] In the eighties its role was almost exclusively one of backup to NRU for isotope production. On March 13, 1986, NRX was designated as a nuclear historic landmark by the American Nuclear Society. The plaque that was presented hangs in NRX and reads: "NRX is the world's longest-operating research reactor, beginning operation July 22, 1947 – an important facility for isotope production, fuels and materials testing, and basic research."

NRU Highlights

The success of NRX, along with uncertainty regarding its useful life, led to consideration of a replacement as early as the end of 1947. It was also expected that the sale of plutonium to the United States would greatly defray the costs. By the end of 1950, the government had approved proceeding with an improved high-power, heavy-water research reactor, to be known as NRU. Design and site preparation were well advanced by 1952, and construction was completed in 1957. Startup was in late 1957, with criticality attained on November 3.[17]

While NRU was similar to NRX, in that it was a heavy-water-moderated reactor designed to be fueled with natural uranium, there were fundamental differences:

- Power – at 200 MWt the power rating was five times that of NRX,
- Flux – the maximum thermal neutron flux level was five times that of NRX,
- Size – the effective core size was 40 percent larger in volume than NRX,
- Coolant – the reactor coolant was heavy water, not light water (which NRX used), one result being that there were no calandria tubes (there are seals between the cylindrical reactor vessel and the top and bottom headers),
- Fueling – fuel could be replaced on-power, whereas NRX required a shutdown, and
- Reflector – light water was used as the reflector, not graphite (which NRX used).

The main features of interest for production and R&D activities included: 227 lattice sites, of which 221 were available for use; three lattice sites with access from top and bottom for engineering loop experiments; horizontal holes of various sizes (max. 30 cm diameter), of which two pass through the lattice; a thermal column; and forty-six available positions in the J-rod annulus. Special tray rods, capsule rods and fast-neutron rods were developed for irradiations. NRU also had a pneumatic carrier serving the chemistry laboratories and the universal cells.

Close control of heavy-water spills and leakage is important not only because heavy water is very expensive, but also because when it is irradiated for long periods, as research-reactor moderator is, the concentration of tritium builds up and presents an internal radiation hazard. Immediately following NRU startup, heavy-water leakage was detected from the lower seal between the vessel and the bottom header. This problem was solved by tightening all vessel bolts, imposing a limit on the temperature difference between the heavy-water moderator and the light water in the reflector, and maintaining a helium pressure on the bottom interseal space.

Heavy-water leakage continued to be a problem, a result of the structural complexity arising from the provisions made for neutron experiments and irradiations. In 1959 a heavy-water leak from one of the through tubes into the J-rod annulus was found. It was repaired by rolling in a long, closed tube (referred to as a thimble), and a similar leak that developed later in the other through-tube was repaired in a similar manner. In 1962 a major leak was traced to cracks in stainless-steel pipes, located in a position with limited access, that carry heavy-water coolant

[16] So successful was NRX that two copies were built, with Canadian help, in foreign countries: the Canada/India Reactor, CIR (later renamed CIRUS), in the early sixties in India, and the Taiwan Research Reactor, TRR, in the early seventies in Taiwan.

[17] Soon after startup, it was found that there was 33 percent excess absorption in the fuel-coolant tubes, caused by excess boron in the aluminum of the tubes. The result was a loss of seven mk in reactivity. The solution was more stringent specifications for the aluminum.

from the underside of the top header. A seal, either permanent or readily removable, had to be designed to eliminate the leakage. An epoxy resin that could be remotely installed was used. It was subject to radiation damage but could be easily removed.[18] The resin was used for periodic replacement of the seal over a period of about ten years before a suitable mechanical sealing device was designed and installed.

In the early operation of NRU, fuel failures were a major problem. In May 1958, during the removal of a failed fuel rod, a piece of uranium fell out of the removal flask into a pit at the top of the reactor, and caught fire. The building was severely contaminated and it was three months before the reactor could be started again. The cleanup operation involved work in areas of high radioactive fields and required the use of many staff (some of them inexperienced) to keep individual doses within limits. It marked the first time television was used at Chalk River to effectively control and monitor the work of inexperienced personnel.

In the commissioning of NRU, problems were encountered with the heavy-water pumps caused by stainless steel running against stainless steel – the result of a communication failure between designers and manufacturers.[19] The problem was solved by changing the bearing material. The most serious operating problem with the heavy-water pumps was with the shaft seals. The manufacturers had always made water pumps with designed leakage through the seals to ensure lubrication, with a recovery system. With heavy water, a design that allowed for the full recovery of such leakage was needed. Shaft seal development became a continuing program for the heavy-water power reactors.

Early in the operation of NRU, the formation of gibbsite[20] (see chapter fourteen) in the heavy water caused blockages of sample lines. An evaporator was installed in the heavy-water system and the sludge that accumulated was periodically removed and sent for further evaporation treatment to recover the heavy water, with the final sludge being processed as waste.

Late in 1958 the absorber of a control rod became disconnected from the drive mechanism and remained leaning at an angle in the reactor core on startup. This led to the introduction of shroud tubes around control and adjuster rods. Subsequently, a shroud tube buckled, jamming the absorber, which led to a limit being placed on the number of shroud tubes permitted in the reactor around control and adjuster rods.

About 1960 it became obvious that the efficiency of the heavy-water heat exchangers was deteriorating. The river water used for cooling came directly to the shell side of the heat exchangers and the cause of the problem was found to be a very persistent form of algae that seemed to thrive in the high radiation fields of the heat-exchanger rooms. After some experimentation, it was found that the most effective way to remove the algae was with a laundry detergent.

In the fall of 1961 a broken vessel bolt was found in the NRU upper service space. Metallurgical examination indicated that all 144 bolts would eventually fail in the same way, caused by the type of lubricant initially used in installation. Inconel-X-stressed sections and a suitable lubricant were developed. All bolts were replaced, some at each shutdown, over an extended time period.

It was decided in early 1962 to convert NRU to operation with highly-enriched uranium fuel. The trigger for this decision,

[18] A technique, similar to sand blasting but using ground walnut shells in place of sand, was used. This technique removed the epoxy and cleaned the metal surfaces without damaging them.

[19] Employee's recollection: "According to an engineer from the manufacturer who took part in the design and manufacture of the pumps and helped with the modification, this was really a security problem. Everything they received was stamped secret and they were not encouraged to ask too many questions. They were well aware that stainless steel could not run satisfactorily on stainless steel under normal conditions, but they were asked to design stainless-steel pumps for a service for which they were not apprised. They just assumed that there was something different about this service in a nuclear facility so they produced the pumps – complete."

[20] Gibbsite is a hydrated aluminum oxide. A more serious problem occurs with gibbsite when there are extensive fuel failures. In this case the gibbsite adsorbs and transports large amounts of fission products, resulting in very high fields around all heavy-water equipment, making maintenance difficult.

the impending loss of the plutonium market in the United States, was the same as that for the conversion of NRX to enriched fuel. However, for NRU, a different ground rule was adopted in arriving at a preferred fueling scheme. This was to maintain the thermal-neutron-flux level while minimizing the fueling cost. In general, with highly-enriched uranium fueling (compared to natural-uranium fueling), the thermal-neutron flux level could be maintained at a lower reactor power and with the use of fewer fueled lattice sites (freeing more sites for experimental and production use). On the other hand, the fast-neutron flux levels were, in general, considerably lower. The scheme chosen used uranium-aluminum-alloy fuel rods (initially with a uranium-235 content of 0.5 g/cm over the nine-foot (2,743 millimetre) active length), with the uranium enriched to 93 percent uranium-235 in uranium. A fuel-management scheme was adopted in which fresh fuel rods were loaded into the outer regions of the core and moved progressively (normally twice) towards the centre of the core before being removed. Fast-flux levels were maintained where they were required by experimentalists, using natural-uranium, fast-flux-peaking rods. This scheme satisfied the ground rule, and provided flexibility for increased reactor loads (with associated increases in power and fueling costs) and higher thermal flux, if desired.

The conversion was carried out from November 1963 to March 1964, with NRU shut down (contrary to the procedure with NRX). Immediately after conversion, NRU was operated at 60 MW, compared to its pre-conversion operation at 220 MW (both figures being exclusive of loop powers), with the same usage. It soon became apparent that user demand justified greater capability. The power and uranium content of the enriched fuel were both raised over time to about 130 MW and 1.5 g(uranium-235)/cm, respectively (with roughly the same neutron flux level).

The conversion was extremely successful. The expanded capability and relative cost-effectiveness greatly extended the useful life of NRU.

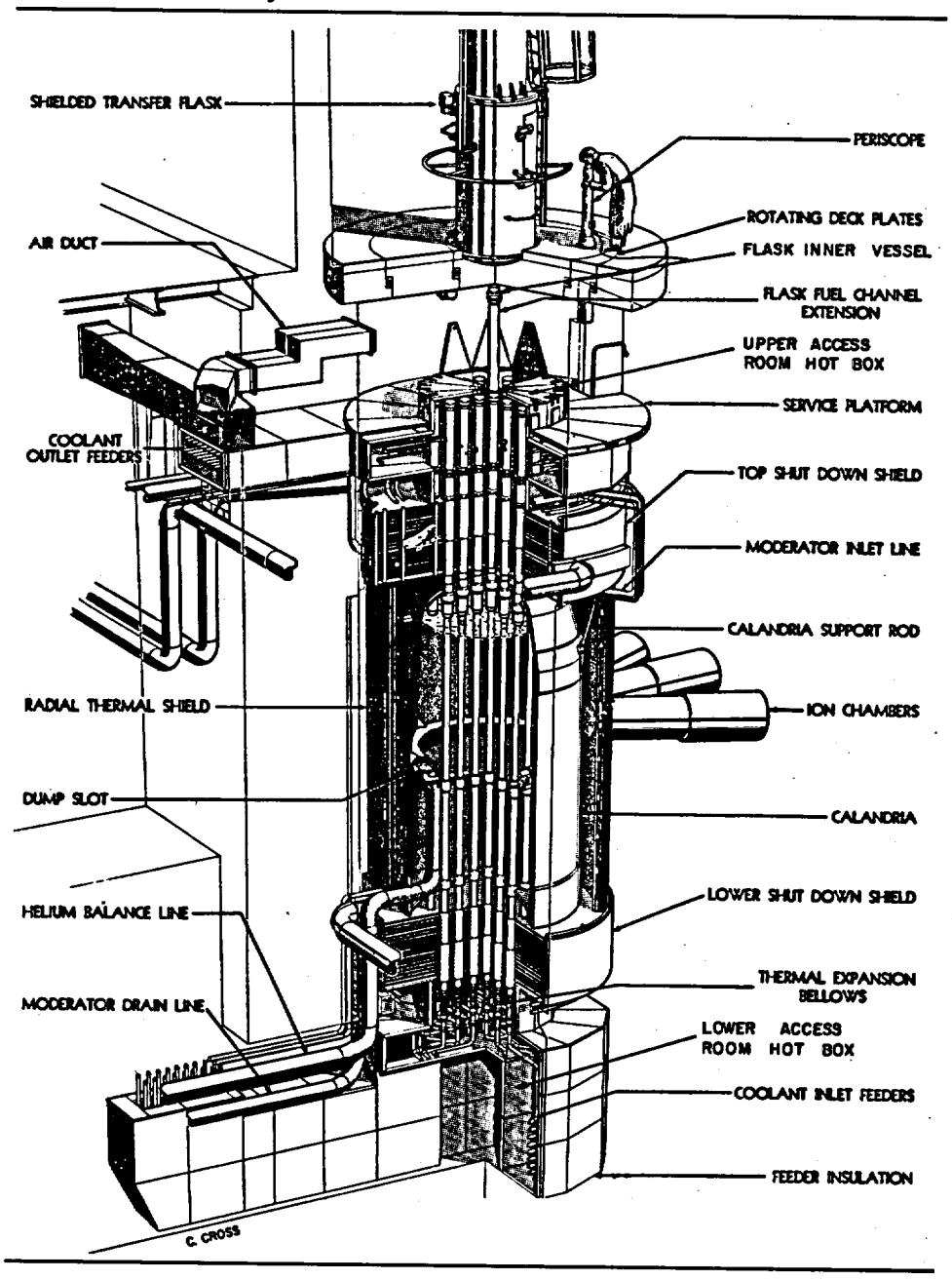
Around 1970, a hole developed in one of the expansion bellows of the NRU reactor-vessel side-wall. A mechanical plug and washer device was designed and installed to successfully seal the hole. Over the next several years, more holes developed, with gradually increasing frequency. The sealing of each hole required several weeks of reactor shutdown, hastening the decision to replace the reactor vessel. A new vessel had been ordered in 1958 and was delivered to the site in 1966. In the years before its installation, a great deal of work was done on the vessel at Chalk River. Over this same period, a project team had been making preparations that would facilitate the replacement when it was required. Of particular significance was their development of a very complex, large, remotely operated tool for inspecting, cleaning and repairing the stainless-steel surfaces in the reactor, to which the aluminum side wall must seal to contain the heavy water. NRU was shut down for vessel replacement in June 1972 and restarted in August 1974. Operation then continued with a very high usage and production for many years.

WR-1 Highlights

AECL interest in the concept of an organic-cooled heavy-water-moderated power reactor (CANDU-OCR) was initiated by a suggestion from engineers at Canadian General Electric, who were following the American program on organic reactors. AECL commenced a development program for this new concept, to provide a backup alternative to its main candidate – the pressurized, heavy-water-cooled, heavy-water-moderated power reactor. Building a small reactor to test the concept was deemed the best way to focus the development effort; such a reactor could also serve a valuable role as a materials-and-fuel-test reactor. In 1960, a proposal was approved to build such a reactor (to be known as WR-1) as the main initial facility at a new research establishment in Manitoba: the Whiteshell Laboratories. Construction of WR-1 began in 1963 and criticality was attained on 1 November 1965.

WR-1 differed in several fundamental ways from NRX and NRU, besides having organic as the coolant. The reactor coolant could operate at high temperature (up to 425°C at the outlet), although still at relatively low pressure. No neutron beam tubes were provided, so there was no external access to the neutrons produced, and hence no basic-physics program. Moderator

FIGURE 2.16
WR-1 Reactor Cutaway



level, moderator dump and addition of soluble neutron absorber (boron) to the moderator were relied on for all power control and shutdown functions; there were no control or shut-off rods. Provision was made in the design for four loops: three organic-cooled and one water-cooled.

Figure 2.16 shows a cutaway view of WR-1. The reactor is of a vertical pressure-tube design, with provision for fifty-five fuel sites divided among three coolant circuits, and having a nominal thermal power rating of 60 MW. However, the initial configuration did not use this full capability.

In the original commissioning there were thirty-seven fuel-rod sites, each with an aluminum calandria tube, and the nominal power rating was 40 MW (thermal). Fuel channels, the in-core portion of which were pressure tubes (I.D.= 8.29 cm), were installed in each of these fuel sites, to support fuel assemblies and to contain and direct the flow of coolant (inlet pressure = 2.17 MPa) over the fuel. Stainless-steel pressure tubes were installed in thirty-one sites, and zirconium-alloy pressure tubes in the other six. The primary coolant was a partially hydrogenated mixture of terphenyls (Monsanto's tradename: HB-40) that is fluid at room temperature; the coolant is discussed further in chapter sixteen.[21] Two independent primary cooling circuits served nineteen and eighteen fuel-rod sites, respectively. The fuel was 2.4 wt. percent (uranium-235 in uranium) uranium dioxide clad in zirconium-2.5 percent niobium alloy (Zr-2.5Nb).[22] Each fuel string comprised five eighteen-pencil bundles on a central shaft that formed an eight-foot-long fuel assembly.

WR-1 commissioning proceeded smoothly, with no serious difficulties and only one major delay.[23] As might be expected with a new reactor design, some modifications were required in the pre-critical phase to make the various systems function as intended. For example:

- to prevent gas-locking of the moderator pumps during a moderator dump, it was necessary to extend the discharge pipe across the dump tank and install baffles to keep the entrained helium from the pump inlet, and
- it was necessary to re-route the ion-chamber cables to eliminate false signals due to "cross-talk" from the crane control circuitry.

The first approach to critical was carried out with fuel in the inner nineteen sites only, to obtain physics measurements with the moderator near its equilibrium level. The startup was smooth and uneventful; the low-power commissioning was carried out throughout November 1965 with WR-1 operating almost continuously at 0.01 percent of full power. This allowed for measurements of nuclear coefficients (change in reactivity with changes in moderator temperature, power, coolant temperature, and coolant void; change in reactivity due to insertion of driver fuel in the central site; reactivity change in the first second of a moderator dump), neutron fluxes, coolant dosimetry, and regulating-system performance. After these measurements and tests were completed for the nineteen-site core, six additional sites were fueled for the high-power core. The major physics measurements and tests were repeated for the twenty-five-site core. Satisfactory agreement was obtained between design values and measurements, except that the output from the ion chambers was found to be too high relative to the system design, due to a significantly lower flux gradient in the ion chamber cavities than predicted. The ion chambers were withdrawn sufficiently to permit full-power operation, some six weeks after first critical.

The main problems encountered during high-power commissioning of WR-1 were difficulties in obtaining satisfactory response from the automatic temperature control system, and in obtaining satisfactory performance from the thermal power control system. The use of standard modules in a relatively complex control circuit proved to be unsatisfactory, and many were replaced by higher quality versions.

Early in the operation of WR-1, a small defect occurred in a fuel element. The failed-fuel-detection system performed as designed, and the fuel element was located and removed without difficulty.

Over the next four years WR-1 operated well, with an average availability factor of 85 percent. Much of that operation was at a coolant outlet temperature of about 370°C, and long, uninterrupted periods of operation were obtained. The operators

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[21] Employee's recollection: "HB-40 was originally intended to be used only as a startup coolant in WR-1, to be replaced later by a product composed of unsaturated terphenyls. HB-40 had the advantage of being liquid at room temperature, but it was thought that its rate of decomposition would be too high to allow it to be used as the permanent coolant. However, as WR-1 operated it was found that the rate of decomposition in-reactor was much lower than predicted and the coolant type was never changed. The HB-40 did indeed decompose, but many of the decomposition products recombined to form other compounds that behaved equally well as reactor coolant."

[22] Employee's recollection: "CGE's reference fuel cladding material for WR-1 was sintered aluminum product (SAP - Al + 10 percent Al_2O_3). However, every test of SAP-clad fuel in the X-7 loop in NRX failed because of the low ductility of SAP. Within about twelve months of the start of WR-1 there was no satisfactory cladding and CGE were about to revert to stainless-steel cladding (which, if used in an organic-cooled power reactor, would make the use of natural-uranium fuel impossible). Some zirconium coupons had been tested in organic coolant and the results were promising. Trefoils of zirconium-alloy-clad fuel were made, to the Douglas Point design, for testing in the X-7 loop. After a three-month test it was found that the fuel cladding remained structurally sound (all the hydrogen that had entered the cladding had migrated to the cooler wire wrap that was part of the design). On the basis of this one test, Zr-2.5Nb fuel cladding was chosen, and there was no cause to regret this decision over the life of the reactor."

[23] This delay was associated with difficulties in aligning the main coolant pumps and motors. It was found that the base and springs on which these were mounted were designed for an 8,000 lb. motor, whereas the weight of the motor had been revised to 12,000 lb. New springs had to be manufactured and the commissioning of the heat-transport system was delayed for twenty-eight days.

concluded that the commissioning and operation of an organic-cooled reactor was more trouble-free and straightforward than that of a pressurized-water-cooled system. They attributed this to two characteristics of the organic system: high coolant-outlet temperature is attainable with relatively low operating pressure in the primary system, and the radiation fields near the primary piping, feeders and headers are very low, minimizing the problems of access control for normal operation and maintenance.[24]

One potential problem with the organic coolant is the fire hazard it presents. Organic coolants are flammable, and appropriate precautions must be taken in handling them. Experience with WR-1 showed that use of organic coolants is feasible. In this four-year period there were a few small fires, confined to glowing insulation, and two significant spills of hot organic coolant (at 350°C), with no attendant fire. The keys to prevention of serious fires in organic coolant reactors are:

- Good housekeeping – the thermal insulation on pipes must be carefully installed so that air access is minimized; if any insulation does become soaked with organic coolant, it must be replaced immediately.
- Elimination of normal ignition sources by use of explosion-proof fittings and thorough grounding of all piping.
- Provision of smoke detectors and protection of areas containing organic circuits with fog and water sprays.

Over this period it was also demonstrated that fouling of heat-transfer surfaces and hydrogen uptake by zirconium alloys in the primary system can be controlled by coolant chemistry. For the former, the coolant must be free of chlorine and low in metal oxides, and this is aided by maintaining a small amount of water in the coolant and by side-stream circulation through clay columns. For the latter, maintenance of an oxide film on the zirconium provides an excellent barrier to hydrogen migration into the material, and is achieved by maintaining a small amount of water in the coolant and low chloride contamination.[25]

With WR-1 operating satisfactorily, there was considerable incentive to make improvements that would demonstrate the feasibility of a CANDU-OCR, provide better conditions for experimenters and lower the fueling costs. Towards this goal a number of changes were made, so that by 1975:

- the stainless-steel pressure tubes (relatively high absorbers of neutrons) were all replaced by zirconium-alloy tubes,
- a third organic primary coolant circuit was added to service seventeen of the remaining sites, which were then commissioned as fuel sites, and
- the fuel material was changed from uranium dioxide to uranium carbide at a suitable enrichment.

At the same time, a great deal of effort went into development of inserts for the loops and the fuel sites, to best exploit the R&D capabilities of the reactor.

WR-1 was shut down and decommissioned in the mid eighties due to budget pressures.

Research Reactor Fueling

The requirements for research-reactor fuel are generally quite different than those for nuclear-power-reactor fuel. In power reactors, the primary objective is the generation of thermal energy at a high temperature (to get efficient conversion to electrical energy), which means, for water-cooled power reactors, that the fuel must operate in a high-temperature, high-pressure coolant. On the other hand, research reactors, such as NRX and NRU, are built to provide neutrons and a neutron-rich environment,

[24] Employee's recollection: "One of the very pleasant surprises about the operation of WR-1 was the absence of any activity transport. In all of the water reactors, and even in water loops, corrosion products are transported by the coolant through the reactor where they become radioactive. They then deposit in the out-reactor circuits, making maintenance difficult. The operation of WR-1 showed that the organic did not corrode the circuit and, even after twenty years of operation, one could do maintenance work on any part of the out-reactor circuit in very small radiation fields."

[25] Employee's recollection: "When it was decided to use zirconium alloy as fuel cladding, it was also decided to add a trace (100 ppm) of water to the coolant, to maintain an oxide layer on the cladding to reduce hydrogen ingress. It was found that the addition of water also substantially reduced coolant-decomposition-related fouling on the fuel and other hot surfaces." [Fouling was a problem that plagued the first American efforts to use organic liquids as reactor coolants.]

and operate most efficiently with low-temperature and -pressure coolant. The situation is somewhat different for WR-1, which was built partly to test first-hand the concept of an organic-cooled CANDU. Since, at temperatures suitable for electricity production, the pressure of organic coolant is very much less than that of water, it proved to be feasible to operate WR-1 fuel under conditions very similar to those suitable for a power reactor. Thus the experience gained from the WR-1 driver fuel is much more directly relevant to possible power-reactor fuels for a CANDU-OCR than experience with NRX and NRU driver fuel is to CANDU-PHW fuel.

In all cases, research-reactor fuel performance is important in providing reliable operation of an important R&D facility. The provision of research-reactor fuel can also be an important factor in building up the infrastructure for a nuclear fuel industry. However, there is a relatively small market for research reactor fuel, and so most of the development discussed here was done in-house.

NRX Fuel

For the first fifteen years of NRX operation, natural-uranium-metal rods were the standard fuel. These consisted of a solid uranium-metal core 1.36 inches (34.54 millimetres) in diameter and 10 feet (3,048 millimetres) long. The uranium was clad with a drawn aluminum sheath, which was either 0.08 or 0.04 inches (2 or 1 millimetres) thick. The clad uranium core was inserted into an outer aluminum coolant tube to provide (post-1952) a water-cooling annulus of 0.071 inches (1.8 millimetres).

Difficulties were experienced with the uranium-metal rods, mainly due to dimensional instability during irradiation: changes in length and diameter, transverse fractures, and longitudinal splits. These conditions caused restrictions in coolant flow and failures in the aluminum cladding, which required removal of rods before their scheduled burnup (approximately 30 MWh/kgU).

The uranium-metal cores were formed by hot rolling, starting with large uranium-metal billets and using multiple passes to achieve the final desired diameter. Work to determine the effects of fabrication procedures on uranium stability under irradiation continued over the years. Factors investigated included type of billet (vacuum-cast or forged), rolling temperature, diameter reduction per pass, heat treatment and finishing following rolling, and inspection methods. Although, during their whole period of use, approximately 23 percent of the rods had to be removed from the reactor before reaching their scheduled burnup, the later batches of rods had a much-improved performance due to the increased knowledge and experience acquired.

By 1961 it was decided to introduce new fuel designs into NRX, and 10-foot (3,048 millimetres) long, solid-pellet uranium-oxide rods[26] were designed and fabricated for irradiation. The sheaths were made from an aluminum alloy, to provide the greater strength required by the buildup of pressure during irradiation from the fission product gases released by the uranium dioxide. These rods were confined to the lower-neutron-flux regions of NRX, to limit the temperatures in the centre of the uranium-dioxide pellets. The irradiation performance of the solid uranium-dioxide rods in NRX was very satisfactory. No significant changes in rod length or diameter were experienced.

Annular pellet uranium-dioxide rods that were cooled internally and externally were developed for the hotter (higher-neutron-flux) positions in NRX. The pellet had an outside diameter of 35.8 millimetres (1.410 inches) and a bore of 15.24 millimetres (0.60 inches), and was approximately 22.2 millimetres (0.875 inches) long. Several rods were examined after irradiation and no significant changes in rod length or diameter were observed.

A seven-element booster assembly was developed for NRX to maintain the reactivity with increased loads. Enriched uranium (93 wt. percent uranium-235) metal pieces were dissolved in molten aluminum at 1,050°C in a graphite or ceramic crucible,

[26] Uranium-oxide pellets had been developed and commercially produced for the NPD demonstration power reactor. The uranium-oxide pellets for the NRX rods used similar fabrication procedures and specifications, but were larger. They were 1.41 inches (35.8 millimetres) diameter and 0.8 inches (20.3 millimetres) long.

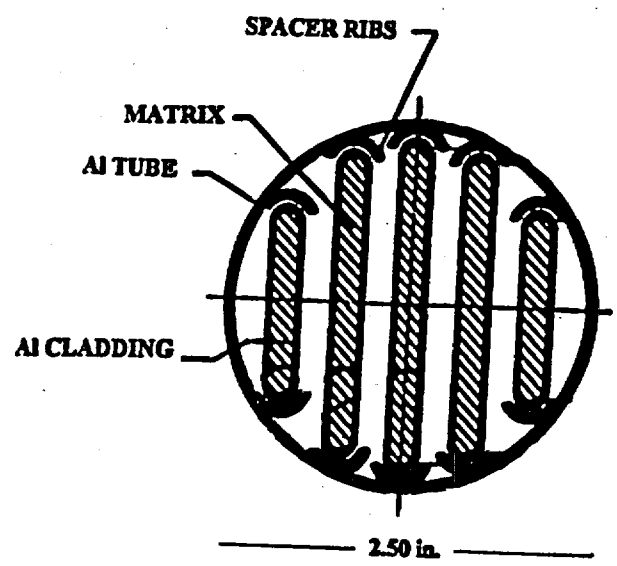
to yield a uranium-aluminum alloy. The melt was cast into water-cooled moulds to provide billets 38.1 millimetres (1.5 inches) diameter and 89.0 millimetres (3.5 inches) long. The billets were extruded at 540°C to 7.0 millimetres (0.275 inches) diameter. The extruded cores were drawn cold to 6.35 millimetres (0.250 inches) diameter in one pass of 17.5 percent reduction. The core was then scanned with two scintillation crystals and photo-multiplier tubes, to determine the dispersion of uranium-235 in the core. Aluminum end plugs were attached to the uranium-aluminum core and the core assembly was clad with 1.14 millimetres (0.045 inches) of aluminum by an extrusion process. An eddy current test was developed to detect voids and inclusions in the clad element. The aluminum-coolant tube that contained the seven elements had a bore of 35.46 millimetres (1.396 inches) and a wall of 2.29 millimetres (0.090 inches). The seven elements were located in the coolant tube by six spacers.

The standard fuel in NRX was subsequently converted to enriched uranium-aluminum fuel. The NRX booster assembly was modified, and although the dimensions of the cladding and fins were changed, the fabrication process for the fuel elements was essentially the same as the booster assembly. The coolant tube was modified so that no internal spacers were required. These enriched fuel assemblies performed very well in NRX.

NRU Fuel

When NRU started up in 1957, the initial fuel assemblies consisted of natural-uranium-metal flat bars clad in aluminum sheaths, with no bonding between the bar and the sheath. The uranium flat bars were fabricated from vacuum-melted ingots. The ingots were hot rolled to oval billets and the billets were then hot rolled to provide flat bars of finished thickness. The flat bars were roll straightened, and the width of each bar machined to the finished dimensions. The length of all bars was 10 feet (3,048 millimetres). The 0.04 inch (1.0 millimetres) thick aluminum sheaths were extruded close to finished size. These extruded sheaths contained spacing fins, which formed the coolant channels. The uranium flat bars with attached aluminum end plugs were inserted by hand into the sheaths.

FIGURE 2.17
Cross-section of Flat Type NRU Natural Uranium Rod



The clad flats were then drawn through a draw die, to sink the aluminum cladding onto the uranium flat bar. The ends of the sheath were welded to the aluminum end plugs, and the flat fuel elements were assembled into a round coolant tube.

The irradiation history of the non-bonded fuel assemblies in NRU was not good. Failures occurred at an unacceptable rate. In many cases the uranium metal shortened longitudinally under irradiation, causing transverse wrinkles in the aluminum sheath. These wrinkles significantly impeded the coolant flow. Other failures were traced to the poor heat transfer between the uranium fuel and the unbonded aluminum sheath. It was obvious that a fuel assembly of improved design was required.

Fortunately, the development of a bonded fuel assembly was initiated before the startup of NRU (see figure 2.17). The most successful development was the cladding of the flat uranium bars with aluminum by extrusion. The machined uranium bars were degreased, nickel plated, and vacuum dried. The extrusion cladding operation consisted of extruding a rectangular

ribbon of solid aluminum at 535°C, and then introducing the uranium bar, pre-heated to 350°C, into the flowing aluminum as a floating, self-propelled mandrel. The extrusion operation provided a cladding thickness of 2.3 millimetres (0.090 inches), which was subsequently machined down to 0.63 millimetres (0.025 inches). It took about ten minutes to machine one clad bar, and an eddy-current instrument was used to inspect the final cladding thickness. The flat fuel elements were assembled by hand into an extruded aluminum coolant tube, which contained spacing fins along its length to position them.

Over 12,000 nickel-plated and extrusion-clad uranium bars were irradiated in NRU, and no fuel failures were detected. However, the burnup achievable with this fuel was limited to about 43 MWh/kgU, and work started on development of an alternative design of natural-uranium-metal fuel (an assembly with nineteen round elements). This work was terminated when it was decided to fuel NRU with enriched fuel assemblies.

In 1962 it was decided to change NRU from operating with natural-uranium fuel to operating with enriched-uranium fuel. The objective was to have a complete loading of enriched-uranium-aluminum fuel assemblies by 1964. Two approaches were considered. The first was to purchase an existing co-extruded tubular design manufactured in the United States. The second was to develop a design that could be made in Canada without a major development program. The experience gained from the fabrication and irradiation of the enriched seven-element assembly in NRX indicated that a multi-element assembly for NRU could probably be developed without a major effort. Various design configurations were investigated, including the number of fuel elements, coolant flows, coolant temperatures, sheath surface temperatures, and allowable surface heat flux. The investigation showed that a twelve-element fuel assembly, nine feet (2,743 millimetres) long, of small diameter elements, would meet the design requirements.

The enriched-uranium-aluminum-alloy billets were fabricated by essentially the same process used for the NRX fuel. The alloy was 9.8 wt. percent uranium and the uranium was 93 wt. percent uranium-235. Similarly, the uranium-aluminum fuel cores were clad with 0.03 inch (0.76 millimetres) thick aluminum by the extrusion cladding process. The extruded aluminum-alloy coolant tube was circular in shape. Six spacers were required to locate the fuel elements along the length of the assembly.

Post-irradiation examination of several twelve-element assemblies showed no significant corrosion, erosion, or fretting wear at the design coolant flows and reactor residence times. The twelve-element assembly became the production design for the enriched-uranium-aluminum fuel for NRU.

WR-1 Fuel

Fuel development work for both WR-1 and CANDU-OCR concepts was carried out in the X-7 and U-3 loops at CRL. However the operation of both standard and experimental fuel in WR-1 itself contributed greatly to the inventory of knowledge regarding fuel performance with organic coolants.

As previously mentioned, it was possible to operate WR-1 with coolant conditions similar to those suitable for a power reactor. Of the three separate organic coolant circuits in WR-1, the 'A' and 'B' circuits were generally operated at a coolant inlet temperature of 290°C. However, the 'A' circuit was operated at an inlet temperature of 375°C (outlet temperature about 400°C) from 1971 to 1977. The 'C' circuit inlet temperature was 320°C.

The initial driver fuel used in WR-1 was of an eighteen-element uranium-dioxide design similar to the Douglas Point nineteen-element bundle, except that the centre element was omitted so that the bundle would fit over a central support shaft. The uranium was enriched in the range 1.2 to 2.4 wt. percent uranium-235 in total uranium. Two fuel types of this general design were made: one used Zr-2.5Nb-sheathed fuel elements assembled into 19.5 inch (495 millimetres) long bundles, with each reactor fuel string having five bundles suspended on the central hanger tube; the other used sintered aluminum product (SAP), sheathed fuel elements assembled into 32.1 inch (813 millimetres) long bundles, with each reactor-fuel string having three bundles suspended on the central hanger tube. About half of a full reactor charge of the SAP-clad fuel had been

made by the time WR-1 started up, and this was eventually irradiated, but no more was made.

WR-1 operated with the uranium-dioxide driver fuel from startup in 1965 until 1973, when uranium dioxide was gradually phased out in favour of uranium- carbide fuel. Experience with the uranium-dioxide fuel in WR-1 was not good compared to the performance of uranium dioxide in water-cooled reactors[27] and to the later experience in WR-1 with uranium-carbide fuel performance. The overall average string burnup for all the uranium-dioxide fuel irradiated in WR-1 (about 1,100 bundles) was 128 MWh/kgU, which satisfied the original target of 120 MWh/kgU, but little more. The number of fuel strings removed due to failure or sticking exceeded those retired without incident.

Experimental irradiations of uranium-carbide fuel started in 1966, and in 1973 the irradiation of uranium carbide as the reactor-driver fuel, in quantities sufficient to obtain statistical information, commenced. Conversion of WR-1 from uranium-dioxide to uranium-carbide driver fuel was essentially completed by the end of 1977.

The driver-fuel bundles for WR-1 consisted of one ring of 14 Zr-2.5Nb-clad, uranium-carbide fuel elements, and five of these were assembled on a hollow central hanger to form a fuel string. The uranium was enriched in the range 1.3 to 2.25 wt. percent uranium-235 in total uranium.

The performance of the uranium-carbide fuel was excellent both from the viewpoint of burnup achieved and failure rate. Average burnup of the first 125 bundles retired was 253 MWh/kgU, whereupon the target burnup was raised from 240 MWh/kgU to 360 MWh/kgU. Not only was the failure frequency low, but the consequences of failure were not serious. It was found that while activity releases were high enough to detect failure, they remained low for a long enough period (six weeks or more) to continue operation of the failed fuel until the next scheduled shutdown before removing it. A significant problem was encountered in the first few strings of uranium-carbide fuel operated in the 'A' circuit during the period of its high-temperature operation. Hydrogen migration to the bundle end plates caused their embrittlement, and six bundles had to be retired due to end-plate breakage during shuffling operations. The problem was solved in the short term by changing the fuel management scheme. A longer-term solution, involving larger hydrogen-sink volumes in the end-plate region, was developed.

Research Reactor Loops

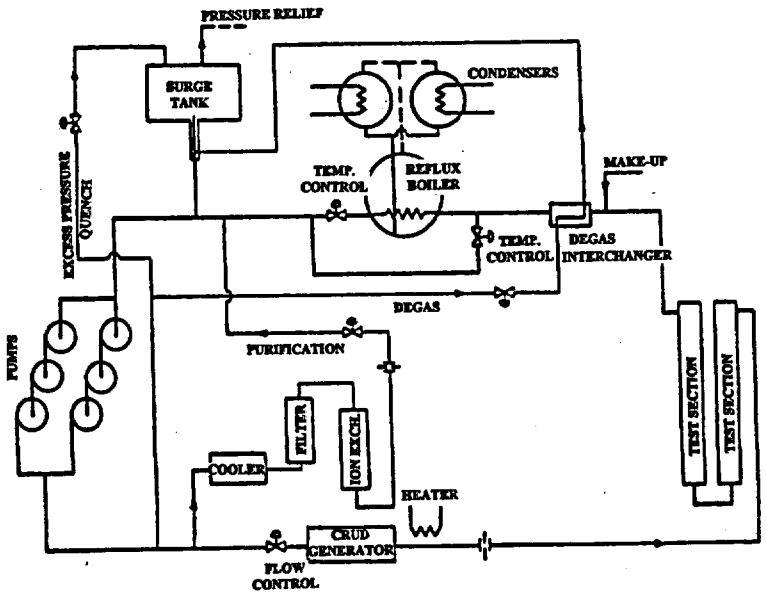
An in-reactor loop is a closed-circuit hydraulic system that provides a coolant and radiation environment similar to that in a nuclear power reactor. It consists of pumps to circulate the coolant, heaters to heat the coolant, coolers to remove the excess heat, pressurizer to pressure the coolant, test sections for the test assembly, piping to interconnect the components, and the electrical and process control system to power and control the process. Figure 2.18 shows an example of a loop circuit.

The concept of high-pressure, high-temperature, light-water loops with an in-reactor test section for fuel development, water chemistry, materials testing, etc., was introduced to CRL in 1950 by the United States Atomic Energy Commission and the Westinghouse Atomic Power Division (WAPD). The first high-pressure, high-temperature water loop started operating in the central thimble of NRX in 1951 and was removed in 1952. This was followed by the installation of the WAPD CR-IV and WAPD CR-V loops starting in 1954.

Many loops were installed in AECL research reactors over the years, and these were often modified over the course of their use. They were an extremely important resource for the Canadian power-reactor development program. Table 2.2 lists

[27] Two factors make the behaviour of organic-cooled uranium-dioxide fuel elements significantly different from that of water-cooled elements: the relatively low system pressure and high-sheath surface temperature in the organic-cooled case. These conditions allow the fuel to swell more readily and, at the same time, the higher fuel temperature causes a greater fission-gas release from the fuel, which tends to reduce the fuel-to-sheath heat-transfer coefficient. The situation is unstable and eventually causes failure at a hot spot where diametral growth, bowing or surface deposits have restricted coolant flow.

FIGURE 2.18
U-2 Loop Simplified Flowsheet



key characteristics of the main AECL loops, circa 1980, to indicate the scope of the loop facilities that were available for use in power-reactor development. Table 2.3 lists the uses made of the loops.

Fuel testing in NRU loops could be done with full-scale power reactor fuel assemblies, whereas tests in NRX were limited to single- or few-element fuel. The following points will supplement the basic data listed in table 2.2:

- X-1 was one of the WAPD CR-IV loops installed between 1954 and 1957. A major modification was made in 1964, when both the main flow and the heat rejection capacity were increased. Boiling was limited to low outlet steam qualities.
- X-2, although originally a WAPD CR-IV loop, was completely redesigned and rebuilt by 1977 as an extended-fuel-defect-and-safety-related studies loop. The modifications included:
  - filters at the outlet to trap fuel particles released from failed fuels, special valves for loss-of-coolant tests,
  - extensive on-line gamma-spectrometry equipment to monitor sections of coolant piping for radioactivity, and
  - an improved ventilation system to minimize radioactive releases to the environment.
- X-3, another original WAPD CR-IV loop, had its flow and heat-rejection capacity uprated in 1964, and later modifications were made to permit operation with two parallel in-reactor test sections. The loop was largely dedicated to the Westinghouse fuel program.
- X-4 was originally designed and built by the English Electric Company Ltd. for the United Kingdom Atomic Energy Authority as a pressurized water loop, and installed in 1957. Modifications were made in the period 1962 to 1966, which extended the operating range and permitted its use as a multi-mode water loop (as table 2.2 indicates).
- X-5, the largest loop in the NRX reactor, was designed and fabricated for WAPD as the WAPD CR-V loop, and installed in 1955. Until about 1979, it incorporated a test section of a 62-millimetres internal diameter. It was found that the smaller-diameter test section listed in table 2.2 was better suited to later programs.
- X-6, originally similar to the other WAPD CR-IV loops, was modified and uprated as a boiling-water loop in 1963.
- X-7 was operated in NRX until the organic-cooled-reactor program was terminated in 1972, at which time it was decommissioned. It was left as intact as possible to facilitate recommissioning if required. X-7 was used for tests associated with WR-1 development.
- U-1, as originally installed in 1964, operated in four coolant modes: pressurized water, boiling water, steam-water inlet, and steam inlet. In 1974 the loop was uprated to operate with two in-reactor test sections in series, and this modification limited operation to the pressurized and boiling-water modes.
- U-2 was installed in 1959 as a pressurized water loop, but subsequent tests showed that limited boiling to about 10 percent outlet steam quality was possible. In 1974 the loop was uprated to operate with two in-reactor test sections in series.

TABLE 2.2
General data for AECL research reactor loops – circa 1980

REACTOR	LOOP	COOLANT & MODE *	DESIGN			TEST SECTION I.D. (mm)	EFFECTIVE LENGTH (mm)	THERMAL OR FAST FLUX (10^{14} n,cm^{-2}.s^{-1})
			PRESS (MPa)	TEMP. (°C)	HEAT (kW)			
NRX	X-1	PW,B	17.3	330	300	38	3030	0.8 (thermal)
	X-2	PW	13.9	336	200	38	3030	0.8 (thermal)
	X-3	PW,B	17.3	330	600	38	3030	0.8 (thermal)
	X-4	PW,B,TPI,S	15.3	335	250	38	3030	0.8 (thermal)
	X-5	PW	17.3	330	550	38	3030	0.8 (thermal)
	X-6	PW,B	18.6	359	300	38	3030	0.8 (thermal)
	X-7	organic	2.9	430	220	38	– **	–
NRU	U-1	PW,B	13.9	354	12000	103.9	3640	3.0 (thermal)
	U-2	PW,B	13.9	354	8000	103.9	3640	3.0 (thermal)
	U-3	organic	4.2	430	4500	103.9	– **	–
	U-5	PW	17.3	354	140	63.25	3640	0.2 (fast)
WR-1	1L2	PW	9.0	330	400	47	2250	1.2 (thermal)
	1L4	organic	3.8	427	4500	76 ***	2250	1.0 (fast)
	1L5	organic	3.8	427	4500	76 ***	2250	1.0 (fast)
	1L6	organic	8.6	427	4500	76 ***	2250	1.0 (fast)

* PW pressurized water; B boiling; TPI two-phase at inlet; S steam

** decommissioned

*** This is the diameter of the support tube for the standard fast neutron fuel assembly insert normally used. Without the insert the diameter available is 104 mm.

- U-3 was operated in NRU until being decommissioned in 1972. It was left as intact as possible to facilitate recommissioning if required. U-3 was used for tests associated with organic-cooled reactor development.
- U-5 was designed for materials testing, not fuel testing. Light water at the required chemistry, pressure and temperature is supplied to as many as six in-reactor test sections operating in parallel, and normally installed inside hollow fast-neutron rods.
- 1L2 was normally operated in the pressurized mode, although boiling was allowed in the fuel channel with outlet steam qualities up to 15 percent.
- 1L4,5,6 were essentially identical, except for a difference in some of the piping in 1L6, which allowed it to operate at a higher pressure than the other two. A fast-neutron fuel assembly was normally inserted. The loops were used primarily for materials-irradiation experiments at high temperature in a high fast-neutron flux.

Loop experiments impose demanding data-gathering requirements that need flexible and highly reliable systems. Computer systems were installed at NRX and NRU for data acquisition in the early sixties, but by the seventies they were becoming increasingly unsatisfactory. In the mid-seventies there was a growing interest in the development of computer networks for data processing and control (i.e., distributed systems). It was felt that CANDUs might benefit from such systems, as replacements for the dual central computers, through reduced cabling costs. To evaluate this new technology, it was decided

to develop a distributed data acquisition system, to be known as REDNET, for the NRX and NRU loops, to replace the old system.

REDNET development proceeded slowly. The complexity of distributed system concepts had been underestimated, and the project had started at a time of dwindling funding for AECL Research, and of changing priorities (see chapter twenty). By the time it was completed, about 1985, it represented the largest software project undertaken at CRL (sixty-five person-years) and it was required only in NRU. A local network with five processors gathers, processes and stores the data from more than one-thousand data points, which are scanned twenty times per second. Terminals in NRU and other buildings allow users to monitor data, to request displays, and to modify data-processing rates or display modes. Archival data is transferred to the computing centre for permanent storage. High reliability is achieved through sharing of tasks amongst processors, and automatic adjustment of this sharing under failure conditions. The system is flexible and capable of considerable expansion; it should be readily adaptable to new hardware and software.

Isotope Production

Chapter five describes the production and marketing history of radioactive isotopes in Canada. The following will give a brief account of the CRL contribution to that history.

TABLE 2.3
Uses made of loops

USES	LOOP														
	NRX							NRU				WR-1			
	X-1	X-2	X-3	X-4	X-5	X-6	X-7	U-1	U-2	U-3	U-5	1L2	1L4	1L5	1L6
Fuel development	✓				✓	✓		✓	✓		✓				
Pressure tube development	✓			✓		✓		✓	✓						
Activity transport	✓			✓	✓	✓		✓							
Water chemistry	✓			✓	✓	✓		✓	✓						
Irradiation of metallurgical samples	✓			✓	✓										
Materials testing											✓		✓	✓	✓
Corrosion - in reactor	✓			✓	✓	✓		✓	✓						
Corrosion - out reactor				✓	✓	✓		✓	✓						
Fuel defect studies		✓													
Heat transfer						✓									
Critical heat flux				✓				✓	✓						
Fuel moving				✓											
Fuel power ramping								✓	✓						
Fission product release		✓				✓		✓							
Fuel power cycling								✓	✓						
Fuel irradiation in: pressurized water				✓				✓							
Fuel irradiation in: boiling water				✓				✓							
Fuel irradiation in: steam/water				✓				✓							
Fuel irradiation in: steam				✓				✓							
Minor fuel sheath defect tests	✓				✓										
Instrument development				✓											
Organic-cooled reactor development							✓			✓					
Westinghouse fuel program			✓												

Driven by demand, production of radioisotopes started in NRX as soon as the reactor was operational. An isotope production branch (IPB) established within operations initially used existing laboratory space. At the same time, planning started for construction of new facilities for the production and separation of isotopes and their distribution to universities and others.

By September 1947, millicurie quantities of phosphorus-32, iodine-131 and sulfur-35 had been extracted from seventy-five samples irradiated in the central thimble of NRX, and two external requests were in the process of being supplied. The first actual radioactive isotope shipment (of cerium-144) was made to the University of Saskatchewan on 31 October 1947 (three months after NRX startup).

By 1949, routine shipments were being made of iodine-131 to pharmaceutical companies for medical treatments, and of phosphorus-32 and sulfur-35 to university laboratories across Canada. Many service irradiations were done by request. Separation processes were being developed[28] to extract tritium from irradiated lithium fluoride and carbon-14 from irradiated potassium nitrate. Cobalt was being irradiated to produce cobalt-60.

During this period, isotope production in NRX was facilitated by changes in the reactor and by new techniques. Originally, isotope production in NRX was restricted to the central thimble (high flux, but serviced only during reactor shutdown) and the horizontal self-serve units (low flux, but serviced during reactor operation). Additional space for high-flux, in-core irradiation became available when it was found that three of the four control rods were not required. This led to the development of tray rods to position isotope targets in reactor fuel sites, which, after modification of the fuel-rod flask, permitted their transfer in and out of the reactor during a short shutdown.

In 1950 the IPB, with the same small professional staff but more process operators, moved into a new, large building devoted solely to isotope production, and the isotope program was immediately expanded. A thermal diffusion column was built for the tritium production program, with the final product being absorbed by uranium turnings in special containers. Carbon-14 was separated. Mixed fission products were transferred from the plutonium separation plant and were selectively separated using ion exchange methods (e.g., strontium-90, cerium-144 and cesium-137).

The value of cobalt-60 for both radiography and radiotherapy was recognized very early. In the fall of 1949, one-curie sources for radiography were produced for Dominion Bridge Company and Toronto Iron Works. The first irradiation of cobalt to produce cobalt-60 for beam therapy units (which was to become the dominant use of this isotope) started in the fall of 1949, and shipments of two 1,000-curie sources were made in 1951.

Around 1950 it was agreed that the commercial products division (CPD) of Eldorado would take on responsibility for the sale of NRX-produced cobalt-60, and in 1951 this agreement was extended to include most NRX-produced isotopes. When CPD was transferred to AECL in the fall of 1952, CPD took on responsibility for marketing all AECL-produced radioisotopes. In the period 1952-55 there was a transfer of some IPB staff to CPD in Ottawa, and some CPD staff to CRL, to work on processes. The IPB at CRL was disbanded about 1955, when the operations for isotope separation were transferred to Tunney's Pasture in Ottawa under CPD.

The reactors at Chalk River remained the major source of bulk radioisotopes for CPD,[29] henceforth referred to as RCC (radiochemical company), throughout the period to 1985 and beyond. The CRL operations group continued to play an important role in the success of RCC, not only in the supply of

[28] The aim was to develop methods for the production of "carrier-free" isotopes; i.e., free of isotopes of the natural element. That is why potassium nitrate, containing no carbon, was used to make carbon-14 initially. For phosphorus-32 the target was a compound of sulphur. Any amount of phosphorus-32 could be produced by the (n, gamma) reaction with natural phosphorus, but it would be much diluted with natural phosphorus and hence less valuable as a tracer in agriculture or medicine.

[29] Reactor control rods (those used for xenon-poison override, whose function required neutron absorptions over long periods at power) for the production of high-specific-activity cobalt-60 were put to efficient use. Dr. Lewis was fond of pointing out to visitors how this function of the control system was performed by the production of an isotope subsequently used for medical purposes.

radioisotopes, but in ensuring the reliability of this supply and in adapting to requirements for expansion of production, both in terms of volume and type of product. The advent of NRU with its capacity for large-scale isotope production was an important element in the ability to satisfy these demands.

The two radioisotopes constituting the bulk of RCC sales were cobalt-60 and molybdenum-99. In the eighties responsibility for separating molybdenum-99 and xenon-133 was moved to CRL under its operations group.

CHEMICAL OPERATIONS AND OTHER TECHNICAL SUPPORT

Reprocessing

In line with the original mission of the laboratories, a plutonium separation plant and a uranium-233 separation plant soon followed the NRX reactor as CRL facilities.

Initially, the plutonium-separation plant used a process developed earlier at the Montreal Laboratory. This involved dissolving the aluminum sheathing from the spent uranium-metal NRX fuel rods using caustic soda, dissolving the uranium in nitric acid, conditioning the dissolver solution with ammonium nitrate, extracting the plutonium with "trigly" (triethylene glycol dichloride), scrubbing the trigly extract with a solution of ammonium nitrate and nitric acid, and precipitating the plutonium as a phosphate. The precipitation stage of the process never worked properly and, shortly after starting up the plant, it was replaced by a process involving the use of hexone (methyl isobutyl ketone), and a final purification step using thenyltrifluoracetone (TTA) and concentration by evaporation. While the modified process operated well, with an overall recovery of 92 percent of the plutonium in a 95 percent pure product, the continued use of ammonium nitrate was undesirable, since it constituted a serious explosion hazard.[30] The process was further modified, using a scheme that replaced the ammonium nitrate with uranyl nitrate and the hexone with tributyl phosphate (TBP). This process operated reasonably well, but was shut down in 1954 when development work on an alternative process showed more promise.[31]

In this alternative process the plutonium was separated from the uranium and fission products by an anion-exchange resin. A pilot plant was built that provided two passes of the dissolver solution through ion-exchange resin, with intermediate conditioning between passes to reform the anionic complex. After some minor early problems, largely associated with the intermediate conditioning step, the process worked well. Although the decontamination factors approached 10^5, the final product still contained about 1.5 percent uranium, and enough radioactive niobium to make further extensive handling hazardous without shielding. Thus the TTA purification process was retained for final purification.

The uranium-233 separation plant also used a process based on work at the Montreal Laboratory. The feed for the plant came from the thorium rods that were irradiated in the J-rod annulus of NRX. The process developed at the Montreal Laboratory for uranium-233 separation involved dissolving the aluminum sheathing from the thorium-oxide rod with caustic soda, dissolving the thorium in nitric acid with some hydrofluoric acid added, extracting the uranium with diethyl dithiocarbonate in hexone and backwashing with 10 percent ammonium carbonate. This reduced the thorium/uranium ratio from about 9,000 to 30-40. Time had not permitted the chemists at the Montreal Laboratory to take the process further, so a method for further processing had to be developed at Chalk River. This involved a controlled thermal decomposition of ammonium carbonate in the backwash solution, followed by anion exchange. The overall uranium-233 yield in the process was about 95 percent.

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[30] An explosion of ammonium nitrate in an evaporator in December 1950 resulted in the death of a worker.

[31] In about 1950 a Chalk River scientist, while looking for a better method of analyzing solutions from the plutonium separation plant for plutonium, discovered that plutonium could be effectively separated from most other materials by absorbing it from a strong nitric-acid solution on anion-exchange resins, which were just then becoming commercially available. He reasoned that this could provide the basis of an effective separation process. This led to further laboratory investigation and the development work mentioned.

By 1956, arrangements had been made to sell the used fuel from NRX and NRU to the USAEC, and all reprocessing facilities at Chalk River were shut down. While they operated, they produced a total of about 17 kg of plutonium and about 500 g or less of uranium-233. The first 3 kg of plutonium was sold to the United Kingdom for use in their military program before they had product from their own facilities, and the remainder was used in the development and fabrication of booster fuel rods for NRX.

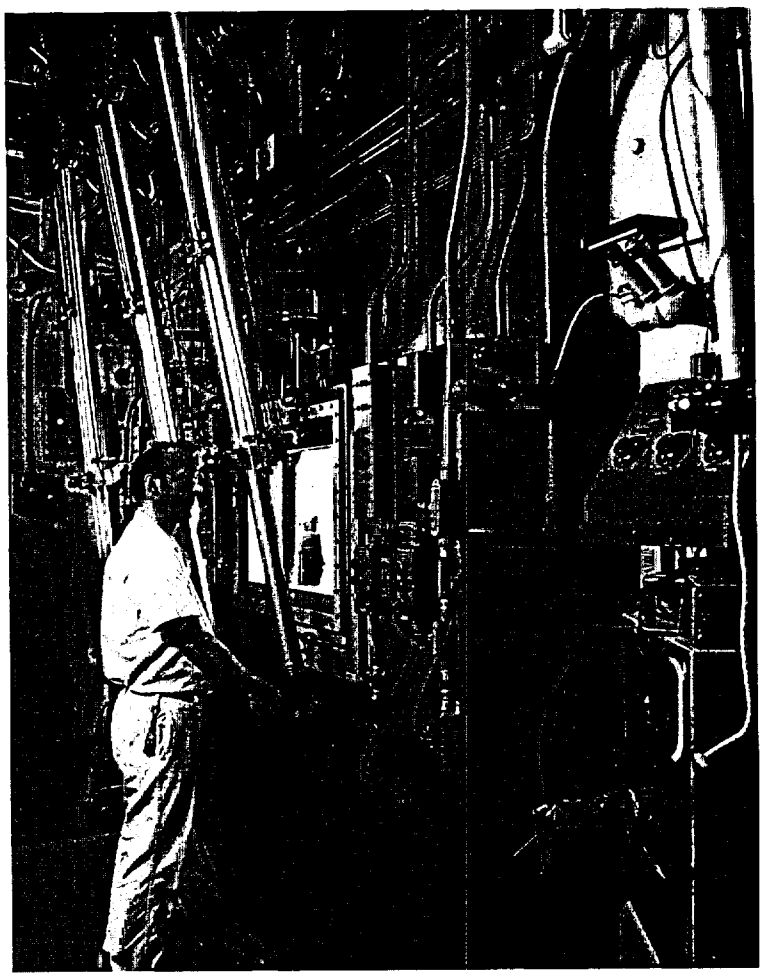
Hot Cells

Shielded facilities for the remote handling of highly radioactive materials, using manipulators, are commonly known as "hot cells". The first AECL Research hot cell was built in the metallurgy building at CRL, and was operated by the R&D arm of AECL Research. This hot cell, known as the "cave", was used for all early examinations of highly radioactive materials at CRL (apart from cursory examinations that could be carried out in the NRX fuel bays, where spent fuel was stored underwater). In the mid-fifties a second cave was built in the same location, to provide increased capacity and the ability to handle more highly radioactive materials.[32] Over time, metallurgical facilities were added adjacent to the metallurgy caves, to provide expanded examination and measurement capabilities. The whole facility was used primarily for R&D, and has always been operated by the R&D arm of AECL Research.

In the mid-fifties it was decided to build another hot-cell facility at CRL specifically for the handling of irradiated cobalt, the examination of irradiated fuel, the selection of metallurgical samples, and the repair and modification of irradiated or contaminated equipment. This facility was called the universal cells and was the responsibility of the operations arm of AECL Research.

The first section of the building that houses the cells was constructed in 1956 and two cells were installed, over the next year or two, in this section. Figure 2.19 shows a front view of the two cells. As well as the cells themselves, the building includes isolation rooms, a shipping and receiving area, change rooms and an office. These facilities soon became inadequate to meet the requirements of the expanding cobalt-60 production,

FIGURE 2.19
Front View of Cells 1 and 2



[32] These metallurgy cells were "dirty", in that the floors, tables, walls and ceilings were covered with radioactive debris arising from cutting open radioactive specimens. The facilities would operate in this condition for stretches of about five years, with installation of new equipment and repair and maintenance having to be done remotely. The cleanup at the end of the five years was a major undertaking, requiring involvement of personnel from throughout the plant.

and of the fuel-development program. An extension to the building was built in 1962 to accommodate a third cell, used chiefly for the examination of full-length fuel assemblies.

Equipment was installed in the universal cells for various purposes, including dissolving the aluminum sandwiches that contained nickel-plated cobalt pellets irradiated in NRU control (adjuster) rods before the pellets were shipped to RCC in Ottawa. Later, uranium-235 targets irradiated for molybdenum-99 production were dissolved in the cells for xenon-133 production before being shipped to Ottawa for processing. The cells also served as a backup to RCC facilities in Ottawa for molybdenum-99 processing.

The facilities were available for use by all CRL in-reactor loop users, including CRL experimenters and those from external laboratories. They allowed fuel tests from the in-reactor loops to be examined between irradiation periods, as well as after completion of their irradiation. Relevant equipment included various optical devices and photographic, machining, cutting and welding capabilities. In many cases preliminary inspection of tests was done in the universal cells, where smaller samples were selected and prepared for shipment to the metallurgical cells for more detailed examination.

At WL the functions of the universal cells (apart from the production functions, for which there was no need) and the metallurgical cells were combined in the hot-cell facility (HCF). The HCF was built to provide post-irradiation nondestructive and destructive examination for the various WL programs, fuel management and component handling for WR-1, and remote handling services and facilities, as required. It started operation in 1965 and consisted initially of eleven hot cells, to which a twelfth cell was added that began operation in 1976. Of special note was the dedicated use of four of the cells for an extended period for the thorium-fuel reprocessing experiment (see chapter eighteen).

Heavy-water upgrading

In the early fifties, a heavy-water upgrading plant was built adjacent to NRX, using electrolytic cells obtained from the plant run by Cominco at Trail, British Columbia. The operation was expanded in 1952, first using more cells obtained from Trail, as well as newer and better cells developed at CRL. A new plant was built and, for some time, ran at full capacity (4 MW of electrolysis or 1,000 Mg deuterium oxide/a at a feed concentration of 65 percent deuterium oxide), with a large backlog, upgrading heavy water primarily from NRX, NPD and Douglas Point. In the eighties, upgrading was done on a commercial basis for the utilities and the heavy water plants,[33] but the demand was not sufficient for full-capacity operation.

Site Waste Management

Chapter nineteen describes the history of AECL Research site waste management. The actual implementation of an effective waste-management strategy for the sites involved considerable skill and dedication from the operations groups. The activities for which they were responsible included:

- management of the waste-management sites, including construction and preparation of the relevant facilities at those sites for storing the wastes, collecting the site wastes and transporting them to the waste-management sites,
- emplacement of the wastes at the waste-management sites,
- security of the sites,
- maintaining appropriate records,
- operation of the waste treatment centre (WTC) at CRL, and
- operation of the underground research laboratory (URL) at WL.

Additional responsibilities

The CRL operations group was also responsible for operating a number of other facilities. For example, operation of the larger accelerators used for nuclear-physics research, mentioned in

[33] The production rate at Glace Bay and Port Hawkesbury was limited by the capacity of the water distillation final-enrichment step, and was increased by producing 94 percent heavy water, which was raised to reactor grade (99.8 percent) at CRL.

chapter eight, was the responsibility of the operations group until relatively recently, when the responsibility was transferred to the R&D group.

There were also a number of activities managed by the R&D arm of the organization, involving operation of facilities or provision of services, that were similar in some ways to activities under operations. Some examples are:

- the operation of the lattice test reactors, ZEEP and ZED-2, and the pool-test reactor, PTR,
- the operation of the Computing Centres (until 1986),
- analytical chemistry,
- electronics and instrument development,
- nondestructive testing, and
- glass blowing.

These were under the R&D arm, in general, for at least one of two basic reasons:

- the facility or service was used almost exclusively by one specialized group (examples being the operation of ZEEP, ZED-2, and PTR, and glassblowing) and/or
- the operation was of a highly specialized and technical nature, often with a significant R&D component (examples being the computing centre, analytical chemistry, electronics and instrument development, and non-destructive testing).

ACKNOWLEDGMENTS

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RADIATION PROTECTION AND NUCLEAR SAFETY

Chapter Three

Radiation Protection

A.M. MARKO
D.K. MYERS

GENERAL PRINCIPLES

Ionizing radiation in the form of X rays was first discovered in 1895, and natural radioactivity in 1896. Within the first decade of the discovery of X rays it had been established that X radiation was useful in medical diagnosis and in the treatment of disease; however, the hazards of skin damage, of bone marrow damage and of malignancy following high doses had also been noted. The need for protection against exposure to high doses of ionizing radiation was thus apparent.

Radiation protection was an ad hoc affair for many years and was directed primarily towards the prevention of reddening of the skin, since there was no acceptable method to measure small radiation doses. An ionization chamber developed in 1925 made it possible to measure small exposures accurately in terms of the amount of ionization they produced. An international committee on radiation units and measurement was founded in 1925 and this committee subsequently adopted units of radiation dose based on these physical measurements. A second international committee on radiological protection was founded in 1928. The latter was the forerunner of the International Commission on Radiological Protection (ICRP). Both committees operated under the auspices of the International Congress of Radiology (concerned originally with medical applications of X rays and radium) and both committees, although reorganized and renamed in 1950, remain to this day the most important sources of international recommendations concerned with radiation protection. Canadian scientists from a variety of organizations, but particularly from AECL, have participated for many years in the work of these committees.

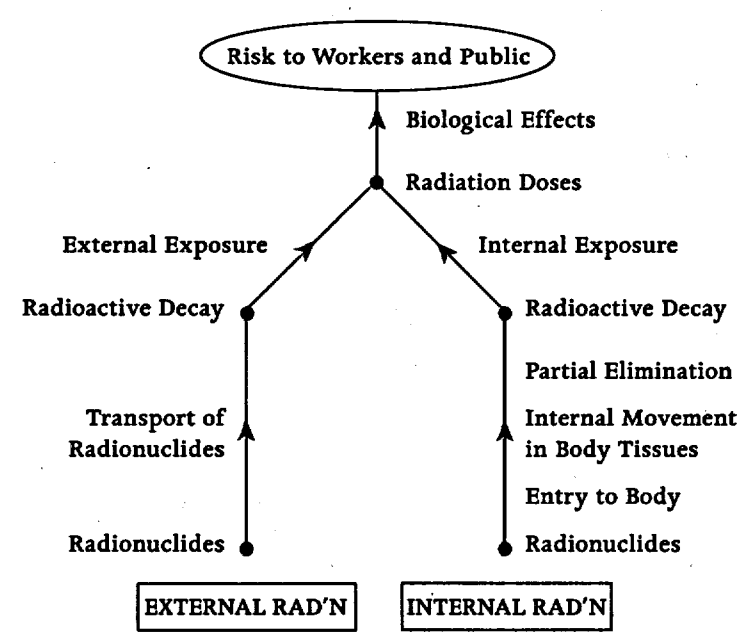
In 1977 the earlier units of dose were replaced by those of the International System of Units. The unit of "dose equivalent" of ionizing radiation is the sievert

(Sv), which is the absorbed dose, in joules per kilogram, multiplied by a weighting factor that expresses the relative biological effectiveness of the radiation. The weighting factor is unity for X rays, beta rays and gamma rays, and is currently estimated to be 20 for alpha particles and 5-20 for neutrons, depending on their energy. A convenient unit for use in radiation protection is the millisievert, mSv, equal to one thousandth of a sievert. Formerly, the dose equivalent unit was the rem and 1 sievert equals 100 rem.

A useful reference point is the natural background dose received from cosmic rays and naturally occurring radioactive materials. It varies from about 1 mSv per year to about 10 mSv per year at various locations in Canada. The average is about 2 mSv per year.

Radiation doses to people can be external, internal or both (see figure 3.1). External radiation doses in people result from exposure to sources[1] outside the body. Internal doses result from radionuclides that have entered the body through ingestion, inhalation or absorption through broken or intact skin, and are distributed, metabolized or deposited in organs or tissues of the body (see figure 3.2). Elimination of radionuclides from the body occurs primarily via urine and feces. Physical decay of the radionuclides in the body must also be taken into account – especially for the short-lived nuclides. Mathematical models are used to assist in estimating the resulting doses. Radiation dosimetry uses a variety of methods to measure or estimate external and internal doses resulting from radiation exposure. Environmental research is concerned with the definition of the major pathways of distribution of radioactive materials from source to man, and to the formulation of mathematical models to represent their behaviour in the environment.

FIGURE 3.1
Risk of Health from Radiation



Depending on the magnitude of the dose, a series of effects may be produced (see figure 3.3). High levels of radiation (e.g., > 1,000 mSv in a short period of time) may lead to early clinical acute effects, such as death, skin burns and a variety of acute radiation syndromes. The acute effects of high doses are not seen clinically at doses of radiation below a measurable threshold (500 mSv is expected to depress the blood formation by bone marrow). Radiation, even at low doses, may cause delayed effects, such as cancers in the irradiated individual (somatic effects), and hereditary (genetic) effects in the progeny of the irradiated person. These delayed effects are believed to be caused by damage to DNA that is not properly repaired. They are sporadic and occur with a probability that increases with the dose; they are therefore said to be stochastic. Much research in radiation biology is devoted to the study of the biological effects of radiation, and incidentally, of other related toxic agents whose effects resemble those of radiation.

A specific health-and-safety objective in a nuclear establishment is to protect the worker and the public from unnecessary exposure to radiation. Meeting this objective calls for a wide

[1] This term is used by the ICRP to indicate the source of an exposure, not necessarily a physical source of radiation. When radioactive materials are released to the environment as waste, the installation as a whole might be regarded as the source.

FIGURE 3.2
Principal Metabolic Pathways of Radionuclides in the body

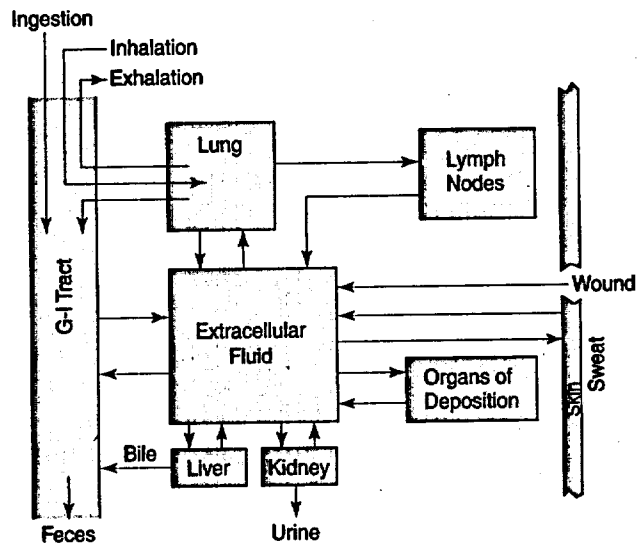
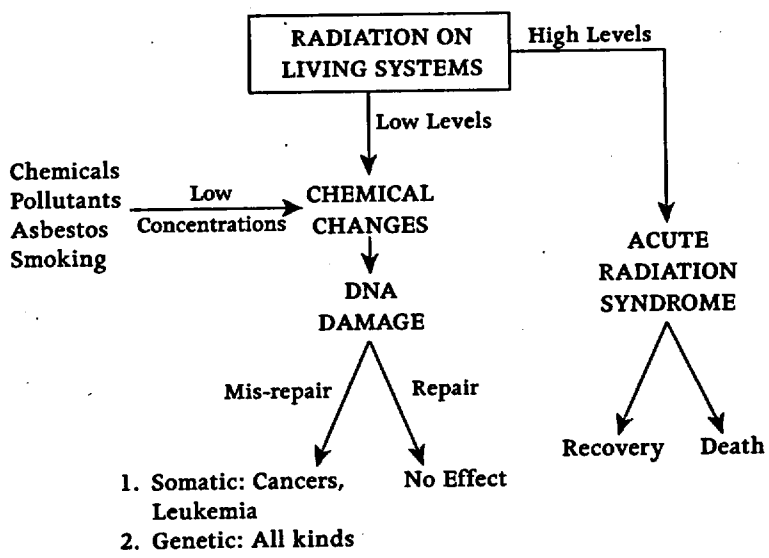


FIGURE 3.3
Biological Effects of Radiation on Living Systems



range of activities in epidemiology, biology, environmental research, dosimetry and operational health and safety. They are discussed in later sections of this chapter, but it is useful at this point to outline how the effects of ionizing radiation on humans have been determined, and how regulatory bodies arrive at recommended dose limits.

The effects (cancers and genetic changes) produced by natural background radiation or by radiation from Canadian nuclear reactors are too rare to be distinguished from the natural incidence of these effects. Risk estimates for somatic effects are obtained by following up groups of people who have been exposed to high levels of radiation, and making the prudent assumption that the rate of occurrence of these effects is proportional to the dose received, i.e., that there is no threshold below which exposure to radiation is without risk. The best information is provided by some 40,000 Hiroshima and Nagasaki survivors, who have been followed up since 1950. Estimates of the doses they received have recently (1986) been revised downwards, with a corresponding increase in the estimated risk per unit dose. The revised estimate is that the lifetime risk of fatal cancer from an accumulated dose of 1,000 mSv is 4 percent for workers (5 percent for the whole population), which is added to the normal risk, for Canadians, of about 26 percent as given by vital statistics. The risk coefficient is given for workers from age eighteen to sixty-five years, while for whole populations it is stated for exposure over a complete lifetime.

There is no evidence of any significant increase in hereditary defects in the children of the atomic-bomb survivors, or of any other group of irradiated humans that has been studied, but there is no biological reason to expect that radiation will not produce genetic effects in humans, just as it does in all other organisms that have been studied under carefully controlled laboratory conditions. Estimates of human genetic risk indicate that an accumulated dose of 1,000 mSv to parents prior to reproduction will produce a 0.4 percent chance of serious genetic effects in their children or a 2.5 percent chance of serious genetic effects in any of their progeny summed over all generations. A serious genetic effect is one that requires clinical attention at

any time in the life of the afflicted individual. These risks may be compared with a normal incidence of about 25 percent in each generation in humans.

Recommended dose limits are set by legally constituted national authorities (in Canada, the Atomic Energy Control Board), who generally follow the risk estimates developed by international bodies. Occupational limits, i.e., for atomic-radiation workers, are derived from consideration of the risks of death incurred in other industries, which range from about 1 in 1,000 per year, for forestry and fishing, to less than 1 in 10,000 for safe industries. In making such comparisons it should be recognized that occupational deaths in these other industries occur on average at age thirty to thirty-five, whereas radiation-induced fatal cancers would appear at an advanced age. The Canadian legal limits, which have not yet taken account of the revision of the bomb-survivor data, currently specify 50 mSv per year for atomic-radiation workers, and 5 mSv per year for individual members of the public. The factor of 10 or so lower for the public is believed to provide an adequate margin of safety.

As well as being subject to these dose limits, atomic-energy activities in Canada operate under the ALARA principle, enunciated by the ICRP, that "all exposures shall be kept As Low As Reasonably Achievable, economic and social factors being taken into account." Thus, at Ontario Hydro nuclear-power stations occupational doses average about 2 mSv per year (that is, equal to the average background exposure from natural sources), and the maximum dose to individual workers does not exceed 20 mSv per year under normal operating conditions. The maximum exposure of any member of the public due to Canadian nuclear-power stations is about 0.05 mSv per year, and the average to all members of the public living within 50 km of a power station is roughly 100 times less than this maximum.

EARLY HISTORY: INTERNATIONAL ACTIVITIES

Radiation protection was a major concern from the beginning of the Canadian atomic energy project. In 1944 the United States Manhattan Project provided "information necessary for guarding the health of the operators at the Montreal Plant," and in 1945 a research laboratory was set up. A 1945 document from the Montreal Laboratory is remarkable for the advanced level of expertise that was available, and for the postulate that somatic effects might be proportional to dose, as genetic effects in fruit flies apparently were. This consideration led to a decrease in the accepted dose limits for the Canadian and British projects to 150 mSv per year, the corresponding figures being 500 internationally, and 300 in the United States. However, even at that time attempts were made to keep actual radiation exposure of workers as low as possible and well below limits.

Information from the Manhattan Project was provided on a need-to-know basis. In order to obtain additional information on the toxicity and metabolism of ingested radionuclides, one of the Chalk River biologists (G.C. Butler) commenced experiments, exposing rats to uranyl nitrate in 1946. After this preliminary study, Chalk River representatives visited the biomedical staff at the Manhattan Project in New York. After several hours of scientific discussion, the biomedical group seemed satisfied with the competence and credentials of the Canadians. Files on experimental animal studies at Berkeley were made available. With this preliminary information on uranium, plutonium and other radionuclides, Butler was able to compile one of the earliest reports on the topic in 1947, entitled "The Biological Basis of Maximum Permissible Exposure for Workers in the Field of Atomic Energy."

The purpose of Butler's report was to calculate the intakes of uranium, plutonium, strontium-90, iodine-131 and radioactive isotopes of krypton and argon that would be equivalent to one of the two "tolerance doses" (a maximum of about 150 mSv per year of radiation from external sources or a maximum of 0.1 microgram of radium-226 fixed in the body, primarily in the bone) accepted for workers at that time. This calculation required a fairly detailed understanding of the probable concentrations of each radionuclide in different tissues of the body and of the length of time that the various radionuclides were likely to remain in those tissues; much of the required information on these topics was obtained from United States' files.

In 1949 a tripartite conference on radiation-protection principles was held at Chalk River with participants from the United States, the United Kingdom and Canada. The notes and minutes of this conference provided a multitude of concepts, ideas, questions and resolutions to problems that set the scene for decades to come in international and national recommendations and regulations. There were eight Canadian representatives, including a Chalk River scientist, G.C. Laurence, who was to become a member of the International Commission on Radiation Units and Measurements in 1950, and later became president of the Control Board.

The conclusions of the tripartite conference had great impact on the recommendations of the ICRP. At least one person from Chalk River staff has participated in the deliberations of the ICRP or its committees since it was reorganized and renamed in 1950. C.G. Stewart served for two terms (1969-77) as chairman of the ICRP and was awarded a gold medal for his services.

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) was established in 1955. Its purpose was to collect and evaluate reports on the levels of ionizing radiation and radioactive substances in the environment, and on the biological effects of ionizing radiations on humans. The ICRP relies heavily on the work of UNSCEAR for risk estimates and thus for deriving its own recommendations on corresponding dose limits. Many Canadians, including AECL staff, have made contributions to the work of UNSCEAR. Chalk River staff have contributed to UNSCEAR since its inception.

Because of the close liaison AECL scientists had with international organizations such as ICRP and UNSCEAR, they were familiar with developments and thinking at the forefront of radiation protection. Consequently, AECL management had confidence in these scientists when they advised on principles of radiation protection, risk estimates and dose limits. AECL adopted ICRP recommendations before they were incorporated into Canadian regulations, and sometimes even earlier than their formal publication by ICRP.

The rest of this chapter will outline AECL's contributions to the subject of radiation protection. Many of these contributions were directed to specific Canadian needs. However, from the early days it was realized that Canada could not alone do all the necessary research. To maximize access to the international pool of knowledge in this area, AECL published work of importance to others. Information exchange in the life sciences concerning radiation was rather free and open on the international scene.

EPIDEMIOLOGY

Significant effects have been observed in the epidemiological studies of miners who were in the past exposed to very high concentrations of radon and its decay products. Four of the eight major studies around the world relate to mining sites in Canada. Chalk River personnel have been involved with the radiation protection of uranium miners since the fifties, and have periodically reviewed the results of these epidemiological studies. An appreciable excess of lung cancers is observed in these early miners. Human population studies by Health and Welfare Canada failed to reveal any measurable effects of much lower concentrations of radon-decay products from natural sources on lung-cancer incidence in various Canadian cities, where the mean concentrations in the air in homes varied by tenfold. The effects of low concentrations of radon are too small to be measured in human populations.

A Chalk River scientist instigated an important epidemiological project on excess breast cancers in Canadian women exposed to multiple fluoroscopic examinations during treatment for tuberculosis in the thirties and forties. Accumulated average doses to the breasts were about eight times greater in the Nova Scotia portion of this study than in the Japanese bomb survivors. The study on excess breast cancers was carried out by the National Cancer Institute of Canada and has been widely cited by international scientific committees concerned with risks of radiation-induced cancers. The doses to the Nova Scotia patients were greater because they faced the fluoroscopic machine, whereas in Ontario they did not.

A variety of studies has been carried out in various countries, including Canada, on workers who were occupationally exposed to whole-body radiation, and on populations who live in areas where natural background radiation is higher than

average. Those studies have been inconclusive; if there is any effect, it is too small to be detected against the normal high background of cancers attributable to other causes.

Epidemiological records were studied of Chalk River employees who had died between 1950 and 1989. Separate follow-up studies of 412 Chalk River workers who received an average of about 20 mSv each year for twenty-two years, and of 1,090 Chalk River personnel involved in the cleanups of the 1952 NRX and the 1958 NRU accidents, were also carried out. No excess cancers were observed in these studies and there was no excess of leukemias, over and above those expected in the general population. For example, there were 153 cancer deaths observed among all Chalk River male employees (including retirees) in 1966-89, compared to an expected value of 172 for the general population of Ontario; total deaths from all causes in the same group were 561 compared to 676 expected. Both Whiteshell and Chalk River staff were involved in follow-up studies of all 14,000 AECL employees who were employed up to July 1980. The data, as analyzed by the National Cancer Institute of Canada, are yielding similar results. Personnel from the University of Ottawa undertook follow-up studies of the armed-forces personnel who were involved in the cleanup of the NRX and NRU accidents; they did not reveal any excess cancers. These results are reassuring; no effects of radiation exposure were detected. The primary value of this type of study has been to allay concerns about the health effects of low-level radiation and to demonstrate that these effects are not being grossly underestimated by the ICRP risk estimates.

In order to estimate the health impact of radiation-induced mutations in humans, we need to know a) the normal incidence of genetic disorders in human populations, b) the normal rates of mutation (or genetic change) that are responsible for this normal incidence, and c) the increased rates of mutation that are induced by radiation exposure in experimental studies on mice and other organisms. The results of these experimental studies on mice are extrapolated to humans. To obtain quantitative information on radiation-induced mutation rates, in the fifties and sixties the Oak Ridge National Laboratory set up the multi-million dollar mouse experiment in which the frequency of coat-colour mutations in mice was scored after partial irradiation. This enormous and expensive study was funded by the United States' Atomic Energy Commission. AECL biologists were able to make important contributions to items a) and b) within their limited budgets.

As a result of his involvement in UNSCEAR discussions on the genetic burden in human populations from fallout from atmospheric testing of nuclear weapons, and the need for data on the normal frequency of genetic disorders in human populations, one of the Chalk River geneticists turned his attention to British Columbia, which had a good vital-statistics registry of births, deaths and marriages, plus a first-rate registry of handicapped children. These registries were separate and needed to be merged together to obtain the required information. Up to the late fifties, such consolidations were carried out manually, but he decided to develop a computer-assisted record linkage for this purpose. He received great help from computer experts at Chalk River in his early endeavours. The first problem was to devise means by which computers could recognize records pertaining to the same individual, even when the birth dates, spelling of names, and such did not agree precisely, as was frequently the case. By adding other bits of information, for example, mother's maiden name, ages, addresses, and so on, it became possible to identify specific individuals. Once the programming of methods to compare names and other data was established, it was possible to carry out record linkages between the registries of vital statistics and the registry of handicapped children. In this way quantitative information on the frequency of genetic disorders in the British Columbia population, their heritability and the longevity of afflicted persons, was obtained. The resulting data were extensively quoted in the 1977 UNSCEAR report on genetic effects of radiation. The principal investigator pointed out that the same methods of records linkage could be used to study the health of workers exposed to radiation and other carcinogenic agents, and worked together with collaborators at Statistics Canada to carry out studies of this kind. The technique of record linkage is now widely used by universities and other government agencies, such as Statistics Canada and the Royal Canadian Mounted Police.

Another Chalk River geneticist carried out a careful re-examination of the published literature on spontaneous mutation rates for various genetic disorders in humans. His review and corrections of accepted mutation rates was published in 1982 and cited extensively in the 1990 report on genetic hazards by the United States Committee on the Biological Effects of Ionizing Radiation (BEIR).

LINEARITY

The assumption, recommended by the ICRP, that the probability of radiation-induced cancers and genetic changes increases in direct proportion to any increase in radiation exposure is generally regarded as reasonable and prudent for purposes of radiation protection, but is very difficult to prove or disprove. The data on radiation-induced cancers from the bomb survivors and the early uranium miners are compatible with this assumption, but do not have sufficient precision to prove that it is true at the low radiation doses that are relevant to the application of nuclear energy. Other data, notably epidemiologic data on bone cancer in humans exposed to low doses of radium, and animal studies on lung cancer in rats exposed to low concentrations of radon, can be interpreted in terms of a threshold for prolonged exposure at low dose rate.

The best evidence for linearity comes from studies of genetic change in lower organisms. In 1955, a French scientist showed that the frequency of a genetic change involving the liberation of a bacterial virus was proportional to radiation dose down to 3 mSv. A 1980 Chalk River study of the frequency of genetic change in yeast cells showed linearity over a very wide range of doses - from several tens of sieverts down to 10 mSv, below which level the effects were too small to be measurable.

It is of interest to study more complex organisms than bacteria and yeast. One of the Chalk River biologists used wasps (Dahlbominus), which readily provide enough offspring for adequate statistical accuracy, to study the frequency of eye-colour mutations following parental irradiation. A total of half a million wasps was examined, and a linear dose effect was established down to 150 mSv. Reaching 75 mSv would have required another two million wasps, and this was not attempted. To test for linearity experimentally in complex organisms, high doses of radiation need to be used to obtain significant statistical results on a limited number of organisms.

A further attempt to probe linearity was carried out using a fish (trout), which is a vertebrate and thus more closely related to humans. To execute this experiment, a senior technologist travelled to a fish hatchery in Jasper, Alberta. He took along a portable cobalt-60 irradiator and thousands of phials to hold the fish embryos. On his return to Chalk River in 1967, the technologist scored the observed malformations in the fish embryos. A linear dose-effect relationship for these genetic endpoints was established down to 250 mSv in fish embryos from irradiated sperm. It took 40,000 fish embryos to establish the 250 mSv point. The natural mortality and infertility were reduced by 250-500 mSv of radiation, even though genetic malformations were increased. Not all of the biological effects of low doses of radiation are harmful. Hormesis is a term applied to the beneficial effects of radiation and there are reports of such effects in the scientific literature, but it remains a controversial topic. In 1994 UNSCEAR reviewed this topic as "Adaptive Responses to Radiation." The term is broader and is not restricted to beneficial results only.

Another study on radiation-induced genetic changes examined the intelligence of rats, as measured by their ability to learn and to remember how to find their way through complex mazes. However, despite repeated high doses to the male gonads in each of several generations, no significant effects on the average learning ability or on the distribution of learning abilities could be detected. The first results from this study were published in 1961. Although innate learning ability is pre-determined by the action of many genes, it was, reassuringly perhaps, found not to be affected by repeated doses of radiation to the gonads in successive generations. Lower doses were not tested, since no effect was found at high doses.

MOLECULAR BIOLOGY

Early files (1944-47) of the Canadian Atomic Energy Project reveal that comprehensive plans for biomedical research programs had been made. These plans involved studies on the mechanism of action of various types of radiations on cells, with special emphasis on nucleic acid and nucleoprotein components of cells. These were considered important because of the concept that DNA (one of the nucleic acids) was in fact the genetic material in the nucleus of the cell. In the first half of the 20th century it was thought that DNA was a simple organic molecule composed of four nucleotides containing the bases: adenine, thymine, guanine and cytosine. With the introduction of mild and gentle techniques for isolation of DNA in the forties, it was recognized that DNA was not a simple molecule but a large polymerized, complex molecule which could serve as the cellular genetic material.

In the fifties a growing body of evidence indicated that genetic effects of ionizing radiation and ultra-violet (UV) light were the consequence of damage to DNA in organisms. One of the Chalk River researchers in the fifties started to chemically characterize the radiolytic products of the bases in DNA. In the early seventies, many publications on the identification of radiolytic products began to appear, but this pioneer work at Chalk River predated those studies by a number of years.

In the late sixties, scientists at Chalk River became concerned that empirical quantitative studies of the biological effects of high doses of radiation, epidemiological follow-ups, and costly, large-scale animal experiments were not going to elucidate the effects of low doses and low dose rates of radiation. It was felt that more detailed information on the mechanism of radiation-induced damage in DNA and its subsequent enzymatic repair would provide insight into the effects of low doses and low-dose rates of radiation in living systems. AECL recruited a team of molecular biologists to work on DNA damage and repair. The objective was to gain more detailed information on mechanisms, which would complement the data on observed biological effects of high radiation doses and facilitate extrapolation down to low doses at low-dose rates, where the biological effects cannot be measured directly. These workers produced some notable firsts in biological sciences, which were recognized by awards from Canadian and American scientific societies.

The AECL researchers worked with radiosensitive and radioresistant strains of bacteria, yeasts and cultured human cells. The initial amount of damage (such as strand breaks and base damage) that could be measured in the DNA immediately after exposure of living cells to ionizing radiation was found to be independent of the type of cell used. The differences between radiosensitive and radioresistant strains of the same type of cell could be attributed to natural mutations that had inactivated one or more of the repair systems that normally remove most of the radiation-induced damage in living cells. The same principles that were found to govern the relative radiosensitivity of bacteria and yeast cells were found to apply to cells derived from humans with two specific hereditary diseases. One of these diseases (ataxia telangiectasia) rendered the persons, and cells derived from them, more sensitive than normal to the toxic effects of the high doses of radiation used in cancer radiotherapy. The second (xeroderma pigmentosum) rendered the persons suffering from this particular disease, and cells derived from them, more sensitive to the effects of UV light. In both cases, the disease was shown to be associated with inherited deficiencies in one of the DNA repair systems. This work, which was supported in part by a large contract from the United States National Cancer Institute, contributed to an important understanding of the relation between DNA damage and cancer development (chapter twenty).

A micro-organism isolated from food that had received very high levels of radiation proved to be very radioresistant. It was known as *micrococcus radiodurans*. It was found that the radioresistance was not inherent, but due to the extremely rapid enzymatic repair of the lesions induced in the DNA by radiation.

A Chalk River scientist in collaboration with a researcher from the University of Toronto found that a wide variety of organisms, simple as well as complex, contained long stretches of polypyrimidine tracts in their DNA. The polypyrimidine tracts consisted of a series of cytosine and thymine bases

joined together without any purine bases being involved. Although the discovery of polypyrimidine tracts contributed to the knowledge of the structure of DNA, the significance of the structure is largely unknown.

A scientist at Chalk River was the first to demonstrate that phorbol esters known for tumour promotion caused damage of cellular DNA. He was able to demonstrate this damage because he had developed a fluorimetric technique to measure single and double strand breaks in DNA of cells. Attempts to market this assay were unsuccessful.

In risk analysis of energy production from a variety of sources it is commonly assumed that the average biological risk to people is constant. It was demonstrated by a Chalk River researcher that the risk of tumour causation in mouse skin by carcinogens was not constant, but could be altered by varying the regimen of initiators and promoters, such as their concentrations and the time between applications of the initiators and promoters to the skin.

Attempts were subsequently made at Chalk River to develop a test on white blood cells to identify cancer-prone and radiosensitive persons in the general population. Both ionizing radiation and chemical carcinogens were used in these studies. Further effort to develop a reliable test is ongoing. A simple and reliable test for the differences in radiosensitivity of individual humans would be particularly useful in cancer radiotherapy, and many studies on the radiotherapeutic aspects are being pursued in other laboratories around the world. The lack of reproducibility of these sensitivity tests in the normal work force has resulted in the postponement of these studies at AECL. Furthermore, the utilities who have funded part of this work are not interested in continuing support of this type of research.

The medical biophysics group at the Whiteshell Laboratories complemented work on the molecular biology of DNA by research efforts on early events occurring in fractions of a second following the deposition of energy into the system. These early physical events include, for example, the production of free radicals (see chapter seven), which cause chemical damage in DNA and in other biological constituents of cells. The chemical mechanism of the interaction between radiation and chemical carcinogens was investigated. The role of naturally occurring enzymes, such as superoxide dismutase and peroxidase, was studied to learn how these enzymes protect biological structures from being damaged by the highly reactive free radicals. It was found that these enzymes degraded the potent free radicals to less toxic molecules. This work has greatly improved our understanding of how radiation produces biological changes in living organisms.

The mechanism of action of chemicals, known as radiosensitizers, which increase the sensitivity of cells to the lethal effects of radiation, was also investigated. The ultimate aim of these studies was to improve the radiotherapy of cancers. The lack of cures for, and the recurrences of, cancers are believed to be due to the fact that cells in the centre of tumours are not killed because they are anoxic or lack oxygen, and for this reason are radiation resistant. Although radiosensitizers are effective in counteracting the radiation resistance of anoxic cells, cancer treatment centres have found that radiosensitizers have considerable toxic side effects. The sensitivity of mammalian cultured cells to heat and radiation was also studied. Results of these studies have implications for the use of hyperthermia in the treatment of cancers. Preliminary promising therapeutic results were obtained on clinically terminal cases.

Another focus of research at Whiteshell Laboratories was work on membranes. Membranes are important in cell functions, because they control the exit and entry of important food stuffs, waste products, minerals and water, while retaining macromolecules such as DNA and proteins. For this reason it was suggested that damaged membranes could lead to delayed effects (cancers and genetic defects).

One of the scientists at Whiteshell used model membranes prepared from biological materials to study the integrity of the membranes after exposure to low doses and low-dose rates of radiation, as well as to compare the effects of X rays with those of tritiated water. From his studies, he claimed that as the dose rate was decreased, the effects on the model membrane *per unit dose* increased. This observation, which received some publicity in the popular press, is at times called the "inverse dose rate effect," where the effectiveness of low-dose rates is greater than

higher dose rates. The interpretation of these data generated considerable controversy within and outside AECL. Most of the AECL scientists did not accept the results or the interpretation of the experiments. The applicability of results on simple membrane models to complex living systems remains questionable. In 1980 the United States National Research Council's BEIR committee reviewed this work and concluded on page 464 of its report that "although it is well recognized that membrane integrity is essential for normal cell function, there is inadequate basic understanding of membrane structure and function on which to base a detailed theory of radiation-induced damage mechanisms." The work on model membranes was not reviewed by UNSCEAR, because of the work's absence of evidence that it was relevant to carcinogenesis, and because there was more direct evidence from animals on the induction of cancer at different dose rates.

TRITIUM

Tritium is a radioactive isotope of hydrogen that is produced naturally in small amounts by cosmic radiation. Large amounts were also released into the atmosphere during the testing of nuclear weapons in the fifties and early sixties, and spread around the world in the form of tritiated water in the atmosphere. This increased the natural concentration of tritium in the Ottawa River and in lakes by a factor of more than 100. Due to its radioactive decay (radioactive half-life 12.3 years) and gradual transfer into the ocean depths, the concentration of tritium in rivers and lakes has been decreasing steadily since it reached its peak in 1963-64, although the concentrations are still much higher than the natural levels.

Large amounts of tritium are formed in heavy-water reactors. Although every reasonable effort is made to contain this tritiated water, inevitably small amounts leak out, resulting in some exposures of workers, the public and the environment. Tritium is thus of special concern both to AECL and to the utilities that operate CANDU power reactors.

Exposure of workers at Chalk River to tritium has been systematically measured by urinalysis since 1956. In general, tritium exposures have been very small at Chalk River, but radiation doses from tritium at the Pickering plant increased to a point where they represented a third or more of the total occupational dose. This was a major reason for the construction of tritium-extraction plants to remove tritium from the heavy-water moderator (see chapter fourteen).

To test the various metabolic parameters applicable to tritiated water (HTO) in humans that were used by the ICRP in the sixties, volunteers (primarily research scientists in health sciences) were exposed to small amounts of HTO vapour in a special exposure chamber. A critical finding was that the rate of uptake of HTO by lungs was equal to the rate of uptake by skin. The effective half-life in the body was found to be ten days. This value was subsequently adopted by the ICRP. It was also observed that the ten-day half-life could be further reduced by a factor of three or so by drinking copious amounts of fluids. The effectiveness of protective clothing on the uptake of HTO by workers was also studied. For employees who need to work in an atmosphere of low levels of HTO, the most practical protective clothing is a ventilated plastic suit with a fresh air-line mask.

Improved stationary and portable monitors, compensated for gamma radiation, have been built by AECL to measure tritiated water vapour in air. An automated tritium urinalyzer was also built so that workers could monitor themselves at nuclear power stations. Ontario Hydro personnel claimed the use and maintenance of these urinalyzers was too fussy for routine use and they were therefore discontinued.

Studies on human volunteers and animals at Chalk River confirmed that, while HTO is actively metabolized and thus retained in the body, inhaled tritium gas (T_2) is not retained in the body but is quickly breathed out again. Very small amounts of inhaled tritium gas are oxidized in humans to HTO, and only by micro-organisms in the gastro-intestinal tract. The net result is that tritium gas is 10,000 times less radiotoxic than HTO. With the building of the tritium extraction plant, which removes tritium from tritiated heavy water and processes it in the form of a gas, it became necessary to be able to measure tritium gas and tritiated water separately. Monitors were developed using Nafion (a plastic film that is permeable to HTO but not to T_2),

which provided the capability to measure these two forms of tritium separately. AECL research on the measurement and relative toxicity of tritium gas is also applicable to monitoring in the fusion research facilities that use tritium.

It was commonly believed by most scientists that tritium gas (T_2) is oxidized very slowly in the atmosphere, but some doubts had been expressed. One of the Chalk River scientists arranged an international collaborative experiment to measure the oxidation of T_2 released into the air. Scientists from Germany, France, United Kingdom, United States, and Canada cooperated in this experiment at Chalk River. It was confirmed that the rate of oxidation of T_2 in air proceeded very slowly. However, when the T_2 was deposited on the ground, it was oxidized rapidly by micro-organisms present in the soil.

Studies on the relative biological effectiveness of tritium beta-rays were carried out in Chalk River at the request of the health and safety division of Ontario Hydro. Various reports in the literature suggested that the relative biological effectiveness of tritium beta-rays range from one to four, depending on the biological endpoint and whether X rays or cobalt-60 gamma rays are used as the reference radiation. This creates considerable problems in the conversion of absorbed tritium doses to dose equivalents in units of mSv. It was decided to investigate this matter using an endpoint that is directly relevant to radiation protection, namely induction of cancer. The reference radiation used for comparison was X rays, which account for more than 90 percent of all man-made exposures to the general population. At Chalk River, two long-term studies were carried out over a period of about ten years by staff members from dosimetric research and radiation biology. The first study explored acceleration of the appearance of breast cancers in female rats. The second, funded in part by the Control Board and the utilities, explored the induction of myeloid leukemia in male mice from a special strain obtained from the Harwell laboratories in the United Kingdom. Both studies showed that the relative biological effectiveness of tritium beta-rays was about 1.2 compared to X rays. Recent recommendations by the ICRP are that tritium beta-rays and X rays should be considered to be equally effective for practical purposes in radiation protection.

AECL staff doing research in dosimetry, environmental sciences and radiation biology have all been involved from time to time in tritium research, and considerable expertise in this area has been acquired. Chalk River staff were thus invited by the Control Board Advisory Committee on Radiological Protection to prepare a report on the toxicity, metabolism and dosimetry of tritium; this report was approved for publication by the Control Board in 1987.

ENVIRONMENTAL RESEARCH

AECL's environmental research program has always been concerned with how radioactive material moves through the environment (pathway analysis), and its uptake by plants and animals. Mathematical models are designed to explain the dispersal of radioactive materials in the environment, and the validity of these models is tested by the analysis of radioactivity in environmental samples. Routine environmental surveillance was carried out on samples of water, air, plants and soil to ensure that releases of radioactive materials from facilities at Chalk River were below specified limits. The geographical locations of Chalk River and Whiteshell provide unusual opportunities for research studies of this kind, in that short- and long-term experiments can be carried out in field stations located in natural terrestrial and aquatic ecosystems. Both research sites are located in remote fenced-in areas, and are protected from external disturbance. The fact that the field facilities are in close proximity to research laboratories is also unusual, which attracts national and international scientists from government agencies and universities to undertake collaborative studies with AECL.

Perch Lake Basin is the main location at Chalk River for research on environmental radioactivity. This area contains two experimental disposal sites, where low-level liquid radioactive wastes from Chalk River operations have been deposited since the late forties. Since that time, small amounts of radioactivity have migrated through the surface and ground-water systems to other areas of the basin, including Perch Lake, and have provided an unusual opportunity to study the behaviour of radioactivity in natural ecosystems.

For example, researchers undertook studies of the migration of cesium-137 and strontium-90. These two radionuclides are of biological significance because they are readily taken up by living organisms; their migration rates are influenced by the geochemistry of the system through which the water travels. It was found that the migration rate of strontium-90 is 3 percent of that of ground water in Chalk River soils, while for cesium-137 it is 0.3 percent. This type of information is critical for the waste-management program (see chapter nineteen).

Before the migration rates of strontium-90 and cesium-137 were determined, it was thought that large amounts of radionuclides might enter Perch Lake and escape into the Ottawa River via the Perch Creek outlet. Preliminary plans were made to build a treatment plant on the Perch Creek outlet to remove the radioactive substances from the water by absorption onto a special clay. However, measurements indicated that the rate of movement of strontium was so slow that it would take about 300 years for large amounts of strontium to reach Perch Lake from the disposal sites, and by this time the radioactive strontium-90 (its half-life is twenty-nine years) would have decayed by a factor of about a thousand. Therefore, a treatment plant was not necessary.

In another research project associated with waste management, high-level fission products were incorporated into nepheline syenite glass blocks and buried in the water table in the Perch Lake basin in 1959. In a continuing study since that time, the leaching of radioactivity from these blocks has been measured. Although ground water has flowed past the blocks for more than thirty years, the amounts of radionuclides released are small, being less than 0.003 percent of the total amount originally buried. A small amount of the released radioactive material became incorporated into micro-organisms and transported biologically in this form for short distances. The minute amounts of radioactive materials leached from the glass blocks point to the feasibility of disposing of high-level wastes by incorporating them into glass blocks and emplacing them in deep, underground repositories. Chapter nineteen discusses this topic in more detail.

Perch Lake water contains very small, but measurable, amounts of radioactive cobalt-60 and strontium-90, introduced by migration via ground water from the experimental disposal sites. The lake supports a rich population of plant and animal life. In 1955, a research program was begun to study the processes that distribute radioactivity throughout a lake ecosystem. Much of the cobalt-60 and strontium-90 entering the lake is absorbed by organic particles and deposited on the lake bottom, but some remains in solution and is taken up by vegetation and animals living in the lake water. Bio-discrimination, the process by which living organisms reject one chemical element in favour of another, plays an important role in the exclusion of radioactive substances from organisms. It was found at Perch Lake that there was a progressive *decline* in the concentration of cobalt-60 and strontium-90 as they are transferred to higher levels of food chains. In other words, predators, such as carnivorous fish, have lower concentrations of cobalt-60 and strontium-90 per unit body weight than do forage fish, insects and plants. It is now generally agreed that the same principle is valid for most other radionuclides, with a few exceptions, such as tritium and soluble cesium-137 (an analogue of potassium), which are incorporated without discrimination into all living organisms. Most radionuclides do not bio-accumulate as one progresses up the food chain, thus differing from DDT and other organic toxins. This has great importance in the assessment of the impact of radioactive releases into the environment.

To study the evaporation of water from lakes, the presence of traces of tritiated water in Perch Lake was utilized. To do this experimentally it was necessary to carry out a total tritiated water budget; that is, the amount entering the lake by various routes and the amount leaving the lake in the outlet stream plus the amount being evaporated to the atmosphere. The channeling of the inflow of ground water into Perch Lake via the main inlet was accomplished by building a dyke (which also served as a road) on the northwest shores of Perch Lake. The amount of tritiated water in evaporation was determined by analyzing water collected with molecular desiccant that had been installed on meteorological towers at Perch Lake. The amount of tritiated water entering and leaving the lake in flowing waters allowed long-term parameters (days) to be used in the model, while the evaporated tritiated water was used to account

for short-term components (hours) in the model. This study enabled the researchers to develop a model for the evaporation of water from a lake, and this model was successfully applied to other lakes in Ontario.

In assessing the impact of radioactive releases to the atmosphere from nuclear establishments and nuclear power plants, cautious assumptions are made concerning the dispersion in air. It is assumed that wind speeds are negligible and that there is a temperature inversion, which does not allow the plume to rise and disperse in the upper atmosphere. If releases under such restrictive conditions are less than regulatory maximum permissible releases, then releases under other atmospheric conditions more favourable for dispersion would also be acceptable.

Instead of relying on hypothetical weather conditions, it was decided to measure argon-41 concentrations at ground level and downwind from the main stack, where airborne emissions from the reactors are released. Argon-41 is produced by neutron activation of argon in the cooling air as it passes through the NRX and NRU reactors. Staff in dosimetric research were able to build a monitor to measure argon-41 specifically. These monitors were located in semicircles on the ground at varying distances from the stack. Ground concentrations of argon-41 were recorded continuously over short time intervals. In this way one could analyze the argon-41 concentration and obtain statistics about the frequency of minimum dilutions of argon-41 emitted from the stack. Correlation of these concentrations with observed weather conditions gave a more realistic model of dispersion under various weather conditions.

As part of the overall environmental program, it was important to obtain experimental data on the dispersion of radioactive material released into bodies of water. Since some of these bodies of water were public areas, fluorescent dyes rather than radioactive materials were used as tracers. Chalk River staff released concentrated solutions of fluorescent dyes and, at different times after the release, measured the concentrations of dye downstream in a river or in surrounding waters in a lake, using a fluorimeter mounted in a boat. Such dispersion studies were made in the Ottawa River at Chalk River, in the Winnipeg River at Whiteshell, and in Lake Huron at Douglas Point. The data was used to establish dispersion models at these various sites. The concentrations of tritium measured downstream in several places in the Ottawa River after an accidental release of tritiated water from Chalk River in 1988 was in good agreement with the values predicted by the model.

Environmentalists considered it important to undertake preoperational surveys in areas surrounding nuclear facilities before they were commissioned. A minimum acceptable survey would determine natural-background radioactivity, type and abundance of plant and animal life, and the dilution capacity of waters accepting discharges from the facilities. It was felt that there would be more credibility to the study if it were carried out by an independent outside body, rather than by AECL. Accordingly, a contract was negotiated with the Great Lakes Institute of the University of Toronto to carry out a preoperational survey at Douglas Point. It was known that as a result of normal thermal shocks in Lake Huron, fish die by the thousands and are thrown up overnight on the beaches of the lake. If we did not have this phenomenon well documented, critics might blame the fish deaths on radioactive releases at Douglas Point. The preoperational survey was carried out and reported by the Great Lakes Institute.

Water used to cool power stations is discharged into rivers or lakes, causing an increase in temperature of the receiving waters. This increase is often referred to as thermal pollution. However, to explore the beneficial uses of this waste heat in cold Canadian waters, an experimental facility was installed at Lake Maskinonge in the Chalk River enclosed area. Optimal conditions were determined for the growth of living organisms. Heated water is now being used for commercial greenhouses and for commercial fish-rearing ponds at the Bruce and Pickering Nuclear Generating Stations, respectively.

Using the Tandem Accelerator Superconducting Cyclotron, very small amounts of carbon can be analyzed for carbon-14 content. Using this technique it was possible to estimate the age of ground waters containing carbonate, from a variety of places in the Canadian Shield, in connection with the assessment of waste-disposal concepts (chapter nineteen).

At Whiteshell, two studies of the effects of ionizing radiation on plants and animals in the environment were carried out. The FIG (field irradiator gamma) and ZEUS (zoological environment under stress) projects were designed for that purpose. Field-irradiation studies had been carried out previously in the United States. The aim of the FIG project was to detect any ecological changes in plant communities that are continuously irradiated under the more severe conditions of growth on the Canadian Shield in Manitoba. Part of a natural forest, one kilometre in diameter, was enclosed with a fence and a large radiocesium source was placed in the centre of this area on a tower. The annual doses in the FIG area ranged from approximately 460,000 times background near the source to thirty-five times near the fence. Since 1973, observations of radiation effects have been continuously recorded. The most radiosensitive of the trees are pines and these died off in the high-radiation areas. Annual exposure rates required to kill the pines were about 100,000 times the background exposures from natural sources. No effects were observed at lower radiation doses, where dose rates were still much higher than those due to natural background. The FIG project was terminated in 1985. The ZEUS project, concerned with irradiation of natural populations of meadow voles, did not yield statistically conclusive results, but no catastrophic effects were observed.

Many other studies have been carried out by the environmental research group at Whiteshell, mostly concerned with the nuclear-fuel-waste-management program.

In the philosophy of radiation protection, the ICRP has long believed that if individual humans are protected, then the survival of species of other organisms in the environment will also be ensured. In nature, the species, as distinct from the individual, is of paramount importance. A Chalk River scientist's detailed review of the literature – including highly relevant data from the FIG project at Whiteshell – confirmed that the effects of radiation on environmental organisms other than humans need not be a critical consideration for the Canadian nuclear-fuel-waste-management concept. A recent review by the International Atomic Energy Agency has supported this conclusion, although this review noted a need for caution in the case of endangered species.

RADIATION DOSIMETRY

The instrumentation for measurement of radiation levels, radiation contamination, etc., was fairly elementary at the beginning of the project and has been under continuous improvement ever since. Radiation produces detectable physical and chemical effects in gases, liquids and solids that may be used to obtain estimates of radiation doses. The physical effects in particular allow the detection of very small doses.

Until 1971, the exposure of workers at AECL to external radiation was routinely monitored using photographic films. Each worker wore a film badge for two weeks and then the film was processed and developed in a standard manner. The optical density of the processed film, which is increased by exposure to radiation, was determined and converted to dose by a calibration curve, where optical density was plotted against known exposures to different radiation doses. The use of films for routine monitoring of over 3,000 AECL employees plus special extra films for particular types of work (finger and extremity film) was very labour-intensive and did not lend itself to automation. Fortunately, the phenomenon of thermoluminescence could be exploited for this purpose. When radiation is absorbed in certain crystalline substances, such as lithium fluoride, energy is stored in the crystal-lattice structure. If the crystal is heated later to an appropriate temperature, it releases the stored energy as light, whose intensity is proportional to the radiation dose and can be measured using a photomultiplier. One of the researchers at AECL worked on thermoluminescence dosimeters (TLDs), and developed an automated TLD reader. This made possible routine monitoring of workers; TLDs replaced film badges at AECL in 1971. TLDs are incorporated into the AECL personal dosimeters that all radiation workers are required to wear. AECL, Health and Welfare, Ontario Hydro, and Hydro-Quebec have used the automated TLD reader developed at Chalk River. Only the Point Lepreau personnel in New Brunswick use a different commercial TLD reader. Owing to recent developments in the miniaturization of electronics, it is now possible to construct reliable direct reading dosimeters. When these dosimeters become

sufficiently rugged and commercially available, they are likely to replace the TLDs.

Neutrons having a wide range of energies are produced by accelerators and reactors. Measurement of doses from neutron sources is particularly difficult since no single detector has been found to have a response that matches the biological effects of neutrons of different energies. Some detectors are highly sensitive to low-energy neutrons and have negligible response to high-energy neutrons; others have the opposite characteristic. Chalk River personnel studied and developed the use of CR-39 (a polycarbonate plastic film) as a dosimeter for monitoring neutrons, and it is used throughout Canada for this purpose. CR-39 is damaged by exposure to fast neutrons and this damage is enhanced to form visible holes in the plastic by electrochemical etching. The density of holes can be measured by a reader and converted to neutron dose. Slow neutrons are detected by placing the CR-39 film adjacent to boron or lithium; these materials produce alpha particles when exposed to slow neutrons, and the alpha particles in turn produce detectable damage in CR-39.

Another staff member developed a novel and sensitive method for the detection of neutrons. Small droplets of Freon liquid are incorporated in a transparent polyacrylamide gel. On exposure to neutrons, Freon gas is released to form bubbles, which grow in size to become visible to the naked eye. The number of bubbles formed is proportional to the neutron dose. This device became known as the "bubble detector". The physicist involved had no experience in the making of polyacrylamide gels, but a radiation biologist working in the same building made them routinely to separate DNA fragments in the gel by electrophoresis. The biologist quickly trained the physicist and his staff to make these gels. This transfer of technology illustrates the advantage of working in a laboratory, where scientists of different disciplines are able to cooperate closely to solve technical problems. The bubble detector attracted a fair amount of attention inside and outside AECL and it was finally organized as a separate business venture, known as Bubble Technology Industries (BTI) (see chapter twenty).

It is also necessary to measure the intensity of radiation doses in working areas, so that work can be planned and monitored to avoid undue exposure. Semiconductors and scintillators, as well as geiger counters and ionization chambers, have been applied to measure radiation fields from beta and gamma sources. Staff in dosimetric research developed a series of different kinds of instruments. These included hand and foot monitors, which quickly verified whether or not a worker was contaminated with radioactive materials. These radiation protection instruments are excellent and have served their purposes within AECL very well.

The maximum permissible uptake of plutonium by workers is too small to be measured directly by bioassay or *in vivo* monitoring. Protection of workers is achieved by area-monitoring instruments. In any process dealing with plutonium, it is necessary to measure very small concentrations of plutonium in air containing naturally occurring radon. The problem has been solved using semiconductor detectors that can distinguish the differences in energy between alpha particles emitted by plutonium and those from radon and its progeny. This type of instrument has enabled the operators to demonstrate compliance with regulatory requirements on annual limits on intake of plutonium by workers.

Internal contamination by radioactive substances is a potential risk for some AECL employees, because of their work. It is the function of internal dosimetry to estimate doses to organs and tissues of individuals following intakes, or suspected intakes, of radioactive materials, and to ensure that these doses are kept to a minimum. It is also necessary to carry out research and development in internal dosimetry, monitoring, and modelling to improve internal dose estimation procedures for radionuclides associated with present and proposed nuclear programs in AECL (see figure 3.2).

One of the most sensitive and useful methods of screening employees for an intake of radioactive materials by inhalation, ingestion or through the skin is radiochemical analysis of excreta (bioassay), particularly of urine. Urine analysis is commonly used for the detection of tritiated water, strontium-90, radiocesiums and other radioactive contaminants, as needed.

When workers were internally contaminated by inhalation of insoluble particles of relatively pure carbon-14 during the retubing at Pickering, the residue of ashes from fecal samples from the workers was analyzed for carbon-14 content. Studying the excretion patterns in workers of the inhaled insoluble carbon-14 material, researchers at Chalk River, together with Ontario Hydro, quickly established a metabolic model of the behaviour of this hitherto unstudied material. The model allowed for the estimation of doses to workers who had been exposed to this source. All doses were within permissible limits.

In vivo monitoring by a whole body counter is required to complement bioassay monitoring and it is a very useful part of the program for the radiation protection of workers. It provides a measurement of radionuclides in organs and tissues of individuals by determining externally the number and energies of photons (gamma and X rays) that escape from the body; this requires efficient photon detectors and good energy resolution. The background counting rate should be as low as possible and when a special facility was relocated and rebuilt on the Chalk River site, but remote from radioactive materials released at the Chalk River laboratories, materials for construction were selected on the basis of their low background levels. Several models of artificial humans called phantoms are used for calibration purposes. Workers were monitored in the whole-body counters to detect the presence or absence of contamination from internally deposited radionuclides.

Metabolic modelling, as it relates to internal dosimetry, is the establishment of a mathematical model that describes the uptake of a radionuclide by organs and tissues, and its subsequent retention and excretion. Once the amount of radioactivity in the organs and tissues of an individual is known from bioassay or *in vivo* monitoring, the resulting radiation dose can be calculated from these metabolic models. Chalk River researchers have made major contributions to international development of a lung model and developed a computerized program known as GENMOD for use in the calculation of doses from internally deposited radionuclides.

OPERATIONAL RADIATION PROTECTION

Operational radiation protection, at times called operational health physics, is a very important topic in nuclear establishments and is devoted to ensuring the safety of workers. Radiation fields and contamination levels in working areas must be determined and records of radiation exposures accumulated in previous work must be available. Protective clothing ranges from simple gloves (to limit hand contamination) to full protective suits, possibly provided with supplies of clean air. Radiation-protection personnel provide valuable advice to operating staff who bear responsibility for safety. Consultations between radiation-protection personnel and operating staff are needed in the planning of work, so that exposures can be minimized (in other words, kept below individual dose constraints for the site). Most tasks will be governed by attention to the time spent on a task, the distance from the worker to the source, and the use of shielding to limit direct radiation exposure. Careful judgments have to be made to optimize procedures so that the total dose will be minimized during the task, and to ensure that individual doses will be as low as possible. In order to comply with dose limits or to reduce individual doses, time of exposure may be reduced by work-sharing. However, work-sharing does not decrease the collective or total occupational dose; in fact, the total collective dose may increase if work-sharing involves less experienced people who take longer to perform a given task. Radiation-protection personnel also have an extremely important role in bringing operational problems to the attention of designers of instruments and facilities. Important contributions are made to the design of the plant, in the layout of major components, and to facilitate maintenance work so that the plant can be operated safely with the least amount of radiation exposure.

Operational radiation-protection personnel are key players in the implementation of the ALARA process. In many areas Chalk River staff practiced the principle of ALARA long before it was formally recommended by the ICRP.

Operational radiation protection in AECL Research has for many years been combined with the more conventional aspects

of industrial safety. The objectives in considering all occupational risks (rather than radiation risks alone) are to reduce the total health risk from all causes and to use funds spent for health protection as effectively as possible.

An examination of annual radiation exposures over the years provides an indication of the good control in AECL, as a result of sound operational radiation protection practices. The average annual dose of Chalk River staff was about 7.5 mSv in 1958, which gradually decreased to about 1.4 mSv in 1989 (the legal limit was 50 mSv per year throughout this time).

With careful planning, practice with mock-ups and rehearsals emphasizing effective use of time, distance and shielding, it was possible to remove and replace components from reactors or hot cells even in very high radiation fields. Thus the collective dose for the replacement of the NRX calandria (1970) was 2.7 person-Sv, while that for the NRU replacement (1972) was 3.6 person-Sv. The replacement of a cracked leaded window with a new one, in the hot cells, took only 0.01 person-Sv.

The medical surveillance of radiation workers in AECL has always been carried out by physicians and nurses. Medically qualified experts in radiation protection may be designated as "medical advisors to the Atomic Energy Control Board" (see the Atomic Energy Control Regulations). Few specific diseases or medical conditions render a worker unfit to be an atomic radiation worker. Determination of fitness is made on the basis of purely occupational or medical issues; i.e., fitness to perform the duties demanded under circumstances that require the use of respiratory protection or use of plastic protective suits in high heat and humidity. Clinical assessment of potential and real overexposures of workers to radiation is carefully carried out. The low frequency of acute radiation effects in AECL workers is another indication of good safety practices. Since 1944, there has occurred only one serious burn of the fingers, when a worker inadvertently picked up a heavily irradiated capsule that had dropped on the floor from the reactor core. Two other workers were found to have reddening of a small area of their skin because of irradiation from tiny particles of radioactive materials. Skin burns of the fingers of two other workers resulted from overexposure of the skin to soft X rays. Two workers developed malignancies, possibly induced by radiation; one was a case of malignant lymphoma and the other was a case of chronic myelogenous leukemia. Through Labour Canada, AECL reported these two cases for review by the Workers' Compensation Board of Ontario. The estates of these workers are being compensated for the presumed injuries to the workers.

Since the forties, when Chalk River produced radionuclides such as phosphorus-32, carbon-14, strontium-90 and cesium-137 for clinical use and for exploitation as tracers in biological experiments, staff at Chalk River have been involved in the training of university, government and military personnel in the safe handling of these radionuclides. AECL staff are still engaged in this important educational role, because of their expertise in radiation protection. Health-sciences personnel host an annual one-week radiation-protection course at Chalk River. The course reviews basic radiation units and dosimetric concepts, biological and medical effects of radiation, and the latest risk coefficients for fatalities and serious hereditary effects. Environmental pathways for the movement of radionuclides are discussed and the relationship to waste management is noted. Selected topics in operational radiation protection are presented, with passing reference to conventional safety practices. The lectures are interspersed with tours of facilities and demonstrations of important health physics procedures. In addition to AECL lecturers, outside speakers (Control Board, Environment Canada, Health and Welfare) are featured. About twenty to forty participants from all parts of Canada, representing government, military, universities and utilities, attend. Obviously, a one-week course does not make instant experts of the participants, but it does permit them to hear and meet outstanding Canadian experts in this field, and to learn important concepts and topics in radiation protection. Feedback from the participants over the years has indicated that the course is highly successful.

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Chapter Four

Nuclear-Reactor Safety Principles

F.C. BOYD

INTRODUCTION

Despite the many safety devices incorporated in the NRX research reactor, which started operation in 1947, NRX suffered a serious accident in December 1952, as chapter one described. Although there were no known injuries, that early accident served as a stimulus for reviews, analyses and discussions about the safety of nuclear reactors. The result was a distinctive Canadian approach to nuclear-reactor safety, the evolution of which is the subject of this chapter. Through the application of this approach, Canadian nuclear-power plants and research reactors have an excellent safety record – no member of the public has been exposed to radiation resulting from a nuclear reactor in excess of regulatory limits, and no worker has been killed or seriously injured due to a reactor accident.

BACKGROUND

All forms of energy present hazards. For nuclear energy, the particular hazard comes from ionizing radiation. As a consequence, the science and practice of radiation protection has been highly developed.

In nuclear reactors, radiation is produced by the fission process and by the decay of radioactive fission and activation products that result from reactor operation. The fission products that result from the splitting or fissioning of the uranium are almost all radioactive, but largely remain inside the uranium fuel. They constitute the vast majority of radioactive material in a nuclear reactor. Activation products are essentially small bits of corrosion in the coolant system that become radioactive as they pass through the reactor core.

A major safety challenge in the design and operation of reactors is to safely control the chain reaction. Although nuclear reactors cannot explode like an atomic bomb, they can increase their power extremely quickly and can experience overheating of their fuel. The result could be the release of a large quantity of radioactive fission products that could be very hazardous.

Because the potential hazard was recognized at the beginning of the nuclear program, there has always been an emphasis on prior analysis of possible failures. The term "nuclear safety", or, more explicitly, "reactor safety", is generally applied to the work aimed at preventing accidents in nuclear reactors, or to mitigating their consequences.

While reactor safety was not identified as a distinct topic in the work of the Montreal Laboratory when it was set up in 1942, the goal of a safe and controlled reactor was central to its work. This was reflected in the design of the reactor, which became known as NRX. Many studies and analyses were conducted on

safety-related topics, such as shielding against the radiation that would be produced in the fission process, the dynamic behaviour of the reactor, heat transfer from the uranium fuel, and atmospheric dispersion of radioactive material.

The choice of a site for the reactor and laboratory also evoked a number of safety issues. Security (in the sense of secrecy) was a factor in choosing a remote location, but the safety advantage of having little or no population for kilometres around was certainly considered by those involved in the choice. As G. Laurence, senior Canadian staff member of the Montreal Laboratory, later wrote: "Isolation was desirable to avoid supposed hazards to population from possible explosion or release of radioactive dust into the atmosphere, and to simplify control of secrecy."[1]

Shortly after the Chalk River site was chosen, in August 1944, smoke tests were conducted to investigate the potential dispersion of radioactive material that might be released; these were the first of many experiments conducted specifically to provide information necessary to analyze the safety of nuclear activities.

SAFETY OF NUCLEAR REACTORS

The primary hazard presented by a nuclear reactor is the very large amount of radioactive products created by the fission process. These fission products are normally contained within the uranium fuel. Although radioactive material is produced by neutron activation, such as radioactive corrosion products in the coolant, and plutonium is produced in the uranium fuel, the fission products constitute about 98 percent of the total radioactivity in a typical reactor.

If the fuel becomes overheated and its sheathing breaks, the fission products can escape from the fuel assembly. Safety analyses of nuclear reactors, therefore, look at all possibilities of fuel damage. In particular, they examine all potential situations that could lead to the fuel becoming overheated, either by too much energy (too many fissions) being created in the fuel, or inadequate cooling of the fuel. The two major potential initiating conditions are a loss of coolant and a loss of control, leading to an uncontrolled increase in the reactor power.

Fission products generate heat even after the fission process (reaction) is stopped. Therefore, the fuel must continue to be cooled after the reactor is shut down. This is most critical when the power density of the fuel is high; i.e., when the fuel generates a large amount of energy per unit volume, as is typical in reactors used to produce electricity.

The original NRX reactor design included two ways of controlling the fission process – changing the height of the heavy-water moderator in the reactor tank or calandria (changing the size of the reactor), and inserting or removing a neutron-absorbing rod. To be able to stop the reaction at any time, shut-off rods were provided, initially eighteen, later twelve. These boron-carbide-filled steel rods were designed to drop into the reactor, accelerated by air pressure to increase their speed over that from gravity alone. The dropping of the shut-off rods could be activated automatically by signals from several measuring devices as well as manually. As a slow backup, the heavy-water moderator could be drained from the calandria.

That safety was high in the minds of the designers can be inferred from a comment by W.B. Lewis (the director of the Chalk River laboratory) that "the reactor has been characterized as having 900 devices for shutting it down but only one for starting it up."

The NRX accident of 12 December 1952 (described in chapter one) precipitated several reviews and, for Laurence, was the beginning of a preoccupation with reactor safety that lasted the rest of his life. Others were also prompted to examine their approach to reactor-safety design, with particular concern for the systems that control and shut down the reactor. Those who were working on the design of the control and shutdown systems for the 200 MW NRU reactor (construction approved in December 1950) decided to invoke the concepts of redundancy and coincidence through the use of multiple-channel systems.

If there is only one channel (or string of instruments and devices) in a system, any significant failure will disable it. With three (or four) parallel channels arranged such that two out of

[1] Eggleston, *Canada's Nuclear Story*, 125

the three (or four) are required to respond to effect a control or shutdown action, the reliability is significantly increased and there is the added bonus that individual channels can be tested while the reactor is in operation.

A further conclusion was that it was desirable to separate the protective (shutdown) system from the control system, so that failures in the more complex control system would not impair the ability to shut down. Because the NRU reactor was too far along in design and construction, this concept of separation was not incorporated in that reactor, but it has become a basic requirement for all subsequent reactors in Canada.

All shutdown systems of Canadian nuclear power reactors have triplicated channels and are physically and functionally separate from the control systems.

Another concept that emerged was that of a safety bank. The NRX reactor accident led to the recognition that a shutdown reactor was not necessarily safe. A safety bank involves resetting some of the protective devices (e.g., shut-off rods) after a shutdown, so that if any maintenance activity or other work increases the reactivity of the reactor, the rods will fall and prevent a possible accident.

With NRU well under way, attention turned to the power reactors that were being designed or proposed. An important difference between the research reactors (NRX and NRU) and the power reactors is that the latter have high-temperature, high-pressure water-coolant systems to transfer the heat energy produced in the fuel to the electricity-generating systems. From a safety viewpoint this introduces a major concern about a loss of cooling or loss of coolant. If a coolant pipe breaks, the loss of the coolant will deprive the fuel of cooling and the escaping hot, pressurized, water will flash into steam, pressurize the enclosing building and provide a means to carry radioactive material to the surrounding environment.

In 1957 E. Siddall, one of the NRU designers, wrote a seminal paper examining the objective of reactor safety, noting that there was a tendency to demand more and more safety measures without any clear evaluation of their benefit. A study issued by the United States Atomic Energy Commission that year (WASH 740) had caused considerable reaction with its prediction of disastrous consequences if a large fraction of the fission products in the fuel of a large (200 MWe) nuclear-power reactor were released to the atmosphere. This led to the demand for more and more safety measures.

Siddall proposed that a basic criterion for judging the safety of an activity should be the number of deaths per unit time arising from that activity. He observed that society implicitly accepted the risk of many activities, even though the accidental death rate from them is high relative to the general probability – private motoring being the prime example.

From studies of other industrial activities producing output of comparable value, in particular coal-fired electricity-generating plants, Siddall suggested that a death rate for a large nuclear power plant (200 MWe) of 0.17 deaths per year, or about one death per six years, should be acceptable. Although these numbers were never adopted, the concept of an acceptable risk, expressed numerically, has been a fundamental part of Canadian nuclear safety philosophy.

At the same time (1957), Laurence, who was then a senior director at the Chalk River laboratories, presented the first of what would become a series of papers on reactor safety. This was not long after he had been appointed the first chairman of the Reactor Safety Advisory Committee (RSAC) of the Atomic Energy Control Board. At that time the Control Board had only one scientific staff member, and chose the advisory committee approach to deal with the technical safety aspects of reactors then being proposed for operation outside of the Chalk River laboratory: a research reactor at McMaster University (Hamilton, Ontario) and the 20 MWe NPD reactor being designed by Canadian General Electric for AECL and Ontario Hydro. Over half of the original members of the RSAC were senior members of AECL's Chalk River laboratories.

Aware of Siddall's concepts, the designers of NPD proposed, in the preliminary "Hazards Report" for NPD submitted to the Control Board's RSAC in 1958, a basic objective of less than one death per 100 years for the plant, which the committee essentially accepted. This was the first formal "hazard", or, as they would be called later, "safety" report prepared in Canada for a nuclear plant.

The protective system of NPD was separated from the control system and employed the two-out-of-three concept. The design included continuous monitoring and inter-comparison of the three protective system channels and provided for testing, from input signal to shutdown device, during operation.

About two years later, Laurence proposed, as a reference value, an upper limit for the likelihood of a "major accident" of one per 100,000 reactor-years, on the premise that it was unlikely any accident would lead to more than 1,000 deaths. He also endorsed the multiple-channel approach for safety systems, with emphasis on testing to demonstrate failure rates and, thereby, availability. Such a target could be achieved, he argued, with very practical reliability criteria if there were adequate separation between, and independence of, the operating systems and the various safety systems.

Laurence and the RSAC conceived of the reactor design having three divisions: the "process" systems (the essential operating part of the plant); "protective devices" (e.g., to shut down the reactor if there are problems with the process systems); and "containment provisions" (to prevent any radioactive material that might escape from the process systems from being released to the environment). If each of these divisions were properly designed for their function, it would take a failure of all three before there could be a serious release of radioactive material to the environment. Further, if these divisions were sufficiently separate and independent, the frequency of failure of the process systems and the unreliability (or unavailability) of the protective devices and containment provisions could be combined to give the likelihood of a significant release affecting the public. This meant that the reliability requirements for individual systems could be practicable while still achieving a very low risk to the public.

On this basis, Laurence proposed criteria for the maximum frequency of serious failures of the process systems and unavailability of the safety systems. He argued that these standards should not only be easy to achieve but that they could be demonstrated by testing. In particular, the protective shutdown systems, if designed in the two-out-of-three or two-out-of-four arrangements, could be tested with the reactor operating.

Some of these concepts were expressed in the NPD design even though that design was completed before the concepts were formally presented in technical papers. Similarly, in the full-scale (200 MWe) Douglas Point NGS, then under way, the protective shutdown system was separate from and independent of the regulating control system and in addition the reactor was housed in a building specifically designed for containment.

During the late fifties and sixties, the designs and safety philosophy evolved together in a somewhat synergistic fashion. Designers met frequently with the RSAC and its growing technical staff to propose, debate, and eventually agree on various safety concepts. The meetings were often spirited; the designers and the regulators greatly respected each other.

With the confidence arising from the initial operation of the NPD station and the completion of the Douglas Point design, Ontario Hydro proposed building a large multi-unit station (four units of 500 MWe) near Toronto (Pickering NGS). The RSAC was not convinced that the Douglas Point containment design would be adequate for this plant. In response, the designers developed a unique arrangement in which the containment buildings surrounding each unit were attached to a large vacuum building. The concept of this vacuum building arrangement was that the steam and pressure resulting from a loss-of-coolant-accident (LOCA) would be sucked out into the vacuum building and the entire system would remain at atmospheric or sub-atmospheric pressure. Consequently, there would be no pressure to push out any radioactive material that might be released from the reactor systems due to the LOCA. After much debate, this concept was approved and has been used on all of Ontario Hydro's multi-unit nuclear power stations as a cost-effective containment arrangement.

For a LOCA to occur, the assumption is that a major failure (break) of a large pipe in the main reactor cooling system (sometimes called the heat transfer system) would allow the hot, pressurized water to escape. The LOCA had come to be regarded (and still is) as the most serious type of reactor accident, since it has the potential to cause failure of the fuel (due to overheating), which could release large quantities of radioactive fission products; at the same time, the steam produced by the escaping hot

water of the cooling system would pressurize the containment building.

Canada's two single-unit nuclear power plants (Point Lepreau and Gentilly-2) each have a containment building designed to hold the highest pressure that would result from a LOCA. This design has also been approved by the Control Board as being fully adequate.

Basic reactor-safety concepts continued to evolve. By 1972 (after Laurence's retirement as president of the Control Board, to which he was appointed in 1962), the RSAC had adopted an approach in which the reactor plant was conceived as having process systems and several special safety systems – each of which had a specific safety function, such as shutting down the reactor, providing emergency cooling of the uranium fuel or preventing the dispersion of radioactive material. The special safety systems were to be separate and independent of each other and of the process systems.

As performance requirements for the designers, maximum values of radiation dose that might be received by a member of the public (using conservative assumptions and calculation methods) were set for single failures of a process system (such as a large pipe failure) and for dual accidents where such a process-system failure was associated with complete failure of one of the special safety systems.

A maximum frequency of serious failures of the process systems was set at one per three years, and a maximum unavailability of each special safety system was set at one per 1,000 years. (A serious failure of a process system was defined as one that could lead to the release of radioactive material in the absence of safety-system operation.) Assuming adequate independence, the likelihood of a dual accident should, therefore, be less than one in 3,000 per year. Following this approach, a major release could only occur if there were a serious process failure and complete failure of two independent special safety systems, which, if the requirements are met, would have a likelihood of less than one in 3,000,000 per year.

The reference dose value for an individual for a dual accident was chosen as that proposed by the Medical Research Council of the United Kingdom as tolerable to be received once in a lifetime, even though the postulated likelihood of a dual accident was one in 3,000 reactor years. These single-failure/dual-failure criteria are still used by the Control Board in judging designs for nuclear power plants, although additional requirements are now applied.

At the end of the sixties, the RSAC was faced with a challenge by the designers of the then-proposed Bruce "A" nuclear generating station. The proponents stated that they could not design and analyze a practical containment to meet the dual-failure criteria if they had to assume a runaway accident; i.e., an uncontrolled increase in power that could result from a failure of the control system accompanied by failure of the shutdown system. They proposed, therefore, to provide a second shutdown system specifically designed to guard against a power runaway.

After considerable debate over the requirements, the RSAC finally adopted the concept of a second shutdown system, but only if it was as capable as the first – separate and independent of the first shutdown system and of the control system – and used a different physical process. If all of these conditions were met, each shutdown system would be regarded as a separate special safety system and the single-failure/dual-failure criteria would apply; i.e., the reference dose limits must be shown to be met for any combination of a process system failure together with failure of any special safety system. All Canadian nuclear power plants since Bruce "A" have two separate, independent and different shutdown systems. This Canadian approach preceded by over a decade a long, protracted argument in the United States over what was called "anticipated transient without scram" – a power runaway without shutdown.

A major feature of the single-failure/dual-failure concept and the associated reference dose limits was that these were performance criteria, not design requirements, thus leaving the designers considerable flexibility. The major design requirement was a very general one: the need for separation and independence of the safety systems. This emphasis on objective was in sharp contrast to the detailed design rules issued by the United States Nuclear Regulatory Commission (USNRC), which succeeded the USAEC as the nuclear regulatory body in that country.

Members of AECL's scientific staff played a major role in the RSAC and in the development of Canadian nuclear safety philosophy in general, and most of the scientific knowledge and technical data that provides the basis for safety decisions has been generated by AECL's research groups (see chapter fifteen).

INTERNATIONAL

Canadian nuclear scientists and engineers have played an active role in the international field of nuclear safety.

The risk-based concepts outlined above were presented by Laurence and others to various international conferences and meetings as early as 1958. While there was widespread support among professionals involved with reactor safety, most national nuclear regulatory agencies followed the style of the USNRC in specifying prescriptive, detailed design requirements. However, by the early nineties even the USNRC was seriously examining risk-based regulations.

In the seventies, the international nuclear community began speaking of reactor safety in terms of defence-in-depth. This refers primarily to the many physical barriers between the radioactive fission products in the fuel and the environment, such as the fuel sheathing, coolant-system piping, and containment buildings. Other aspects, such as the exclusion zone around nuclear power plants, and the extensive analyses and quality-assurance programs, are also sometimes invoked as part of this approach. The defence-in-depth concept is often used when describing the safety of nuclear reactors to the public. The approach followed by Canada, especially that of separate safety systems, provided very effective defence-in-depth long before the concept became internationally popular.

SAFETY EXPERIMENTS AND RESEARCH

Most of the scientific and technical information needed for the safety design and evaluation of a nuclear reactor is the same as, or similar to, that used in the design of the basic plant; e.g., fuel behaviour, effects of radiation on materials, physics of the nuclear reaction. In some cases, the safety analysis raises questions beyond the available knowledge, or the safety design involves devices or techniques for which there is little or no precedent. For such situations special safety experiments have been conducted, particularly for the early CANDU nuclear power plants.

As the nuclear power program matured, most of the obvious questions had been addressed. Attention turned to the more esoteric safety questions involving conditions having a very low probability. To address them, a broad safety-research program evolved in the early seventies.

Chapter fifteen describes both the early, specific experiments and the later, comprehensive safety-research program in more detail.

OTHER FACILITIES

In recent years there has been renewed interest in small reactors, which has led to studies, experiments and proposed criteria specific to that class of reactor. Canadian scientists from AECL and staff of the Control Board have been in the forefront of international developments in this area.

SUMMARY

The safety of nuclear activities has been a high priority since the beginning of Canada's nuclear program. Special attention has been paid to the safety of nuclear reactors, especially the large CANDU reactors used in Canadian nuclear power plants.

A set of conservative criteria was developed over the first decade or so of the CANDU program, with the objective of minimizing the risk of a serious accident in a nuclear power plant. These criteria evolved out of a critical review of the NRX accident in 1952, and therefore reflect a unique Canadian approach. They emphasize separate and independent safety systems that are testable to achieve a very low likelihood of a serious accident. The concepts involved in this approach are now being adopted in many other countries with nuclear-power programs.

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RADIOISOTOPES

Chapter Five

Radioisotopes

B. ULLYETT

BEGINNINGS

Radionuclides that do not occur naturally in nature and are produced intentionally through nuclear reactions are called artificial radioisotopes. The first fruits of the Canadian nuclear program were realized in the production of artificial radioisotopes in the NRX reactor, and in their subsequent worldwide use in the medical and industrial fields.

These developments came just half a century after the discovery of natural radioactivity, when Henri Becquerel, in 1896, motivated by Wilhelm Röentgen's discovery of X rays in 1895, found that uranium emitted a penetrating radiation, referred to at the time as Becquerel rays. In 1898 Marie and Pierre Curie discovered two new elements in uranium ore, pitchblende, which they named polonium and radium. Radium soon came into great demand for cancer treatment in humans. It commanded a very high price; e.g., $75,000 per gram (1930 dollars).

Artificial radioactivity was discovered in 1934 by Irène, the daughter of the Curies, and her husband, Frédéric Joliot. They were studying the positrons produced by bombarding aluminum with particles from polonium. They found that the positron emission did not stop when the polonium source was removed, but decayed with a 2.5-minute half-life. This discovery prompted Enrico Fermi to examine whether neutrons could produce artificial radioactivity. He found, also in 1934, that slowed-down neutrons were particularly effective. By the end of 1935, about one hundred artificial radioisotopes had been identified.

With the advent of nuclear reactors, enormous quantities of radioisotopes could be produced, and a new industry was born. As just one indicator of the scale of this evolution, it was possible to replace the radioactivity of one gram of radium, i.e., 3.7×10^{10} atomic disintegrations per second, at $75,000 (1930 dollars)

with sufficient cobalt-60 to yield the same amount of disintegrations per second at $1 (1977 dollars).

ARTIFICIAL RADIOISOTOPES FROM NRX

By 1944, subsequent to the 1943 World War II Quebec conference of the heads of Allied governments, the decision was made that the Montreal Laboratory should design, build and operate a 10 MW heavy-water, natural-uranium-fueled nuclear reactor to produce plutonium and to provide neutrons for research. The reactor was to be named National Research Experimental (NRX). The importance of radioisotopes to medicine had already been recognized (in particular cobalt-60, which had been identified by 1945 by the scientific community as a possible source of gamma radiation for the treatment of cancer tumours without invasion of the body), as well as their importance to agriculture and industry. NRX was immediately seen as an excellent means for radioisotope production and the reactor was designed with this in mind. Self-serve entry/exit tubes were designed into the reactor shielding walls to permit on-power sample entry for neutron irradiation and subsequent removal.

Before the NRX startup in 1947, there was a joint effort by NRC research chemistry and the isotope production branch (IPB) at Chalk River to develop methods to make and purify those radioisotopes that were expected to come into demand. Within two months of NRX startup, megabecquerel (MBq) amounts of phosphorus-32, sulphur-35 and iodine-131, and kilobecquerel (kBq) amounts of carbon-14, had been produced. Within three months of startup (31 October 1947), the first radioisotope shipment was made to the University of Saskatchewan, and within five months shipments had been made to Canadian university researchers from coast to coast.

With the chemical processing of many radioisotopes in rapidly increasing amounts, the IPB staff had to learn quickly to work safely with radioactive materials. Special processing fume hoods were designed, some with raised floor platforms in front, to increase the distance from hood working levels to the laboratory floor. These platforms were later lowered to floor level, with additional in-hood shielding provided by the design of interlocking modules of lead bricks to permit various shielding configurations.

After two years of NRX operation, it was obvious that Chalk River could produce higher radioactive specific activities (radioactivity of a specific radioisotope per unit mass of isotope target) than could Oak Ridge (United States), or Harwell (United Kingdom), due to the factor-of-ten higher neutron flux available with NRX. With the increasing amounts of radioactivity being produced as reactor irradiation experience grew, the IPB laboratory layout required the marking of floor areas with colour bands and circles, to alert technical staff traversing from a lower-activity area to a higher one. By 1950, the quantities of hydrogen-3, carbon-14, sulfur-35, phosphorus-32, iodine-131 and others were made available to researchers at Chalk River, to Canadian university research centres and to pharmaceutical firms studying the uses of these new products.

By 1949, it became increasingly apparent that cobalt-60 was coming into strong demand as a radioactive source[1] for use in the treatment of deep-seated cancer tumours. Chalk River arranged to sell this radioisotope through the commercial products division of Eldorado Mining & Refining Ltd. (Port Hope, Ontario), as a possible substitute for radium in tumour treatments and in radiographic work.

COMMERCIAL PRODUCTS DIVISION OF ELDORADO

At the time of its agreement (1949) to sell Chalk River's cobalt-60, Eldorado's commercial products division (CPD) was facing slumping profits from its radium operations, because of competition from other radium suppliers (United Kingdom, United States and France). There was also the gradual recognition that nuclear-reactor produced radioisotopes would eventually displace radium in the medical field.

By 1950, NRX had produced sufficient high-activity cobalt-60 in wafers of metallic cobalt to permit fabrication of cobalt-60

[1] Radioactive material encased in a sturdy capsule, without leakage, is defined, for this chapter, as a radioactive source.

sources. With this availability, CPD saw a marketing opportunity for the sale to cancer clinics of medical treatment units containing a cobalt-60 source.[2] Necessary funding was obtained for a CPD design, and a contract awarded to Canadian Vickers, of Montreal, to build the first unit.

This first CPD teletherapy unit, called the Eldorado A, comprised an encased, lead-filled shielding head mounted on a fixed vertical column that could be raised, lowered or angled in several directions. This unit was shipped to the London Cancer Clinic in London, Ontario. Another unit of different design was being constructed, at about the same time, by the Saskatoon Cancer Clinic for similar use. Within a short time of each other,[3] the two clinics began using their respective units for cancer treatment.

These were the first two cobalt-60 teletherapy cancer treatment units to operate in the world. Concerned about product quality, CPD decided to undertake their own manufacture of additional Eldorado A's. By the time that it was superseded by improved models, about fifty had been built and installed around the world by CPD staff. These units sold in a price ranging from $25-$30,000 ($US).

The head of a renowned cancer clinic in Paris who purchased one of the early Eldorado A units has kept the now-retired unit in his clinic as a museum piece. He states that, even today, the Eldorado A remains among the best and simplest of cancer treatment units.

By late 1952, the first rotational cobalt-60 teletherapy unit had been designed by CPD based upon specifications provided by a physicist with the Francis Delafield Hospital of New York. It was called the Theratron Model B. A design patent was awarded and royalty payments were made to the hospital physicist for each unit sold by CPD until 1973. In this type of unit, the cobalt-60 source (fixed within the shielded head) was rotated over an arc of a circle with the beam collimator aimed at the centre. The patient couch, an integral part of the unit, could be adjusted to permit patient location so that the tumour was at the centre of rotation and would alone receive the full radiation dosage. The source to centre of rotation was 75 centimetres. Junior Model B's having a smaller cobalt-60 capacity and a 55-centimetre treatment distance were also designed, and became popular at small clinics.

The Model B Theratron sold at a price that was double that of the Eldorado A units. The first 'B' unit was installed at Francis Delafield Hospital in 1953. The Model B was well accepted and complemented the Eldorado A in many clinics. By the late eighties, vertical units were no longer marketed. CPD later trademarked the name of the cobalt-60 teletherapy units to theratron units, which it now uses exclusively. (From this point forward in this chapter, theratron is used and specifically denotes a cobalt-60 teletherapy unit.) From 1946 until 1952, with the new radioisotope products to sell along with radium products, and the development of a first-class machine shop, CPD staff grew to a total of eighty-four, most being located in Ottawa.

COMMERCIAL PRODUCTS DIVISION OF AECL

CPD was transferred in August 1952 from Eldorado to Atomic Energy of Canada Ltd. The transfer included operations for the sales and production of radium, the new radioisotope products

[2] These units were called teletherapy units, to indicate radiation-beam treatment of deep-seated body tumours from a distance, and to identify the cobalt-60 source as a teletherapy source.

[3] For many years, rivalry existed concerning who had the first cobalt-60 unit, and who gave the first patient treatment. The following information was obtained from Miss Sylvia Fedoruk, who assisted at Saskatoon at the time (she later became Lieutenant-Governor of Saskatchewan), and Miss Elaine Marshall in London, as reported in *The Cobalt-60 Story* (see bibliography):

- Official opening of cobalt unit in Saskatoon by Dean Cronkite was 23 October 1951.
- First patient treatment on cobalt-60 unit in Saskatoon, by Dr. T.A. Watson, was 8 November 1951.
- Official opening of cobalt unit in London was 11 November 1951.
- First patient treatment on cobalt-60 unit in London, by Dr. I. Smith, was 27 October 1951. (Note: Opening date in London was after first patient treatment – Ed.)

and the manufacturing machine shop. Thus, the history of radioisotopes in Canada and their worldwide use, is, in effect, the history of CPD, one of the oldest, and for a long time the least known of AECL's divisions. CPD became simply AECL Commercial Products (CP) by 1968, and AECL Radiochemical Company (RCC) by 1978. For consistency, the name CPD is used followed by (RCC) when appropriate.

Although this new AECL division was to be profit-oriented, immediate profitability was not certain, and in fact there was no assurance of the operation ever making profits. However, the record indicates that in spite of some bad years, there were a few years of modest profitability.

CPD grew rapidly after becoming part of AECL. It was more able to consult with the nuclear physicists at Chalk River and to obtain services, such as glassblowing, radioisotope measurements, design concepts for laboratory equipment, and dosimetry from several of the AECL divisions. Radioisotope markets between nations gradually opened up. CPD established its own R&D branch to explore uses for radioisotopes. The design branch and machine shop at Ottawa expanded rapidly to manufacture theratrons, which were in demand following the London and Saskatoon installations. The small CPD group at Chalk River required increased quantities of radioisotope products from the IPB to meet orders, as well as a variety of newer radioisotopes such as calcium-45, chromium-52, gold-198, iron-59, mercury-203, phosphorus-32, yttrium-90, zinc-65, and the fission products strontium-90 and cesium-137. Several long-term contracts were obtained by CPD for some of these, which resulted in the scheduling of routine processes by the IPB.

One Chalk River official observed at the time that the accomplishments of such a small radioisotope production group in crowded facilities were amazing when compared with the new giants of the day. Oak Ridge National Laboratory (ORNL), in the United States, employed dozens of professional staff and built much larger radioisotope-separation laboratories that required isolation in individual buildings. They were required to process as much as ten times more radioactive target weights, because their reactor fluxes were a factor of ten lower than that of NRX. The Radiochemical Centre Ltd., United Kingdom, and Commissariat à l'Énergie Atomique, France, were each in the same position, as their reactor fluxes were about the same as those of ORNL. Nevertheless, the market for radioisotopes developed rapidly.

CPD staff reached 150 by 1954. Business activities were squeezed into rented basement quarters of an Ottawa store. Research and development operations and the machine shop were crammed into other leased buildings throughout the city. To handle radioactive cobalt-60 for the manufacture of ^{60}Co sources processing facilities were needed. The radium-source production laboratories at Port Hope needed expensive upgrading. The expanding CPD group at Chalk River was crammed amid IPB operations. More space was needed to accommodate the rapidly expanding business. CPD had the understanding that, once becoming an AECL division, new facilities were to be considered to consolidate all operations under one roof.

The CPD manufacturing shop could not wait for these. In late 1953, it moved temporarily into a warehouse on Laperrière Avenue in Ottawa. In 1954, new facilities, designed to accommodate the CPD Port Hope radium-source operations, the CPD radioisotope operations at CRL, the manufacturing shop and the scattered Ottawa groups, were begun in Tunney's Pasture in Ottawa, a federal building site situated close to the city core. The Ottawa choice for CPD was more suitable to commercial needs – such as Customs, transportation, and business contacts – than the more distant Chalk River. The new facilities, costing $1 million, were ready for occupancy by early 1955.

It was obvious that the allocated manufacturing shop space in the new facilities was only a fraction of what was required to meet the demand for fifty theratron units per year, and that number was ever-increasing.

CPD R&D had by then established that monitored, mega-rad dosages of cobalt-60 radiation to certain foods destroyed the enzyme action that was a cause of food spoilage, and left the food safe and palatable. Food-irradiation plants to use large quantities of cobalt-60 were being designed. These would require manufacture, complete assembly in the shop area, and testing before shipping. As a result, the manufacturing shop

elected to stay at the nearby Laperrière location, scheduling three daily shifts that operated two continuous production lines to produce the required theratrons.

When CPD began operations in the new Ottawa facilities, AECL decided that all costs associated with the supply of commercial radioisotopes would be fully accountable by CPD. However, irradiation services from the NRX and NRU reactors would be provided on a marginal-cost basis (see chapter twenty). In this way the viability of the business could be evaluated. During the period 1955-57, the IPB turned over all of its radioisotope production technology to CPD, in some cases providing new glassware and remote-handling facilities. The IPB subsequently ceased its radioisotopes operations at Chalk River.

In the new CPD laboratories, each radioisotope was processed in an isolated laboratory to prevent cross-contamination of products, as opposed to a common area as at Chalk River. A transfer cell was constructed to enable the remote delivery of sealed irradiated targets from shipping containers to specific laboratories. By 1957, CPD began to prepare all of its reactor targets and to chemically process them after irradiation. These included formerly IPB-produced carbon-14, calcium-45, iodine-131, phosphorus-32, and sulphur-35. The list expanded in time to provide approximately fifty such products, many as custom orders. This necessitated division of laboratory space and the locating of some radiochemical processes in the basement.

The radium laboratories constructed in the Ottawa laboratory were built in an isolated section of the building, with facilities designed to avoid contamination of work areas. This had been a constant problem at Eldorado. All radium was now handled within sealed, lead-shielded, cubic metric boxes of lucite, fitted with "ball-joint" manipulators mounted through lead-shielded walls. In-box equipment was articulated with flexible joints and designed to be disposable. Interior air from the boxes was exhausted first through in-box shielded, activated charcoal filters, followed by absolute particulate filters, prior to reaching the final absolute filters in the upper exhaust room. All in-box waste was deposited in shielded waste containers (pre-approved for shipment to CRL), via inter-box transfer units isolated from the in-box operations. A remotely operated drawer vault for holding radium inventory was built.

These facilities operated as clean areas. Exposure of personnel to contamination with possible high-radiation dosages from radium never became a serious problem again. Face masks, double-lined gloves, double shoe covers and full whites were no longer necessary for routine work, but were available from nearby emergency cupboards.

The first product CPD (Eldorado) offered for sale, in 1946, was radium salts enclosed in steel needles and plaques, for interstitial treatment of cancer sites. These products were scheduled for phasing out by 1959 as the use of cobalt-60 encased in needles began to grow.

Radium products that represented significant sales for CPD until the late fifties were neutron sources produced by the homogeneous mixture of radium with beryllium. These became useful as starter sources to provide an initial supply of neutrons for the startup of small nuclear reactors. The nuclear reactors that powered the first two American nuclear submarines, the NAUTILUS and the SKATE, used these starters. Another was used by the British navy. (These could not have been prepared prior to the development of the remotely automated tungsten-inert gas (t.i.g.) welder, described in the next section.) Similar sources, with less radium content, had been used earlier by oil-well explorers (loggers), in the exposure and examination of deeply located substrata to identify oil-bearing sites.

In the late fifties, oil-well logging interests (Wells Surveys Inc.) asked CPD to produce higher-quantity neutron-emitting sources from an actinium:beryllium mixture for oil-well exploration. The exercise to irradiate radium, separate the actinium-227, and fabricate the sources became far too expensive. After the preparation of a number of these sources, the program, being uneconomical, was discontinued.

With the declining interest in radium, all products were phased out throughout the sixties. Borrowed radium was returned. The program was terminated in 1970, and all remaining CPD stocks were given to China as a gift.

Summary of CPD Revenues, Profits (Losses) – 1947 to 1978

PERIOD	REVENUES ($ million)	PROFIT/(LOSS) ($ million)
1947-52	2.66	1.39
1953-59	11.25	(0.286)
1960-69	61.50	3.50
1970-74	53.55	(1.63)
1975-78	81.42	(2.77)

Data in the above table ceases at 1978, the year in which CPD was restructured to become RCC. From 1980 to 1990, annual revenues increased, exceeding $100 million by 1990.

PROCESSING HOT CELLS AT CHALK RIVER AND CPD

The CPD need for larger quantities of cobalt-60, both as high specific activity (HSA: >7.4×10^3 GBq/g (200 Ci/g)) for theratron sources and low specific activity (LSA: <1.8×10^3 GBq/g (50 Ci/g)) for large-scale food and medical irradiation plants, raised problems. The handling of such vast amounts of radioactivity following reactor removal was a concern. Also, Chalk River needed better shielded facilities to handle irradiated reactor fuel elements that were under test for the power-reactor program. To meet these needs, in 1957, Chalk River built a complex of high-radiation-capacity processing units equipped with remote operating devices. These were called 'Universal Cells' (see chapter two).

The increased requirement of HSA cobalt-60 was met by the Chalk River development of a more efficient target. The 1×1 mm pellets of nickel-plated cobalt-59, normally loaded in an annular ring of 3 mm within irradiation capsules holding 30 grams, was revised to the sealing of that weight in one layer within 10 cm square pure aluminum sandwiches. These were curved and tack welded together to form tubes 2.5 metres in length and entered into high-flux reactor locations. LSA cobalt-60 was produced in 2.5-cm-long cylinders of cobalt-59 sheathed in aluminum, stacked vertically on trays that were assembled one above the other, and irradiated within the lower-flux regions of each reactor. Following irradiation, the sandwiches and cylinders required chemical processing in the Universal Cells for removal of the aluminum and recovery of the cobalt-60. About 7.4×10^7 GBq (2×10^6 Ci) of cobalt-60 could be produced annually in the Chalk River reactors.

At the Tunney's Pasture building in Ottawa, CPD had the same need for such hot cells. Construction of two general-purpose cells at the Tunney's building were completed in late 1957. All cobalt-60 source fabrication was subsequently transferred from Chalk River to the CPD cells. The handling of exposed cobalt-60 in the cells at CPD would result in serious in-cell contamination. Rigorous decontamination of sources would be required. Source seals had been made by Chalk River with screw-down metal lids using lead gaskets or force-fit metal plugs, followed by decontamination before shipment to CPD.

To eliminate source contamination and any possibility of source leakage, an improved method of a higher integrity source seal was needed. Following a year of trial units, a remotely automated tungsten-inert gas welder was developed at CPD. It proved to be reliable and at once heralded the dawn of source sealing within a welded inner stainless-steel capsule, followed by ultrasound decontamination and subsequent sealing within an outer stainless-steel welded capsule. The concern regarding the leakage of radioactivity was eliminated. The first all-welded stainless-steel theratron source was No. S-246 in 1960, which encased 2.96×10^5 GBq (8,000 Ci) of cobalt-60, one of the largest therapy sources ever made by CPD. It was sent to a Puerto Rico clinic. This welding technique was ultimately adapted for the sealing of all metal-encased radioisotopic sources.

CPD BEGINS RELOCATION AT KANATA

By 1964, nearly one hundred cobalt-60 theratron units were being produced annually, exclusive of other products. The manufacturing plant on Laperrière Avenue in Ottawa became totally inadequate for such a large volume. Crowded conditions existed at the Tunney's Pasture building and the situation was only partially resolved when the sales department vacated to rented quarters. Relocation to larger facilities had to be considered again.

CPD purchased approximately eighty acres of crown land in Kanata, just west of Ottawa. Phase I consisted of the construction of the largest and most comprehensive machine shop and assembly area in eastern Ontario. It was completed by 1966, along with a five-storey administration building. (These facilities were expanded three times over the next fifteen years.) Sufficient land was reserved for the eventual relocation of the balance of CPD Tunney's Pasture operations.

As business grew, the storage of cobalt-60 inventory at the Tunney's Pasture laboratories caused problems. Bulk cobalt-60 was stored in three-tonne lead- shielding containers that were used for shipments between Chalk River and CPD. High temperatures on container surfaces (60°C), due to the heat of decay of the cobalt-60, raised concern that possible melting or shifting of internal lead could cause gaps in the shielding, bringing about a serious radiation hazard. The quantities of cobalt-60 stored in these containers was accordingly reduced. It was realized, however, that if the demand for cobalt-60 continued to grow, as seemed highly likely, the situation would become immeasurably worse. Subsequent examinations showed that the containers were, in fact, sound. Nevertheless, new cobalt-60 facilities were considered to be vitally necessary.

In the second phase of the relocation to Kanata, a new cobalt building was completed by 1971. It consisted of four high-radiation-capacity hot cells, expandable to eight if required. Cooled deep-water storage pools were built for cobalt-60 storage of inventory and completed sources. The latest models of high-density shielding windows and master-slave manipulators were installed. All heavy fixtures – the eight-tonne hot-cell enclosure doors, 25-tonne bridge cranes for overhead handling of shipping containers, remote in-cell cranes and pool retrieval devices – were motor operated. The facility became a world-wide showcase for isotope-handling technology.

The CPD personnel who remained in the Tunney's Pasture building renovated the vacated cobalt facilities to provide space for the development of new radiochemical processing technology, particularly the separation of molybdenum-99 from irradiated uranium, and for the installation of the Chalk River-developed SLOWPOKE-2 reactor (see chapter twenty), for research.

COBALT-60 FROM POWER REACTORS

By 1965, CPD's food-preservation research program, based on gamma irradiation using cobalt-60, indicated that an expanding market was available. If the demand for the irradiation plants developed as forecasts indicated, then CPD would require much more LSA cobalt-60 than the Chalk River reactors could produce. Most of the annual 7.4×10^7 GBq (2×10^6 Ci) of cobalt-60 would be required to support the need for HSA cobalt-60 for theratron sources if the theratron business continued to expand, leaving less than 50 percent for LSA production. Cobalt-60 irradiators for food and medical-supplies sterilization were expected to be loaded with as much as 1.85×10^8 GBq (5×10^6 Ci) of cobalt-60 each.

An arrangement was made whereby cobalt target rods were irradiated in the reactors at Pickering A and B, Bruce B and Gentilly-2 nuclear power stations. The power reactors, in effect, would be producers of a saleable by-product.

Due to the long production period for cobalt-60 in power reactors (one to two years), and since the primary objective of the CANDU reactor being electricity production, CPD accepted responsibility to receive and pay for all cobalt-60 discharged at each reactor shutdown period (annually, on average), according to a long-term agreement. Any reduction in future production would require advanced negotiations. The potential annual production of LSA cobalt-60 from these flux adjuster rod irradiations was:

Pickering A and B	4.7×10^8 GBq	(1.3×10^7 Ci)
Bruce B	4.4×10^8 GBq	(1.2×10^7 Ci)
Gentilly-2	7.4×10^7 GBq	(2×10^6 Ci)

CPD was ready with their new high-capacity hot cells at Kanata when the first reactor discharge was made in 1972.

CPD/RCC — GROWTH, STAGNATION, PROFIT DECLINE

Theratron Units

CPD made excellent business progress from 1950 to 1970. Sales of equipment requiring cobalt-60 had been remarkable. By 1960, CPD had established basic standards for theratron units.

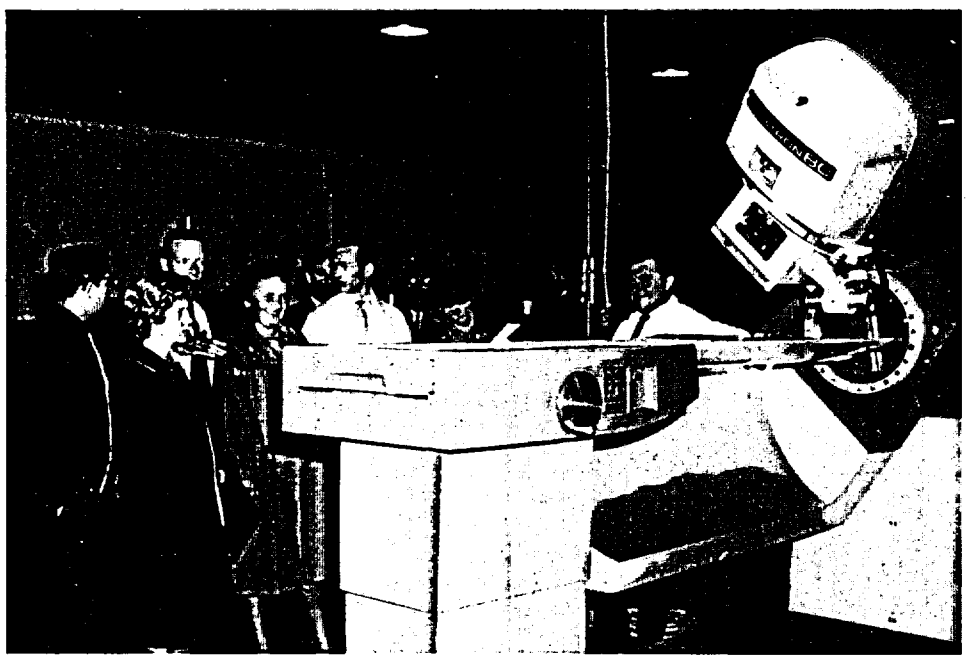
In the following decade, major modifications to theratrons were made as physicists gained experience and requested changes. This resulted in the first of a new series of theratrons for the seventies: the vertical units, Eldorados 6, 8, 76, and 78, followed by modified Model B theratron units, the C, C', F, 60, 80, 760, and 780 (see figure 5.1). Many options became available, with some units being fully computerized for treatment setup. In 1968-69, CPD sales of theratrons reached a peak of 126 units. By 1970, a total of 600 units had been built. (Some models were contracted out for manufacture by CPD-licensed agents in Europe.) All were loaded with AECL-produced cobalt-60 sources[4] and installed by CPD's installation group or their licensed agents. More than 90 percent of the units were exported, yielding about 60 percent of all CPD revenues as foreign exchange.

Annual revenues by 1969 and 1970 exceeded $10 million, but residual profits were not high, dropping to less than $400,000. Orders for new theratrons then dropped alarmingly when the number of suppliers increased, with each offering enhanced technology and performance. Field maintenance problems arose with CPD's new line of theratrons for the seventies (malfunctions in solid-state boards, patient anti-collision devices on couches behaving erratically, and, most seriously, the detection of many cracked rotational arms on theratrons in current operation). About 150 arms required urgent replacement, which put severe strain upon CPD's capabilities. The replacements in the field were costly and caused serious delays on new unit deliveries.

The introduction of medical linear accelerators (LINACs) was a factor in the reduction of the theratron market. Varian Associates, of Palo Alto, in the United States, normally provided research accelerators, but in 1967 began to market its Clinac 4 MeV medical LINAC. This LINAC was only marginally more effective than cobalt-60 for deep-seated treatment of cancer. In spite of their higher cost and dubious reliability, accelerator installations increased rapidly. The French CGR Group began to market a 6 MeV medical LINAC (the Neptune) and a 32 MeV Sagittaire unit. All of this seriously affected the cobalt-60 sales picture.

In 1970, CPD was able to market only fifty theratrons and the large-capacity shop began to look for non-CPD work, such as the manufacture of CANDU reactor components.

FIGURE 5.1
Early Theratron 80



Medical LINACs

The impact of LINAC competition was sufficient for CPD to consider entering the LINAC field. CPD had no expertise with medical accelerators and would need to enter a cooperative

[4] The amount of cobalt-60 loaded into each source was approximately 7.4×10^4 GBq (2,000 Ci). The total cobalt-60 encapsulated into 600 theratron sources by 1970 was 3.7×10^7 GBq, (1×10^6 Ci), all produced at Chalk River, and most fabricated by CPD.

venture with an experienced company. The decision was difficult, for the venture would be the largest ever for CPD, and success was by no means certain. In 1972, a five-year agreement was signed with the French company, CGR.MeV, a subsidiary of the Thomson-CGR Group. This group had been CPD agents for many years, selling CPD's theratrons in Europe. To complement their Neptune and the large Sagittaire lines, a LINAC more suitable to the size of clinics – the Saturne, a 20 MeV unit – was being developed and was nearing the prototype stage. CPD realized that selling these accelerators in the United States would complement its theratron market and, at the same time, allow CPD to compete seriously for American accelerator sales. Before the United States would accept the accelerators, however, major redesign would be required to meet American radiation standards.

At the same time, Chalk River was developing a double-pass, high-energy LINAC that would provide 5 and 25 MeV electron beams, and was anxious for CPD to enter the LINAC field with its product. However, AECL's board decided that Chalk River was not commercially oriented. Chalk River accepted AECL management's recommendation regarding the CGR.MeV contract. The decision, while seemingly correct at the time, was to cause CPD considerable trouble and expense later.

The redesign of the French units, the necessary shop expansion for assembly and testing, plus the extensive technical training for installation, made the program extremely expensive. There were licensing delays, and demanding service requirements. A total of 36 Therac 6s (Neptune), and 20 Therac 20s (Saturne) were installed in North America. They all performed well, but the necessary competitive selling prices resulted in a severe loss situation for this product. In 1978, CPD decided that the expense to market Theracs with continuous losses would decimate all other programs that were emerging. Thus, a withdrawal from the French venture was planned in 1979. The AECL board urged CPD to continue the program with the Chalk River double-pass Therac 25. CPD agreed and installed twelve units, built to Chalk River specifications by 1985. These units initially failed to operate reliably. Six patient overdose accidents occurred, due to software deficiencies of the computer-control system. The product showed no sign of becoming profitable and the manufacture of additional units was abandoned by CPD in 1985.

Industrial

Since the mid-fifties, strong emphasis had been placed by CPD's R&D group on developing products that would use cobalt-60. This led to a concentration on radiation devices to sterilize medical supplies, assist the study of gamma-radiation effects on materials through laboratory experiments, and extend the shelf life and storage time of food. All avenues were explored concurrently.

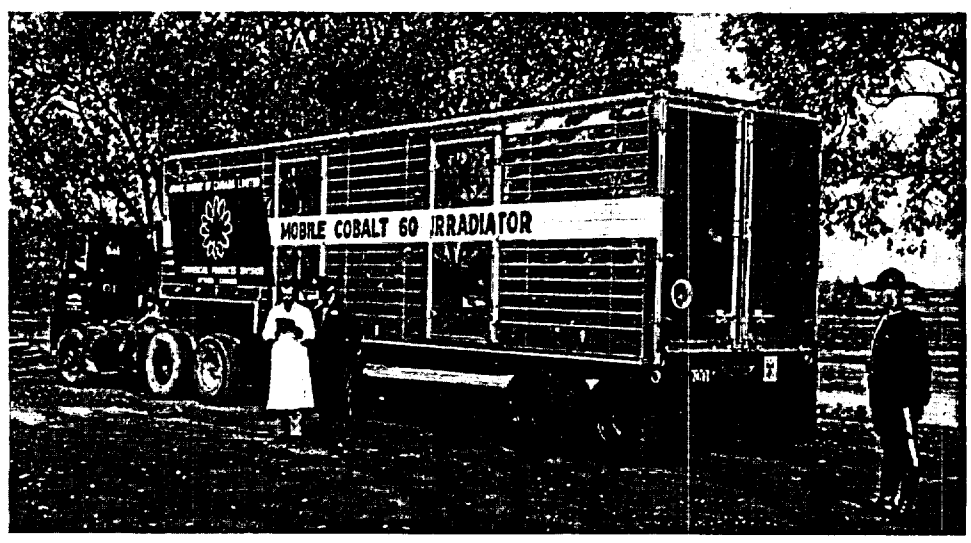
Food preservation has always been important in developing countries. Much of their grain produce is destroyed by insect infestation, and many foods are destroyed by their own enzyme reactions due to limited refrigeration. Studies by CPD R&D proved that the storage life of many foods could be extended by cobalt-60 gamma irradiation, while leaving the food safe and palatable.

Results from these studies prompted CPD to build the Mobile Cobalt-60 Demonstration Food Irradiator (MDI) in 1961 (see figure 5.2). It contained a cobalt-60 source of 5.92×10^5 GBq (16,000 Ci) that required 18,000 kg of poured lead shielding. This was mounted on a heavy-duty road trailer and pallets of food on a conveyor were passed around the exposed source in ferris-wheel fashion within the shield, to provide an approved 10 kilorad dosage. The unit was hauled to eastern Canada, the Maritimes, through Michigan and to California from 1961 to 1964, demonstrating the storage-life extension of irradiated potatoes, fish, berries, onions and grapes.

The American military purchased a CPD-designed cobalt-60 food-irradiation facility to test on its mobile forces. A private enterprise at St. Hilaire, Quebec, purchased one for use as a potato irradiator (it failed for commercial, not technical, reasons). Inroads into the United States were prevented by lack of approval from the United States' Food and Drug Administration (FDA) despite the military testing, the publication of the experimental data, and acceptance in many other parts of the world.

Cobalt-60 irradiation was a proven method of destroying bacterial contamination of medical supplies. However, a large proportion of the medical-supplies industry was locked-in and satisfied with the existing method of exposure of medical goods to ethylene-oxide gas for sterilization, making cobalt-60 irradiation to sterilize prepackaged goods hard to sell. An important breakthrough came in 1962 when CPD was asked by Ethicon Inc. (a Johnson & Johnson subsidiary), in the United States, to bid on the design and supply of a cobalt-60 medical-sterilization plant. Obtaining this contract over a prominent world supplier (Marsh, United Kingdom) was a coup for CPD, as it established them as a world leader in the field. The plant was installed in 1964 in Sommerville, New Jersey. Whereas other parts of the American medical industry were initially slow to adopt this technology, installations were made by other Johnson & Johnson subsidiaries in Australia, Sweden, Italy, Brazil, Mexico, and Peterborough, Canada.

FIGURE 5.2
Mobile Cobalt-60 Demonstration Irradiator Designed and Built 1960/61



The apparent need for small in-laboratory devices to provide high gamma-radiation doses within a shielded enclosure of a 1-to-2 litre volume led CPD to design a variety of units, called Gammacells, that could be loaded with 2 to 8×10^5 GBq, (6-24,000 Ci) of cobalt-60. The first unit was exhibited at the New York Atom Fair in October 1957. These have become popular in research centres throughout the world for studying irradiation effects on biological and non-biological specimens.

A specific variation of this device, configured for the irradiation of human blood to destroy lymphocytes, used for transfusion into immuno-suppressed patients (newborn, leukaemia patients), has a growing market. Another is the "Panoramic Irradiator", capable of irradiating large specimens in a shielded room.

A technical process developed by CPD that failed to gain market acceptance was for hardening of certain woods, such as parquet floors, for use in heavy- traffic areas and bowling alleys. This hardening was achieved by the vacuum induction of liquid methyl-acrylic plastic into wood pores, which were then polymerized by cobalt-60 irradiation. Installations were made in the CPD (RCC) Engineering lobby, and another in the social area of the Nepean Sportsplex in Nepean, Ontario. An installation was likewise made by an American supplier at the Kansas City airport. All of these installations have been in constant use for some twenty years with minimal signs of degradation. The cost of shipping the wood to CPD, the vacuum process, the irradiation and the subsequent return of the product became expensive, and a good market failed to develop.

MAPLE was a device designed to utilize the heat of decay from a large cobalt-60 source to activate a thermo-electric power generator. The first unit, housed in a four-tonne shielded container, was used on the St. Lawrence River to operate a flashing beacon light. A second unit was located at Resolute Bay, to service weather-recording instruments. However, ordinary maintenance, such as replacement of light-bulbs, or replacement of the recording pen and paper at frequent intervals, negated the long-term advantages of cobalt-60. The initial product cost was

very high (cobalt-60 and shielding costs). The units worked well, but after the expiry of the first half-life, they were not replaced.

Radioisotopes

Throughout the fifties and until the mid-sixties, CPD had established itself as a major supplier of bulk radioisotope products to much of the world, including Canada, the United States, parts of Europe, and Japan. The future looked bright.

In the late sixties, however, cancer-treatment clinics began using the many newer radioisotopes that were proving to be more suitable for the diagnosis and treatment of cancer. CPD was not ready for this market and, as a consequence, was relegated to being a bulk supplier of the older radioisotopes.

CPD'S (RCC) RESURGENCE PROGRAM

With annual revenues virtually flat at $10 million from 1970 to 1975, and the high internal expenses to pay for the new facilities plus the expensive LINAC program, the AECL board in 1975-76 had to decide whether to sell CPD to interested American buyers or to support CPD in a recovery program. The AECL board acknowledged that CPD had been good for Canada, that it had been profitable in the sixties and that, foreign ownership of part of Canada's nuclear capability was not acceptable. The board, therefore, decided to support a resurgence program.

Medical Products

Early acceptance of CPD's theratrons in the sixties had given it a leadership role in world teletherapy manufacture. When sales of theratrons began to decrease in the seventies, due, in part, to LINACs, CPD realized that if their market share was to improve, their theratrons would require conceptual changes (simplicity to reduce production expense, a design to meet competition on a feature-by-feature basis, and affordable pricing for smaller clinics).

A second series of major revisions, based on the best features of the series produced in the seventies, culminated in the dropping of vertical units. The new models comprised: the Phoenix – a basic, manually operated, no-frills theratron, with upgradable options; the Theratron 780C – a higher-capacity radiation unit, with computerized options; and the larger Theratron 1000. These reached the market in 1986, and a promising market recovery began.

Accessory devices for theratrons were always part of CPD's program. A patient pre-setup unit introduced in 1977 was the Simulator (now trademarked as Therasim). This device is smaller but similar to the theratron unit. X rays are used to assist in accurate tumour location, and to establish the required cobalt-60 dosage from the theratron. The Therasim minimizes patient stress by reducing treatment setup procedures beneath the large therapy unit, and permits the clinic to handle a larger patient load. Originally, the Therasim was considered a luxury item for clinics. However, in 1982, the 100th Therasim produced was shipped to the Saskatoon Cancer Clinic.

Coincident with the Therasim, a computerized "Theraplan" system was developed as a valuable accessory for the planning of ongoing theratron treatments. Theraplan consists of a tumour-registry system logged into a personal-computer database. It comprises a patient-dose compensating device, simulated with a water phantom and a film-dosimetry recording system, to measure therapy unit outputs and the transfer of these data to the computerized treatment plan for subsequent treatments.

The Montreal Neurological Institute (MNI) became interested in the development of a body-scanning device similar to a rudimentary unit that they had observed at the Bethesda Maryland Hospital. They developed a prototype for study and evaluation. MNI saw that a fully developed device using positron emitters (obtained from cyclotrons) would be useful in brain-scanning research. It asked CPD in 1978 to develop a 3-D Positron Emission Tomography unit (PET) for commercial sale. With a $2 million budget and two scientific teams (from CPD and Chalk River), a prototype unit, the Therescan 3128, was built. The computerized software and the unit were developed by CPD while the resonance detectors were developed by the Chalk River team. The unit was sold as a prototype for about $1 million to

MNI in 1980. In 1980, TRIUMF, of the University of British Columbia, placed an order for a similar unit for delivery in 1981. However, their order was cancelled the following year, due to a conflict of specifications. After some usage of the prototype, MNI requested CPD to develop a more sophisticated unit. CPD realized that few institutes would have the expensive infrastructure (cyclotron, radioactive processing facility) to operate such an assembly, as it was basically a medical research device. Few sales were expected and CPD was not able to finance additional development funds. The project came to an end in 1982. There are five PET units in Canada today.

The unit sold to MNI operated reliably for seven years, and was eventually resold to a Scandinavian group. Various other medical treatment devices were investigated, and prototypes built. Most were technically successful, but due to their expense or lack of market interest all were abandoned. Such devices were the Brachytron (a body intercavity unit for internal radiation techniques), a neutron-therapy unit (see chapter nine), and a cesium-137 source unit.

Industrial Products

The expected approval from the FDA to permit food irradiation in the United States failed to materialize, reflecting public reluctance to accept any food associated with irradiation. This delay and no other available market obligated CPD to accept a large inventory write-off of power-reactor cobalt-60 in 1974, thereby incurring a loss of more than $1 million that year.

While continuing the struggle to obtain FDA approval of food irradiation, CPD believed that a resurgence in this field would come from improved sales of the variety of cobalt-60 irradiation devices that they had developed during the mid-fifties – in particular, medical-supplies sterilization plants plus the sales of laboratory radiation devices, backed up by a program of cobalt-60 resupply to counterbalance decay.

Following the breakthrough in obtaining the medical sterilization contract from Johnson & Johnson in 1962, emphasis continued on the development and sales of larger-capacity plants. These were initially designed to contain as much as 3.7×10^7 GBq (1 million Ci) of cobalt-60, but soon the average design capacity far exceeded that amount.

An interesting adjunct to this growth was that the large quantity of power-reactor cobalt-60 that CPD was required to write off in 1974 had been sold before 1976. By 1974, a total of twenty-seven such plants had been installed by CPD, a few being installed in the United States for several medical suppliers.

Radioisotopes

To support a resurgence in the sales of radioisotope products, NRU operations were modified to permit easier entry/exit of targets to the high-flux regions of the reactor. Chalk River installed a loop in NRU for the irradiation of circulating xenon gas, which permitted selective tapping off of carrier-free[5] iodine-125 in good quantity, and in a more efficient way than the low-yield process CPD had established earlier. Economical quantity production of this radionuclide processed to a usable radiochemical had been a long-time CPD objective. It is used in quantity for radioimmunoassay studies.

Iodine-131, a carrier-free standard product since 1948, is used for thyroid imaging and therapy. A lengthy wet-chemical process was required to separate the iodine-131 from irradiated tellurium metal. It was messy, replete with costly process losses, and was uneconomical to produce. A dry-vacuum-distillation method of iodine-131 separation from irradiated tellurium dioxide had been successfully tested by CPD's R&D, but it required a larger target weight. The availability of the high NRU fluxes overcame this disadvantage without loss of radioactive quantity production. A full-scale extraction system was built that gave excellent and reliable results in a fraction of the usual processing time, and at reduced cost. The tellurium metal method was immediately abandoned.

The new product that CPD most wanted to produce in quantity was fission-product molybdenum-99, having a 66.7-hour

[5] Carrier-free: All atoms of the radioisotope exist in a pure form. It is not diluted, nor contaminated with any other radioisotope, or non-radioactive material, a desirable feature in the irradiation of human tissue.

half-life. Molybdenum-99 decays to technetium-99m, a nuclide that emits a single gamma photon of 140 keV, and has a six-hour half-life. Biological compounds targeted for selective absorption by body organs can be readily labeled with technetium-99m for scanning body functions. Technetium-99m is used in perfusion (blood flow), brain and heart scans, and to detect bone fractures. The energy level is easily collimated, resulting in sharp images by gamma cameras. Technetium-99m has been considered as the almost perfect diagnostic tool and has gained rapid market acceptance.

The product would be very profitable to any major supplier who was able to supply large and routine quantities of carrier-free molybdenum-99. In 1972, NRU was to undergo a lengthy service shutdown and would not be available for irradiations for an extended period. McMaster University (Hamilton, Ontario) assisted CPD to gain a foothold in the technetium-99m market by offering their small reactor (2 MW) to irradiate targets of enriched uranium that CPD would process. A three-year agreement was signed. The reactor power level was raised to 5 MW to increase production. However, yields were irregular, at less than 3.7×10^3 GBq (100 Ci) per week. CPD knew that, to be effective in the marketplace, routine quantities in the order of ten times that amount were needed. The availability of the McMaster reactor, however, allowed CPD to develop its molybdenum-99 process.

As soon as NRU was again operational, CPD switched its irradiation of uranium targets to NRU. Molybdenum-99 production quantities from NRU were easily sufficient to meet the large market demand. CPD began the fission-product separation process at the Tunney's Pasture laboratory. As the demand increased, the fission-product separation process quantities became too great for Tunney's Pasture. Also, the arrangement involved daily shipment of irradiated uranium targets from Chalk River to CPD for processing, and then the return of fission-product residues and the unused uranium-235 to Chalk River. It was decided to switch the first stage of chemical separation to Chalk River's universal cells. Chalk River went back into routine radiochemical processing for CPD. The unrefined fission-product molybdenum-99 was separated there and shipped in bulk to CPD for refinement and quality control.

To ensure reliable availability of molybdenum-99, CPD and Chalk River organized smooth production and transportation schedules. Rapid Customs clearance and the use of chartered aircraft were arranged. Moly-99, as it is called in the trade, provided substantial profits from 1978 on, with sales approaching $20 million annually by the mid-eighties.

Another product resulting from the molybdenum-99 program was the collection of xenon-133 fission product at the molybdenum-99 separation stage. This became a new product, useful in lung scanning.

In time, the accomplishments of CPD (RCC) became respected throughout the scientific world. Over the years, Australia, India, Pakistan, Belgium, Mexico, Thailand and Taiwan have consulted CPD in the development of their own radioisotope products.

Cyclotron-Produced Radioisotopes

Certain cyclotron-produced radioisotopes have special medical uses and have characteristics that cannot be met with reactor-produced nuclides. These are converted by pharmaceutical firms into aseptic products called radiopharmaceuticals. Such products command high selling prices.

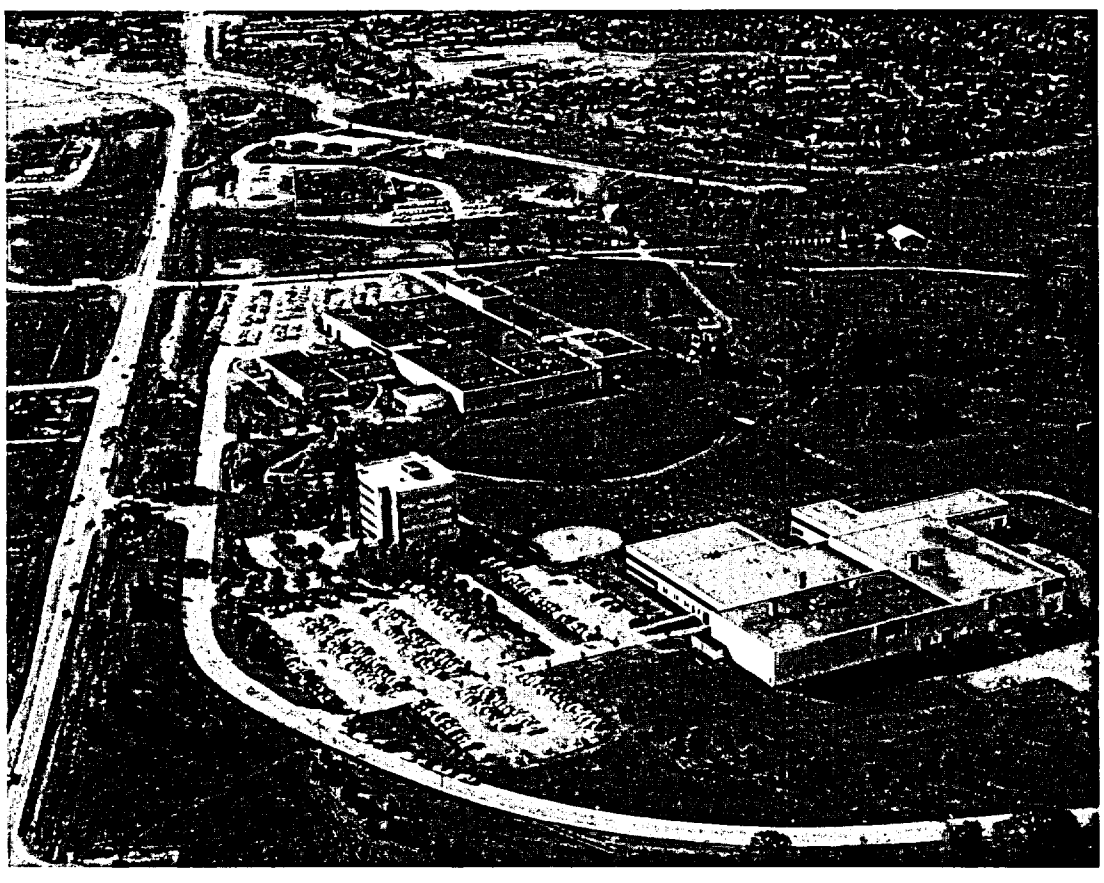
In 1978, the CPD (RCC) product-development group proposed entering this growing segment of the market, to which they had no access. The large TRIUMF cyclotron at the University of British Columbia was made available on a contract basis. In 1980, CPD (RCC) constructed a four-hot-cell processing laboratory at TRIUMF, and ordered a 42 MeV-type CP42 cyclotron. Meanwhile, it shared the use of the TRIUMF 500 and 70 MeV beams. Production of thirteen-hour iodine-123 from enriched xenon-124 in bulk quantities was planned initially. It has been used extensively in heart and brain studies. In 1982-83, the first shipments of radiopharmaceuticals were made to hospitals and clinics in Canada.

The resurgence program for radioisotopes became a total success. CPD (RCC) had become a major supplier of both the older and the newer medical radioisotopes throughout the world.

FINAL RELOCATION FOR CPD/RCC

To cope with the production of the many new products and the higher radioactivity, it was obvious that the Tunney's Pasture laboratories had to be closed down. In 1979, final consolidation of CPD (RCC) at Kanata was started. By 1984, new offices and laboratories were completed (see figure 5.3). All Tunney's Pasture operations and some 180 personnel joined the 600 already at the Kanata site in the $35 million, 12,540 square-metre Kanata Isotope Production Facility (KIPF). KIPF contained twenty new hot cells constructed of lead and iron, instead of high-density concrete, reducing them to about one-third of their former size but having the same radiation-shielding capacity. Each radioisotope was to be processed in dedicated and isolated hot cells. These included cells for iodine-131, iodine-125, carbon-14, xenon-133, iridium-192, and yttrium-90 for radiography sources, and well-equipped facilities for the molybdenum-99/technetium-99m line. Cells were planned to handle special irradiations upon demand, for the R&D group, and to handle waste products. KIPF was built as an integral part of the 1971 cobalt-60 facility. Today it represents the largest and most modern radioisotope-production facility in North America.

FIGURE 5.3
Cobalt and Radioisotope Site, Kanata, 1980



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BASIC RESEARCH

Chapter Six

Basic Research: An Introduction

G.C. HANNA

This introductory chapter explains what is meant by basic research, outlines its historical foundation, and summarizes subsequent developments from an organizational point of view. The research itself is described in the three subsequent chapters.

A very complex terminology has grown up to describe various types of scientific research, much of it redundant and disputed. In a 1972 monograph on "Forty-five varieties of research (and development)", Lord Rothschild argued that "pure", "basic", and "fundamental" are synonyms, all describing research "done solely to increase knowledge without any practical application in view", and that all other research, which may be described as "underlying", "strategic", "mission-oriented" etc., is simply applied research. However, it is difficult in practice to maintain a completely clear distinction. Applied research can be directed to very long-term objectives, and its practitioners can be as curiosity-motivated as pure-research scientists; they in turn are only too happy if their contributions to pure knowledge are promptly applied to practical problems.

This section on basic research is not confined to pure research, but includes some applied programs, generally of a long-term nature, that are not covered elsewhere in this book. It does not include research in the life sciences, which is described in chapter three.

In the early years of the Canada/United Kingdom project, particularly in 1944 and 1945, almost all the research was directed to the design and operation of NRX and the chemical extraction plants. As the immediate problems were dealt with, at least provisionally, research activities began to separate into two conceptually distinguishable types. The first met the requirements of existing major projects and formally-approved new ones (e.g., the NRU reactor). It

was greatly expanded, starting in the mid-fifties, to meet the needs of the power-reactor program, and it is described in other sections.

The second type of research was aimed at obtaining a more fundamental and detailed understanding of the various processes involved in nuclear science and its applications. The question of whether it was pure or applied research was not initially of much concern. Pure research had discovered fission and its application had come very quickly. It was therefore not unreasonable, in the early days of the project, to think that further research in this new area of science could lead to discoveries having some practical importance in the near future. The scientists were eager to develop the tools of their trade, and as they did, their researches, in a rapidly expanding field, did not appear to them as obviously pure or applied. This early period is characterized by the large number of research scientists who were simultaneously engaged in activities that, in retrospect, are identifiable as pure research, applied research, and instrument development. Items such as mass spectrometers, particle detectors, and electronic systems were designed and built by the experimenters themselves; nowadays, corresponding items are readily available from commercial sources, regrettably almost all outside Canada.

Unlike the applied research programs which, as noted above, were greatly expanded, pure research in the physical sciences has been maintained at a relatively constant level. With access to advanced facilities, many of them unique in Canada, the scientists have successfully exploited their opportunities to carry out forefront research of international quality, and AECL has done its best to update the facilities on which the research depends.

That AECL has a strong program in basic research is ultimately attributable to C.J. Mackenzie, who was acting president of the NRC when the atomic energy project was set up. He recognized it as an opportunity, in a prestigious area, for the advancement of Canadian science, which he saw as vital to the future of the country. J.D. Cockcroft, director of the project, was also a strong supporter of broadly based research, as would be amply demonstrated later at Harwell. E.W.R. Steacie, the deputy director, emphasized the importance of fundamental research in an August 17, 1944 memorandum to Mackenzie, an emphasis that characterized his entire career, which culminated in the presidency of NRC (1952-62).

In September 1944, Cockcroft and Steacie proposed that NRC should build an atomic energy research centre in Ottawa to take over from the Montreal laboratory when it was closed down. This centre would be additional to the major facilities and associated laboratories at Chalk River. Mackenzie was sympathetic, but G.C. Laurence, the senior Canadian physicist, argued strongly against dividing up what would be a relatively small research group after the United Kingdom staff returned home. In July 1945 it was decided, perhaps mainly for financial reasons, not to proceed with the Ottawa centre, but to transfer all the Montreal laboratory activities to Chalk River.

NRX, the world's best research reactor at that time, would of course be the centrepiece of the program, but there were other elements. The construction of a Van de Graaff accelerator had been initiated by Cockcroft in February 1945, and this laid the foundation for what became AECL's largest pure research program, nuclear-structure physics. This accelerator was to have been located at the proposed Ottawa centre, and Cockcroft justified it as "a necessary tool for the investigation of nuclear fission and absorption levels, since it can provide a monokinetic neutron source of controllable energy. Such a generator has been in constant use in the American project and it is our intention to build one in England." Not exactly pure research.

The early program also included high-energy physics, using cosmic rays, and this expertise made possible the study, described in chapter nine, of neutron production by high-energy particles. There were even plans, by B. Pontecorvo in 1946, for an underground experiment to detect solar neutrinos. This would have anticipated by some twenty years Davis's celebrated experiment, using the technique proposed by Pontecorvo, in the deep Homestake Gold Mine, South Dakota.

W.B. Lewis, who succeeded Cockcroft in 1946, was a strong and effective supporter of basic research. He demanded high standards, not least in matters of presentation and publication, but left the scientific direction of pure research to the scientists

while he concentrated his formidable abilities on applied-research activities.

When AECL took over from NRC in 1952, the commitment to basic research was maintained. Nor was there any abrupt change in 1955 when the power program was approved, giving a prime focus to what had been an imprecisely defined mission. The reactor-based research continued to flourish, with NRU (1957) displacing NRX for most of the work. The Van de Graaff was replaced by the world's first tandem accelerator in 1959, which was succeeded by a larger one in 1967 (see chapter eight). Research in chemistry and materials science (see chapter seven) was sustained with a variety of advanced equipment, including mass spectrometers, a mass separator, electron microscopes, and electron and positive-ion accelerators. The basic-research program would have been greatly expanded had Lewis obtained approval for his Intense Neutron Generator (ING) project, but this was cancelled by the government in 1968 (see chapter nine).

In 1976, anticipating demands for more detailed accountability, AECL set up, under an R&D program committee, six steering committees to plan, coordinate, and report on R&D activities in i) power reactor systems, ii) the nuclear fuel cycle, iii) environmental protection and radioactive waste management, iv) heavy water processes, v) assessments and new applications, and vi) underlying and advanced systems research.

The purview of this last steering committee was essentially the area covered in the present section (and the R&D described in chapter three); i.e., "underlying" included pure research, and "advanced systems" referred to the accelerator and fusion-related work described in chapter nine. The steering committee justified its research activities, which accounted for almost one-third of the Research Company's budget, as:

- providing AECL's current mission with better fundamental understanding of the processes involved in nuclear energy systems, with a knowledgeable consultancy service, and with a mobile reserve of versatile individuals for dealing with emergencies;
- meeting AECL's need to be aware of, and to be able to respond to, developments elsewhere, by exploiting the "subculture of science" (meaning personal contacts with researchers in other institutions), which could provide intelligence when formal channels were restricted. It was considered particularly important to be able to evaluate without delay the potential of plausible schemes that could otherwise lead AECL into expensive blind alleys;
- providing the basis for future advance.

In retrospect, this rationale did not adequately emphasize the prime purpose of pure research, which is the advancement of human knowledge, and the particular responsibility that developed countries and large laboratories have for pure research – an activity whose benefits to those who fund it are seldom direct and immediate. It could also have been pointed out that achievements in this area added to AECL's overall prestige.

The rationale had some success. The underlying and advanced systems research continued to receive about the same share of the total R&D budget, and the nuclear physicists' superconducting cyclotron project, which was started in 1974, was allowed to proceed to the construction phase in 1978 (see chapter nine).

By this time, most of the more applied programs in underlying research (other than those described in chapter three) were in chemistry and materials science (see chapter seven). They included work on radiation chemistry, high-temperature-solution chemistry, isotope separation, radiation-induced creep and growth, hydrogen uptake by zirconium, and zirconium corrosion.

The pure research program had three major, world-class components. The largest was the accelerator-based nuclear-physics program, which concentrated on the use of heavy ions to study the structure of nuclei (see chapter eight).

The second one, at NRU, investigated a wide range of phenomena in condensed matter physics by neutron scattering. It exploited, in particular, the triple-axis-spectrometer technique that had been pioneered at Chalk River thirty years earlier. Thanks to excellent instrumentation, this program remained fully competitive with programs based on newer higher-flux reactors in Europe and the United States (see chapter eight).

The third research program arose out of the 1962-63 discovery at Chalk River of channeling, which is the ability of energetic positively-charged particles to penetrate further into crystalline

material by following preferred directions (i.e., channels) between the regularly spaced atoms in the crystal. Of the three pure research programs, it provided the most immediate applications, and its use in solid-state studies, especially of semiconductors, was energetically pursued, and attracted visiting scientists from around the world (see chapter seven).

Unfortunately, budgetary pressures were becoming increasingly severe and, rather than continue to squeeze all three programs, AECL decided in 1985 to transfer the channeling program, recognizing that it could, unlike the other two, be successfully continued in a Canadian university environment – which it was, thanks to considerable initial help from AECL.

The neutron-scattering program was continued because Chalk River is one of the few places in the world where such research can be done, and the research represents outstanding value for money, making minimal demands on a facility (NRU) already funded for other purposes. For nuclear physics, the cyclotron was just being commissioned, which would ensure the future of this Tandem Accelerator Superconducting Cyclotron (TASCC) laboratory as one of the world leaders in nuclear-structure research. Both TASCC and the neutron-scattering installations at NRU constitute national facilities used by Canadian universities, who have supplied major pieces of equipment. This was no doubt a significant consideration in 1985 and will remain an important justification for the future survival of these programs.

In the early eighties AECL started to change the way its programs were planned, performed and reported to government. By February 1986, a new organization was in place (see chapter two, figure 10) with four program responsibility centres: PRC 1, for reactor development; PRC 2, for waste management; PRC 3, for physics and health sciences; and PRC 4, for radiation applications and isotopes. Of the R&D activities previously classed as underlying and advanced systems research, those described in chapters three, eight, and nine all went to PRC 3, except for accelerator physics (chapter nine) and the environmental research work at Whiteshell, which went to PRC 4 and PRC 2, respectively. The chemistry and materials-science programs of chapter seven that remained after the channeling program disappeared were considered to be applicable to specific near-term objectives. The materials-science programs went to PRC 1, chemistry at Whiteshell to PRC 2, and chemistry at Chalk River to either PRC 1 or PRC 4.

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Chapter Seven

Chemistry and Materials Science

T.A. EASTWOOD

INTRODUCTION

The basic research programs in chemistry had their origins in the Montreal Laboratory. The task of the laboratory was the exploration of an alternative to the plutonium production route being followed in the Manhattan Project. The American program was based on graphite-moderated reactors and, as a backup, the Anglo-Canadian team pursued the heavy-water-reactor option. A major contribution of the chemists was the successful development of the process used to extract plutonium from uranium irradiated in the reactor. Working independently, they duplicated one of the main achievements of the chemists in the much larger Manhattan Project. In addition, they developed the process used to separate uranium-233 from irradiated thorium carbonate, a secondary objective of the Montreal Laboratory. This work aroused the scientific curiosity of the staff and led quite naturally to the research described here under the headings: Nuclear Chemistry and Heavy-Element Chemistry.

The third main component of the early chemistry-research programs arose directly from the commitment to heavy-water-reactor technology. It was expected that the nuclear radiations in the reactor would decompose the heavy-water moderator. The details of the chemical steps leading to decomposition were not well understood, however, and were of significant scientific interest. Research on this topic in radiation chemistry was launched in Montreal and continued throughout the period of this history.

As the research potential of NRX became apparent, research activities broadened. The reactor was used to make radiotracers for application in a variety of new fields, and it was shown to be a powerful tool for neutron-activation analysis. The effects of reactor irradiation on solids soon attracted attention. These activities are summarized under the headings: Radiotracer Studies, Irradiation Effects, and Analytical Methods. As the nuclear power program advanced, the research programs developed in tandem and are described later in this chapter.

NUCLEAR CHEMISTRY

There seems to be no generally accepted definition of nuclear chemistry. Within the Canadian project, it was considered to be the study of nuclear processes, interdisciplinary with physics, in which chemistry plays an important role. One such process that occurs in nuclear reactors is the synthesis of new chemical elements: the production of plutonium (element 94) from uranium (element 92), the aim of the Montreal Laboratory, is a well-known example.

Shortly after NRX startup, elements of even higher atomic number were made in the reactor. Plutonium was irradiated to yield americium, and it, in turn, was irradiated to make curium (atomic numbers 95 and 96, respectively). These elements had first been synthesized about five years earlier at the University of California. The nuclear properties of the americium and curium isotopes produced in NRX were investigated in the early days at Chalk River.

The question arises: is there an end to the new elements that can be made this way? Indeed, there is. With the progression to heavier nuclides, the half-lives tend to be shorter and, an additional process, spontaneous fission, opens another route by which heavy nuclides can decay. Production then becomes a competition between the processes for formation versus those leading to destruction by decay. The key factor is the neutron flux, and, in this feature, NRX was superior to other reactors for a number of years. That is why it was in demand by scientists active in this line of research, not only at Chalk River but also at other laboratories.

Eventually, NRX was surpassed in neutron flux by the Materials Testing Reactor (MTR) in the United States. This reactor opened fresh possibilities for heavy- element synthesis. In the mid-fifties, AECL chemists cooperated with scientists at the Knolls Atomic Power Laboratory at Schenectady, New York, to investigate the heavy elements in samples of plutonium the latter had irradiated in the MTR. In addition to isotopes of americium and curium, they were looking for isotopes of berkelium, californium, einsteinium, and fermium, elements that had been discovered a few years earlier, with atomic numbers 97, 98, 99, and 100. They thought that isotopes of even heavier elements might be produced, but they knew that they would have to work quickly to study them because the half-lives would be short. The sample that had received the longest irradiation was processed as rapidly as possible after the end of the irradiation, and all the trans-plutonium elements up to, and including, fermium were indeed found. Using the techniques that had been developed at Chalk River, the nuclear properties of the isotopes of these elements were investigated in the co-operative program. The results confirmed and, in some instances, refined the information in the scientific literature. No sign of elements beyond fermium was found. Heavier elements have since been synthesized elsewhere, but in accelerator irradiations, not in reactors.

A major interest of the nuclear chemists in the Montreal Laboratory was the fission process. From a practical standpoint, this is the most important nuclear process on earth, and from the scientific viewpoint, its complexities attracted the attention of experimentalists and theorists alike. In order to study any one of the several hundred different fission-product nuclides, available in only small amounts in the early days, it was first necessary to purify and identify them chemically. This was an extraordinary challenge, because more than thirty different elements are produced in fission. In fact, this chemical hurdle had been an obstacle on the road to the discovery of fission.

The yield of products from the fission of uranium-235 by reactor neutrons when plotted against the mass of the product gives a double-humped curve, with maxima at masses of about 97 and 137. One of the earliest publications of this curve resulted from research on the radioactive fission products in the Montreal Laboratory. A similar curve for the fission of uranium-233 was also published from there a short time later. These were the beginnings of a stream of publications from the Chalk River Laboratories, refining and extending the early work. Measurements of the radioactive products were augmented by mass-spectrometric determinations of stable or long-lived nuclides made at McMaster University (Hamilton, Ontario), some of which were supported by AECL contracts. The yields of many nuclides were put on an absolute basis, as compared with the early relative yields, and the radioactive decay characteristics of the products were also investigated. Measurements made in the fifties revealed small irregularities in the otherwise-smooth yield curve. The Chalk River chemists studied them with special care because it was thought that they could shed light on the mechanism of fission. This turned out to be correct and the irregularities are now generally considered to reflect structure in the arrangement of neutrons and protons in the fissioning nucleus.

Expertise developed from research on the radioactive-decay properties of the fission products was applied later to the wide

variety of other radionuclides that could be produced in NRX. A noteworthy example was the study of the radioactive decay of calcium-41 made at Chalk River around 1950. This long-lived isotope, produced by irradiating calcium oxide or carbonate with neutrons, decays by electron-capture, a rare mode of decay in light elements. Rigorous chemical purification to rid the calcium-41 of radioactive impurities and sophisticated counting techniques were required for the successful conclusion of the work. In another early example, the half-life of carbon-14, the fundamental factor in radiocarbon dating of archaeological artifacts, was determined with greater precision than reported previously from elsewhere.

Measurements of the cross-sections of radionuclides in reactors received special attention at Chalk River as the power reactor program advanced through the fifties. The term cross-section is defined properly in chapter one; in the present context, it can be thought of as a measure of the probability for a specified reaction to occur in the reactor. Cross-sections vary over a range of more than a billion, so yields from nuclear reactions can range from the abundant to the meagre. Cross-sections cannot be calculated accurately from theory; consequently, measurements are required. The cross-sections of the uranium and plutonium isotopes were the foci of a wide variety of measurements, both chemical and physical, for they are of fundamental importance in CANDU power-reactor design, as well as of scientific interest. The cross-sections and yields of the fission products were also determined to calculate the loss of neutrons to the fission products as they accumulate in the fuel with increasing irradiation.

Neutrons abound in nuclear reactors, yet AECL chemists were able to make radionuclides in NRX that are neutron-deficient; i.e., their nuclei contain fewer neutrons than naturally occurring isotopes of the same element. One way was to use nuclear reactions initiated by the energetic neutrons born in fission. When a fast neutron is absorbed into a nucleus, the excess energy may be carried off by two, or in rare instances three, neutrons, yielding a neutron-deficient product. A reaction of the (n,2n) type led to the production of bismuth-208, a long-lived radioisotope of bismuth not known previously.

Another approach was to use secondary reactions: neutron reactions with a light element produced charged particles, which subsequently reacted with other nuclei. For example, carbon-11, a neutron-deficient isotope of carbon, was produced by irradiating lithium borate in NRX. Tritons, produced by the interaction of reactor neutrons with lithium-6 nuclei, react with nearby boron-10 nuclei to produce carbon-11 and two neutrons. Other radionuclides, including beryllium-7, nitrogen-13, fluorine-18, and sodium-22, were obtained this way.

By the early sixties, the nuclear chemists were turning their attention from the reactors to the tandem accelerator. Their main interest was the measurement of cross-sections for reactions between neutrons of precisely known energies, ranging from about 2 to 20 MeV, and a wide variety of target nuclides. The neutrons were produced in the reaction between deuterons accelerated by the tandem and tritium, and the tandem provided the precise control of deuteron energy required for the measurements. The results were of fundamental interest and provided useful information for the development of neutron dosimeters and in radiation damage studies.

Basic research in nuclear chemistry at AECL gradually wound down in the late sixties, as a result of staff changes and the emergence of programs of higher priority.

HEAVY-ELEMENT CHEMISTRY

In parallel with their work on methods for separating plutonium from irradiated uranium, the chemists in the Montreal Laboratory and later at Chalk River, sought to determine the families to which these elements belong. Scientists order the elements in sequence by their atomic number and place them in families, the members of which have similar properties. This arrangement is called the Periodic Table of the Elements. The table was essentially complete at the turn of the century, but the familial assignments of the heaviest elements were somewhat controversial. Few specialists pursued the matter, for, apart from radium, the heavy elements attracted little notice. With the advent of the nuclear age, interest increased greatly and all the nuclear establishments became

active in research on the heavy elements, including the Montreal Laboratory.

G.T. Seaborg, a co-discoverer of plutonium at the University of California, was a strong supporter of the view that all the elements from actinium, atomic number 89, to the undiscovered element 104, belonged to one family. This would include all the synthetic elements mentioned earlier.

He argued that in this actinide family, all members had the same valence (outermost) electron shell, and that an inner shell, with little influence on valence, was filled in sequence as the atomic number increased. In this respect, the actinide family members resembled the lanthanide (or rare earth) family, found near the middle of the Periodic Table, where inner-shell filling was well established. With the family assignment properly established, the properties of new elements could be predicted from the chemistry of other members of their family. Many of the actinide elements existed in such small amounts, however, that proof of Seaborg's actinide hypothesis was elusive.

Uranium was known to exist in several valence states, and the initial research in Montreal explored relationships among the states using electrochemical methods. These experiments were extended to plutonium when it became available. A wide variety of oxidizing and reducing agents was used in related research. Plutonium was found in positive valence states of three, four, and six, with a preference for four in mild oxidizing or reducing solutions. The solubilities of many plutonium salts were measured, partly with an eye to methods for separation and purification. Precipitates containing other elements with which plutonium would co-precipitate quantitatively at tracer levels were sought, and lanthanum fluoride was recommended. Plutonium was found to readily form a complex with inorganic anions, such as sulphate, as well as with a multiplicity of organic molecules. The optical absorption spectra of aqueous solutions of plutonium showed sharp bands comparable to those of uranium, and analogous to those of the lanthanide elements. Scientifically, the knowledge gained through this research was consistent with the actinide hypothesis. From a practical standpoint, it built a firm understanding of methods for separating uranium and plutonium from the fission products and from each other, for purifying plutonium, and for developing analytical methods.

A noteworthy characteristic of this work is that the stock of plutonium amounted to only a few micrograms at first, and slowly increased to milligrams as irradiated uranium became available. The plutonium was recycled through the numerous experiments, and if a test-tube broke or other accidents scattered the sample, the blotting paper placed on purpose to catch the spills was processed to recover the plutonium. Many hours were spent on these tedious recovery operations.

Metallurgical research provided further tests of the actinide hypothesis. As discussed in chapter two, plutonium-aluminum-alloy fuel elements were fabricated at Chalk River and used in NRX in the early years. In research experiments throughout the fifties and early sixties, using the same basic process, many intermetallic compounds were prepared of uranium, neptunium, plutonium, or americium with aluminum and other elements. Some of these compounds make excellent small sources of neutrons; alpha particles from decay of the heavy element produce neutrons when they interact with the light element in the compound. The efficiencies of the various compounds for this purpose were determined. Similarities in the alloying properties and crystal structures of the intermetallic compounds of these heavy elements supported the view that they belonged in one family.

Experiments aimed at reducing plutonium fluoride to the metal, on the scale of hundreds of milligrams, were made using barium and related metals in a high vacuum. The physical properties of the products, such as density, crystal structure, and melting point, were then determined. The techniques were later extended to the investigation of methods for separating plutonium from irradiated uranium using molten silver, silver-gold alloy, certain of the lanthanide metals, and bismuth.

In sum, the AECL results in the forties and fifties were consistent with Seaborg's actinide hypothesis, but did not prove it. Since then, further results from other laboratories have firmly established the validity of the actinide hypothesis. It formed the basis for separating and identifying new synthetic elements heavier than curium and, for example, was invoked in the

separation of these elements for the research mentioned earlier. Work on heavy-element chemistry lapsed in AECL laboratories for several years, but resumed at Whiteshell around 1970.

RADIATION CHEMISTRY — EARLY DAYS

Radiation chemistry embraces the study of the chemical changes produced by high-energy radiation from natural and artificial sources. It has its roots in Mme. Curie's research on radioactivity, and grew rapidly in step with the development of nuclear energy.

In the fledgling Canadian project, radiation chemistry was particularly important, because the heavy-water moderator in NRX was expected to decompose into deuterium and oxygen. NRX was equipped to recombine the gases and return the heavy water to the reactor. About the time of NRX startup, however, research at the Brookhaven National Laboratory in the United States showed that, if water is pure, the decomposition is negligibly small. Steps were taken to ensure the purity of the moderator, and the recombiner unit was not often needed. Although there was no net decomposition, a lot of radiation chemistry was going on and water radiolysis was a prominent program of research at Chalk River.

The radiolysis experiments were aimed at testing a contemporary hypothesis. It held that gamma rays dissociate water molecules, at widely spaced intervals along their paths, into positive ions and electrons. The ion-electron pairs quickly recombine to form water molecules again. These re-formed molecules are in higher-energy states than normal water molecules, and eventually shed the excess energy by decomposing into reactive intermediate species. Thus, clusters of reactive species are formed at intervals along each gamma-ray track. The clusters were estimated to be a few molecular diameters across, separated by much greater distances. The intermediates were thought to be neutral atoms and free radicals.

Atoms and free radicals are reactive by virtue of unpaired electrons in their outer electron shells; they are usually formed by the decomposition of energy-rich compounds, and are well known in other fields of chemistry. The intermediates from the radiolysis of ordinary water would be hydrogen atoms and hydroxyl free radicals, written as H• and OH•, respectively, typically three of each in each cluster.

In some clusters, the atoms and radicals would react to produce H_2, H_2O_2, and H_2O, all of which would eventually disperse in the bulk water. If reaction failed to occur in the clusters, the intermediates themselves would diffuse into the bulk water. There, if no impurities were present, they would react with dissolved hydrogen and hydrogen peroxide to re-form water. This explained why there is no net decomposition of the NRX moderator when it is pure. If solutes were present (metal ions, for example), the atoms and radicals would react preferentially with them; deuterium and oxygen (from the decomposition of the peroxide) would accumulate, and eventually escape from solution.

Most free radicals are too short-lived to have been observed directly with the techniques available at the time, so their identities and roles were hypothetical, based on knowledge from other fields of chemistry. Calculations to test the hypothesis had been reported in the literature, but to make them manageable, the authors had been forced to assume only one reactive species, a mathematical hybrid of H• and OH•, to which reaction and diffusion rates were assigned. To include two species, as required by the hypothesis, was beyond the mathematical techniques employed. For the new breed of computer mathematician at Chalk River, this chemistry problem presented a nice challenge. Using numerical techniques rather than classical analytical mathematics, and the Datatron, the first, and newly commissioned, electronic computer at the laboratory, the Chalk River researchers quickly made the calculations for two species reacting and diffusing as hypothesized.[1] The agreement between the calculations and the experimental results gave encouraging support for the hypothesis.

The calculations did not, however, reproduce all the experimental results. In particular, there was a notable lack of accord

[1] Each calculation took longer (eight hours or more) than any done up to that time. The computer staff wrote a special sub-program to start the calculations on the Datatron at night, unattended, to run to the next morning. This, of course, became standard practice.

with experiments at Chalk River in which deuterium or tritium was dissolved in the ordinary water being irradiated. The hydrogen product, containing both the light isotope, H-1, and either H-2 or H-3, depending on the additive, was then mass analyzed to determine the hydrogen isotope ratios. The mass spectrometers used at that time were home-made, temperamental machines, but were worth the trouble they gave because isotope experiments were very effective tests of theory.

Shortly thereafter, in the late fifties, it was shown from experiments elsewhere that an early step in water radiolysis is the formation of solvated electrons. Some electrons from the interaction of the gamma rays with water molecules were not recaptured by their parent ions; instead, they became associated with several water molecules. Similar hydration spheres are commonly associated with ions in water; sodium and chloride ions are examples. The solvated electrons turned out to have almost the same chemical properties as hydrogen atoms. When the computer calculations were done with reaction and diffusion rates appropriate to solvated electrons in place of hydrogen atoms, the agreement with experiment was much better than that obtained formerly, and confidence in the theory increased accordingly.

AECL radiation chemists and computer scientists have continued to cooperate in developing sophisticated programs to predict the course and outcome of chemical reactions, and the programs have found numerous applications inside and outside AECL.

In the early sixties, the radiation chemists at Chalk River turned their attention to the hydrocarbons. Several aspects of the radiolytic behaviour of these compounds of hydrogen and carbon made them interesting subjects for research. For instance, it had been known for about thirty years that benzene, C_6H_6, is more stable to ionizing radiation than its near-relative, cyclohexane, C_6H_{12}, by a factor of about one hundred. Both molecules have ring structures; benzene has alternating single and double bonds between the carbon atoms, while cyclohexane has only single bonds. It is this stability of benzene-type structure, coupled with their high boiling points, that make the terphenyls, three benzene rings bonded together in a string, effective reactor coolants. It was also known that as little as three percent of benzene in cyclohexane reduced the decomposition of cyclohexane by as much as fifty percent. This intrigued the chemists and appeared to be a form of radiation protection, a matter of special interest to radiation biologists.

The Chalk River chemists concentrated on cyclohexane solutions of benzene and other molecules with similar chemical properties. As in their research on water, they substituted deuterium for ordinary hydrogen in the compounds used in the experiments. Initially, they mass-analyzed only the hydrogen produced in the reactions, but later they were able to account for all the participating atoms. This advance was made possible by gas chromatography, a powerful technique then in its infancy, and the Chalk River chemists pioneered the application of the mass spectrometer as a detector in chromatography.

The experiments were among the first to show that positive ions and electrons are prominent reacting species in the early stages of hydrocarbon radiolysis, as elucidated in a dozen published Chalk River papers. Radiation protection in benzene-cyclohexane mixtures was shown to be due, at least in part, to a process called energy transfer. Benzene accepts energy from excited cyclohexane molecules which otherwise would decompose; benzene molecules, by virtue of their structure, accommodate the energy internally without decomposing. Thus, for a given dose of ionizing radiation, the decomposition of a mixture is less than that observed with pure cyclohexane.

Whiteshell joined Chalk River in research on radiation chemistry starting in the mid-sixties. By that time, instrumentation had advanced to the point that direct observation of the transient species in radiolysis, solvated electrons, free radicals, and so on, became possible.

RADIOTRACER STUDIES

With an excellent reactor in which to make radionuclides and the techniques for handling them well understood, it was natural for the chemists in the early days at Chalk River to apply radiotracers to their research interests. Among the tracer studies were pioneering experiments in which the rates of reaction

of organic compounds enriched in the radioactive isotope carbon-14 were compared with those containing stable carbon-12 or carbon-13; i.e., the so-called isotope effect. Mass-spectrometry techniques were later employed in such experiments and were also used in research on isotope effects in chlorine reactions. The results were used to elucidate what chemical bonds were broken or formed in the reactions (i.e., the mechanisms of the reactions), and used for comparison with theory.

Radiotracer studies with bromine-82 showed that bromine and bromate ion in perchloric acid solution undergo isotopic exchange. The data implied the occurrence of secondary reactions leading to the formation of hypobromous acid. The mechanisms of the reactions revealed in the tracer experiments were later deduced from more conventional experiments. Isotope-exchange methods were also used to compare the properties of complex ions as a function of the position of their parent elements in the lanthanide family. A similar dependence of thermodynamic properties among the alkali metals gave results, unobtainable by prior methods, for comparison with theory. Radioactive isotopes of sodium, of mass 22 and 24, were used to investigate isotope separation using an organic cation exchange resin.

In research on the effects of fast-neutron irradiations on materials, discussed below, there is often a need to know the distance travelled by an atom recoiling from a collision with an energetic neutron. Because the distances are very small and, therefore, difficult to measure, such information was very scarce. In response to this need, a method for measuring these ranges was developed from research that started at Chalk River in the late fifties.

The key was a method for stripping away a series of thin, uniform layers from a metal sheet using a "chemical milling" process. It was first worked out with aluminum and depended on forming a layer of oxide on an aluminum anode in an electrolytic bath. By controlling the conditions, the thickness of the anodic oxide could be regulated precisely and, after removing the sample from the bath, the oxide was dissolved without attacking the metal. The sample could then be returned to the bath to repeat the process as often as needed. In this way, layers only a few atoms thick could be stripped away one after the other. The method was later extended to tantalum, tungsten, silicon and gold.

For the range measurements, radiotracer ions were accelerated to selected energies in the kilovolt region using a mass separator as an ion gun. Mass separators are similar in principle to mass spectrometers, but have no beam-defining slits, so the transmission is very high. The instrument for this program was built in Sweden in 1958, and was the first step to many valuable and enduring cooperative programs with Scandinavian laboratories.

The accelerated ions were directed at the target foil and, after the bombardment, their distribution in the target was determined by measuring the radioactivity remaining in the target after successive layers were stripped away using the technique just described. To ascertain the dependence on ion and target mass, ion masses were varied from 24 to 222 and the target elements ranged in atomic number from aluminum at 13 to gold at 79. Thus, data on ion ranges were obtained for the whole region of interest. The results agreed with a theory developed at about the same time at the University of Aarhus, Denmark, and the common interest formed a strong bond between researchers there and at Chalk River.

In the course of these experiments, it was found that a few ions penetrated much farther than most into aluminum foil. This result was completely unexpected and to demonstrate that it was not an experimental artifact, work on a second method for determining ion ranges was initiated at the Nobel Institute of Physics at Stockholm during a visit by the lead Chalk River scientist in 1962. Radon-222, an isotope of radon that decays with the emission of alpha particles, was implanted into aluminum foil and the energy spectrum of the alpha particles emerging from the foil was measured. Since alpha particles lose energy in proportion to the thickness of matter through which they pass, the spectrum could be interpreted to give the depth distribution of the implanted radon. Here again, it was found that a few atoms penetrated to a greater depth than the average, thus confirming results from the electrochemical stripping experiments. The spectroscopic technique was refined at Chalk

River over the next few years and yielded similar results. These experiments and the range measurements furnished the first experimental evidence for ion channeling, a quite unexpected phenomenon that was studied intensively at Chalk River and elsewhere.

A different application of the nuclear spectroscopic technique was the measurement of the relative mobilities of oxygen and metal ions within a growing anodic oxide. Radioactive marker atoms were implanted in a thin surface oxide layer on the metal specimen using the mass separator. A marker with nearly ideal properties was xenon-135. Xenon is quite inert chemically, and the isotope of mass 135 yields a mono-energetic electron when it decays. The oxide was then grown electrolytically to produce a coat many atom-layers thick, and the position of the marker was determined at several stages during the growth by measuring the electron energy loss. If oxygen is the mobile species, the oxide forms at the metal-oxide interface and the xenon marker remains near the outermost surface. If, on the other hand, the metal ions move, growth occurs at the oxide-electrolyte interface and buries the marker. Zirconium oxide was found to grow mainly by oxygen migration, for example, while in tantalum oxide, both the species are mobile, oxygen with about twice the mobility of the metal. A number of elements in several different families in the Periodic Table were investigated, and the experiments were soon accepted internationally as yielding the first reliable solid-state marker results. The method was brought to a high degree of refinement with tantalum.

IRRADIATION EFFECTS

Reactor irradiation produces physical, as well as chemical, changes, and fundamental research on the effects of irradiation on crystalline solids was started at Chalk River about 1951. It was known that many of the effects are caused by fast neutrons and energetic nuclei, such as fission fragments, which knock atoms out of their normal crystal-lattice positions. This results in the creation of a pair of what are called point defects: the displaced or interstitial atom that becomes wedged among normal lattice atoms, and the vacancy, the hole in the lattice where the interstitial atom used to reside. Defects may migrate through the crystal and will disappear should they arrive at a sink. The important types of sink are: linear defects in the otherwise regular crystal lattice; voids, small holes in the crystal; and grain boundaries. These last are absent in perfect crystals, but some solids are composed of small crystals (e.g., ordinary metals), and the boundaries between the crystal grains are common sinks for point defects.

In the first experiments, ordered alloys of copper-gold were irradiated in NRX. Ordered alloys are characterized by regions in which the atoms of the two elements are in a fixed arrangement. Two competing effects were resolved: disordering by fast neutrons, and what appeared to be an ordering effect of thermal neutrons complicated by the transmutation of gold-198 to mercury-198.

Interstitial atoms are at a higher energy level than those on normal lattice sites, and form a reservoir of energy in the solid. Heating allows the interstitials to return to their normal positions and releases the stored energy. Measurements of the stored energy in irradiated graphite were started in 1953, and, a few years later, measurements were made on irradiated lithium fluoride and several oxides of uranium. In the latter two cases, alpha particles and tritons, or fission fragments, respectively, are responsible for displacing host atoms from their normal lattice sites.

Around 1955, Chalk River scientists began two types of experiments to measure the energy required to form a defect pair, or to move defects in the crystal lattice. In one approach, the specimen was irradiated with fast neutrons at a temperature low enough to freeze the defects. The electrical resistivity of the specimen, a very sensitive monitor of point defects, was measured as the temperature was slowly raised, thus releasing the defects. From an analysis of the resistivity changes, the energy required for a vacancy to migrate in the lattice could be deduced, and the effects of neutron irradiation could be compared with other methods for introducing point defects. In the second type of experiment, the rate of diffusion of a radioactive tracer was measured as a function of temperature of

the specimen. In the simplest experiments, the tracer was an isotope of the element being studied. Here again, the vacancy migration energy could be deduced. The first experiments of both types were made with platinum and later extended to other metals and semiconductors.

When zirconium alloys became the choice for power reactor pressure tubes in about 1957, the expertise developed in the early research activities was used to investigate irradiation effects in zirconium alloys.

ANALYTICAL METHODS — EARLY DAYS

Much of the research just described depended on measurements of the decay rates of radionuclides. In the early days, alpha- and beta-emitting nuclides were generally used in the experiments. Although it was relatively easy to compare decay rates of similar sources of a given radionuclide, comparisons under different conditions, or of different nuclides, were more difficult. Methods for improving the accuracy of these more exacting measurements received a good deal of attention. Experimenters switched to gamma-ray measurements with the introduction of sodium iodide scintillation spectrometers in the fifties, and of solid-state spectrometers about a decade later. These made possible not only the determination of the amounts of radionuclide present, but also identified them by their gamma-ray spectra.

Chalk River chemists analyzed the radionuclides in the debris from weapons tests, beginning with the first USSR nuclear explosion in 1949. The amounts were much smaller than used in the chemistry laboratories and new approaches had to be developed. The keys to success were the elimination of extraneous radioactive material from the laboratories to avoid contaminating the samples, and low background techniques; i.e., methods for reducing the radiation detected by the counters in the absence of the samples. Soon they were able to determine the type of weapon tested, and its approximate yield. Since such information was valuable to the Defence Research Board of Canada, this aspect of the work was later transferred there.

Radionuclides in the fallout entered the food chain, and the techniques were adapted to analyze dairy products, vegetables, etc., from across Canada. When procedures became standardized, this program was moved to Federal Department of Health and Welfare laboratories.

Low-background techniques were equally valuable in AECL programs and became highly developed. Building materials for the rooms in which the measurements were to be made, such as cement, sand, and paint, were chosen to minimize the radioactive content. Even the electric light bulbs used in these rooms were taken apart to remove the small amount of slightly radioactive cement used to join the glass to the metal base. The low counter-backgrounds facilitated measurements of radionuclides accidentally assimilated into the organs and tissues of AECL employees, and in environmental research.

The high neutron flux in NRX was exploited in research on activation analysis during the late fifties. In this procedure, the sample is irradiated with neutrons and the radioactive products are measured, often by comparison with standard samples of known composition processed in the same way as the sample being analyzed. High sensitivity is possible for many elements, depending mainly on their nuclear properties. Resolution of the gamma-ray spectrum of the activated sample, obtained with a sodium iodide spectrometer, for example, often permits several elements to be determined simultaneously, speedily, and without destruction of the sample. It is an especially valuable method for establishing whether samples collected in different places are from the same source. For example, by comparing the trace elements in samples of human hair, the Royal Canadian Mounted Police, in cooperation with scientists at Chalk River, obtained evidence that resulted in a criminal conviction in a Canadian court. This was the first judicial recognition of activation analysis in Canada and, so far as we know, in the world.

The Canadian nuclear program placed heavy demands on conventional methods of chemical analysis. Reactor materials required analysis for neutron absorbers, and methods for analyzing uranium fuel and aluminum sheathing for cadmium, boron, and other strong neutron absorbers had to be developed

from first principles, since standard methods did not exist. The assay of plutonium solutions was an early challenge.[2]

Several mass spectrometers for the precise measurement of atom ratios were designed and built for chemistry research at Chalk River. Work on the second instrument, specifically for the demanding work with elements of high atomic mass, was started in the spring of 1950 and commissioned two years later. Typical of such ventures, not all the applications of the instrument were clearly foreseen when the design was started. It was not until a few years after commissioning that its full value was realized. By then, it was indispensable in both the fuel-development and reactor-physics programs. In the former, the uranium-isotope ratios before and after irradiation were used to calculate the energy produced in experimental fuel. In the latter program, the accuracy of reactor-physics calculations could be tested by comparing the results with the uranium-isotope ratios actually found in discharged fuel. Plutonium mass analyses were also needed in these programs and a third machine with safety features for work with this element was built a few years later. The many high-quality fuel analyses made with these instruments were major contributions in the evolution of CANDU power reactors.

CHANNELING AND ITS APPLICATIONS

In parallel with the experimental measurements of heavy ion ranges at Chalk River, scientists at the Oak Ridge National Laboratory, United States, studied the trajectories of atoms injected into crystalline targets by computer simulation. They found that a few of their simulated atoms travelled to surprising depths in the target, apparently confined to the open spaces (channels) between the rows of atoms in their model crystal by the electrostatic forces they had included in their simulation. They suggested that, in the Chalk River experiments, some crystal grains in the polycrystalline target foils were fortuitously aligned with their channels parallel to the incident ion beam, thus allowing the ions to penetrate further into the target than those hitting randomly oriented grains. At Chalk River, the foil experiments were repeated with single crystals of aluminum, and the results quickly confirmed this hypothesis. Independent verification followed rapidly from other laboratories.

The theoretical framework for the process, which came to be called ion channeling, was developed at the University of Aarhus, Denmark, in 1964. A feature of ion channeling that surprised scientists at first, and one that makes it applicable to many fields of research, is that the yields of nuclear processes that depend on close encounters between the incident ion and the target atom fall almost to zero for channeled ions. It was surprising because, before the discovery of channeling, one would not have expected the geometrical arrangement of atoms in the target, on a scale so much greater than nuclear dimensions, to have any effect on the yield of nuclear reactions. Preliminary tests made at Aarhus in 1964, in which the lead Chalk River scientist participated, and also at AERE, Harwell, United Kingdom, were the first to demonstrate the predicted reduction in nuclear reactions induced by protons. Further experimental confirmation of the theory was obtained over the next few years.

To summarize, channeling occurs whenever a beam of positively charged nuclear particles is incident on a crystal in a direction parallel to prominent rows of atoms forming the crystal lattice; in crystallographic terminology this is called a low-index direction. The particles are steered on helical paths through the channels between the rows by the

[2] These analyses were of more than academic interest. AECL sold the irradiated fuel rods from NRU to the United States for a price based on the plutonium content at about $100 per gram. The rods were dissolved in the United States, and samples of the solutions were analyzed there and at Chalk River. The American analysts used a method based on measuring the plutonium disintegration rate by counting the alpha particles. At Chalk River, the absorption of light by the solution, which is proportional to plutonium concentration, was measured spectrophotometrically. The latter were consistently higher, and the Canadian chemists managed to convince their American counterparts that their method was the more accurate one. Winning the debate was worth about $9 million to the Canadian program.

concerted effects of the electrostatic fields of hundreds of atoms forming the channels.[3]

As understanding of the channeling process improved in the late sixties, possibilities for applying it to other fields of research began to receive attention. At about the same time, a 2-MV Van de Graaff accelerator was installed at Chalk River for use in this program and in radiation chemistry. The first application was in research on ion implantation of semiconductor materials, and was launched during a visit from the head of the implantation group at Hughes Research Laboratories, Malibu, California.

In the joint experiments, each process used to make semiconductor electronic devices was investigated. Ions of the element required to dope the semiconductor and give it the desired electrical properties were implanted using the mass separator. During this process, the ions knock the atoms of the crystalline semiconductor from their normal lattice positions and produce point defects. If the implantation is continued, the concentration of defects becomes so great that they merge and form an amorphous layer. This damage to the perfection of the crystal structure impairs the electrical properties of electronic devices and is therefore undesirable. Point defects and amorphous regions in a crystal also adversely affect the channeling process. In the joint experiments, it was shown that channeling techniques could give a quantitative measure of the damage produced in semiconductor crystals during ion implantation.

The next stage in device fabrication is to heat the implanted crystal for a period of time sufficient for the damaged region to re-crystallize using the undamaged portion as a template. Joint experiments showed that it was also possible to follow this mechanism by channeling measurements. In addition, they gave the ratio of dopant atoms on crystal lattice sites to those in interstitial spaces in annealed devices.

The experiments resolved many questions about the use of ion implantation for fabricating semiconductor devices, and proved to be an ideal subject for testing the applicability of channeling in solid-state research. This valuable collaboration continued for nearly twenty years, although the Hughes scientist changed affiliations during the period. Other laboratories quickly embraced the channeling technique in related research, and by 1973, its use was reported from around the world in over one-thousand technical papers.

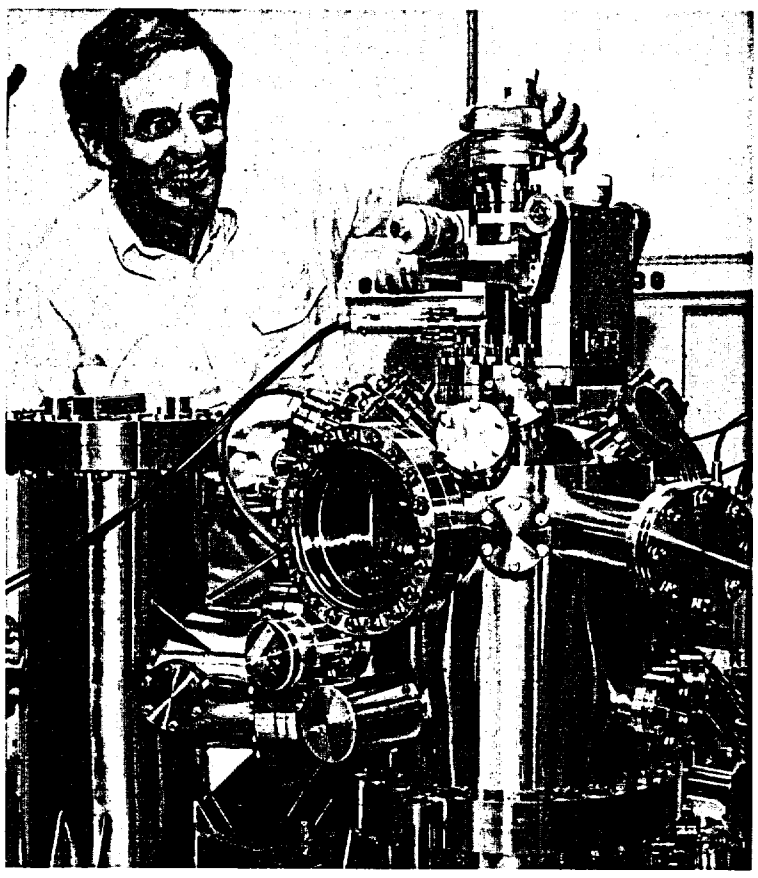
Chalk River scientists were the first to show that channeling is a powerful technique for the study of point-defect behaviour in alloys. Crystal-lattice vacancies are often found in association with the dilute, or solute, component of an alloy, and may influence the positions assumed by solute atoms in the crystal lattice. When these atoms are in, or encroach on, interstitial spaces, channeling techniques can often reveal the configuration of the solute atom-defect cluster. Numerous alloys were studied throughout the seventies, and several types of cluster were investigated, the most complex example of which is probably the tetrahedral arrangement of vacancies around interstitial indium atoms in aluminum crystals.

Beginning in 1974, channeling was used to augment more usual surface-science techniques in research on metal catalysts of the type used for hydrogen isotope exchange (see figure 7.1). Noteworthy results include information about the arrangement of atoms on catalyst surfaces, and correlations between surface structure and the extent to which the surface is covered by adsorbed gases.

In addition to the above, channeling has been applied at Chalk River in several areas of research of somewhat narrower scope. These topics include the measurement of nuclear lifetimes;

[3] In the late sixties, the federal cabinet minister responsible for AECL at the time, the Honourable Jean-Luc Pépin, paid an official visit to Chalk River and was briefed by the staff about program highlights. For a while he listened patiently to an account of the discovery of channeling, which was described with the aid of a "ball-and-stick" model of a simple crystal. Soon he interrupted, however, to exclaim: "Isn't that exactly what one would expect from looking at the model? Why are you so excited about it?" In such crystal models the open spaces between rows of atoms are quite obvious. The preferential penetration of protons in these low-index directions had, in fact, been predicted as early as 1912. The discovery of X-ray diffraction at about that time and the revolution in physics then in progress diverted attention from simple electrostatic steering responsible for channeling. The difficulty of measuring the short ranges of ions in solids was an added impediment.

FIGURE 7.1
Scientists at CRL used this ultra-high-vacuum equipment on a beam line from an ion accelerator for research on the structure of layers of gases adsorbed on catalyst surfaces.



the systematics of heavy-ion stopping powers; inner-shell ionization arising from ion-atom collisions; and the steering of GeV protons by bent single crystals.

MATERIALS SCIENCE

Materials science is at the boundaries of physics, chemistry and, when metals are studied, metallurgy. As the name implies, the subjects for research have practical applications. In 1957, when Zircaloy-2 was first considered for pressure tubes, zirconium alloys became the structural materials of greatest importance to AECL. Materials science covers the spectrum from fundamental studies at one end to applied research at the other. Work of the latter type in AECL is described in chapters twelve and thirteen, while the former is covered herein, although the division is not sharply drawn here, nor was it in the laboratories in which the research was done.

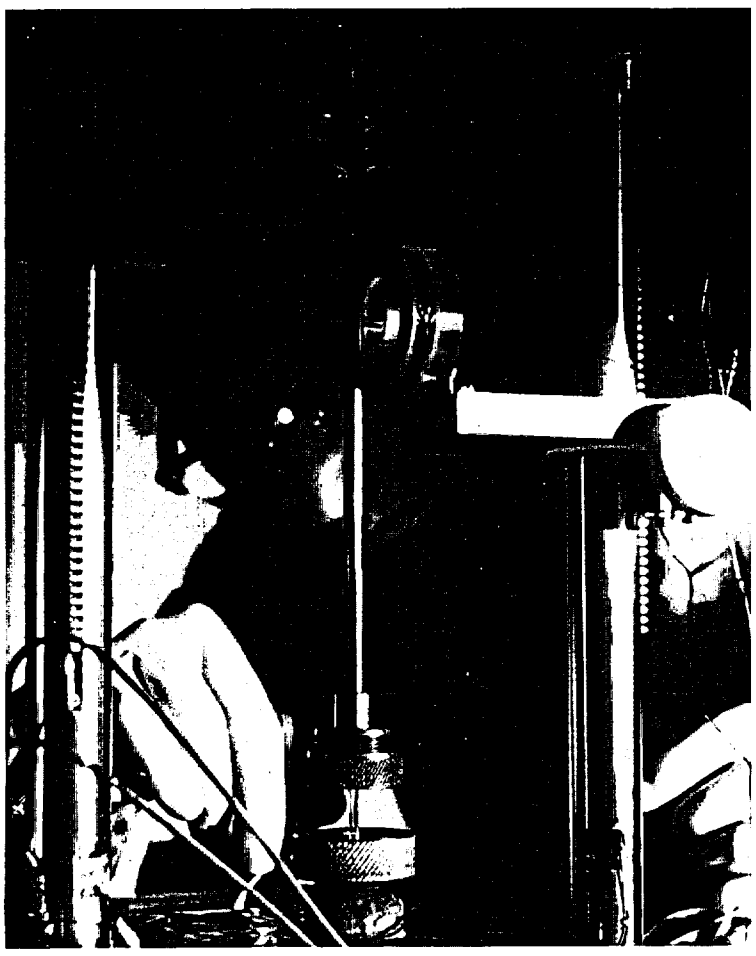
Zirconium was a "new metal" in 1957, attractive to reactor designers because its neutron-capture cross-section is lower than the cross-sections of other strong, corrosion-resistant metals, and there was much to learn about its fundamental properties. It was known that reactor irradiation produces point defects, and the subsequent behaviour of the defects modifies the physical properties of the material. Initially, therefore, much of the effort was aimed at establishing techniques and adapting them to investigate point-defect behaviour in this new metal.

The first experiments used resistivity and tracer diffusion methods to explore defect properties. By the early sixties at Chalk River, and a few years later at Whiteshell, transmission-electron-microscopy techniques were being used. Scanning-electron-microscopy was added in 1970.

The interaction between point defects and dislocations has been studied at Whiteshell since 1970 using the internal-friction technique. This technique depends on very precise measurements of oscillatory vibrations set up in the specimen (see figure 7.2). An example of its application is mentioned later, when zirconium hydride is discussed. Theoretical and computational methods have also been developed at Whiteshell to predict the precise arrangement of the atoms around defects and dislocations.

The use of channeling to determine the configuration of point defects in the neighbourhood of solute atoms has already been touched on in the previous section. Yet another technique introduced at Chalk River in the early eighties depends on the positron-electron annihilation phenomenon, discussed in chapter eight. Positrons tend to survive longer at vacancies than in the perfect crystal, because the electron density is usually lower there. Thus, lifetime measurements reflect vacancy behaviour.

FIGURE 7.2
The extremely small amount of energy required to move point defects in a crystal of reactor-irradiated magnesium oxide (shown at the bottom of the photograph) was deduced by scientists at WL from measurements made with this oscillating pendulum device.



The results accruing from these studies are presented in some two hundred technical papers. Over fifty are directly related to zirconium and its alloys, and contributed to the international store of knowledge about the basic effects of irradiation on these materials.

Pressure tubes in the CANDU reactors sag and elongate in service, as described in chapter twelve. As discussed there, experimental measurements in NRX and NRU were difficult and time-consuming, and the specimens became highly radioactive. Around 1967, complementary approaches to the study of the processes fundamental to these shape changes were started at Chalk River and Whiteshell. Over the next two decades, the effects of electron and heavy-ion, as well as fast-neutron, irradiations were investigated. In addition, the theory of the processes was explored at both sites. By drawing on the experience and knowledge of irradiation effects accumulated over this period, AECL scientists and engineers have, to a steadily increasing extent, based predictions of pressure-tube-shape changes on sound physical principles.

Tests showed that Zircaloy-2 has good resistance to corrosion by water at the operating temperatures of power reactors over a period of a few years. The pressure tubes in CANDU reactors, however, were expected to last much longer, and to try to anticipate deleterious long-term effects, and understand them should any arise, fundamental research on the mechanism of corrosion was done at Chalk River from 1963 onwards.

It was known that in the early stage of zirconium corrosion, oxygen from the water migrates through the surface oxide film to the underlying metal; electrical neutrality is maintained by a counter-flow of electrons. The process was explored in detail using oxygen-17 as a tracer, and the $^{17}O(^{3}He,^{4}He)^{16}O$ nuclear reaction to follow the migration of the tracer during the corrosion process. With polycrystalline specimens, it was found that the rate of diffusion was a thousand times faster through the boundaries between oxide crystallites growing on adjacent crystallites in the metal than through the main body of the oxide. Incidentally, the novel tracer technique used in these experiments is an example of the type of cooperation possible at Chalk River. The chemists were able to satisfy the physicists' requirement for a stable target of oxygen-17 for experiments with the tandem accelerator, and the physicists provided the helium-3 beam from the tandem for the diffusion studies.

The counter-flow of electrons during corrosion was found to depend on the type of alloy and local environmental conditions. A hypothesis embracing both the oxygen and electron observations, and explaining the effects of reactor irradiation on the oxidation rate, has been developed. Additional results indicate that the later stages of corrosion proceed by oxygen migration through pores in the oxide layer, and factors affecting these processes were also established.

Zirconium and its alloys absorb hydrogen, and brittle platelets of zirconium hydride form in the metal if the amount exceeds solubility limits. The formation and orientation of such platelets through stress-induced diffusion resulted in the cracks in some pressure tubes, first observed in the Pickering-3 reactor in 1974. It turned out these tubes had been installed incorrectly. The behaviour of hydrogen in zirconium had been investigated in AECL laboratories since about 1958, and scientists were quickly able to provide a qualitative understanding of the fundamental mechanism by which these cracks developed in service. Within a few weeks, the mechanism was placed on a sound quantitative basis, but the importance of the topic has meant that much additional refinement of the theory has been considered essential to the CANDU program.

The cladding on CANDU reactor fuel is also based on zirconium. In addition to the environmental factors already mentioned, cladding may experience stress-corrosion cracking (SCC): i.e., attack by the fission products that accumulate in the fuel elements, coupled with mechanical stress. The intense investigation organized to find the cause of fuel-element failures, first appearing at Douglas Point in 1971 and later at Pickering, showed that SCC was responsible for many of the failures. Fundamental research on the process started at Chalk River in 1971 and contributed to the team investigation reported in chapter thirteen. As described there, the evidence points to fission-product iodine as the active agent, and a remedy was found to counteract its attack.

Although zirconium is no longer the new metal it was in the fifties, there remains a good deal to learn about the fundamentals of its alloys. Since they are extensively used in CANDU reactors, this area of materials science is particularly important to AECL.

RADIATION CHEMISTRY — LATER PERIOD

The first of the instrumental advances for research on radiation chemistry anticipated earlier in this chapter was the electron-spin-resonance (esr) technique introduced at Chalk River around 1963. As already discussed, the free radicals and neutral atoms, so common in chemical reactions induced by high-energy radiations, possess an unpaired electron. The electron spins as it orbits its parent nucleus, in roughly the same way the earth turns on its axis as it circles the sun. The esr spectrometer searches out resonances between the energy states of the spinning electron and an applied electric field that can be scanned across the microwave region, in the presence of a strong magnetic field. Research in other laboratories had shown that the resonance spectrum can, under favourable circumstances, be used to identify the chemical species containing the unpaired electron.

A wide variety of compounds was studied at Chalk River, including olefins, acetone, several different alcohols, aqueous solutions, nitrous oxide, and amines. From the esr spectra, the intermediate species produced by radiolysis could often be deduced, including free radicals, neutral atoms, and trapped electrons. The results confirmed the production of reactive intermediates postulated to be present on the basis of less direct methods, or introduced new and more explicit evidence. With knowledge of the intermediates, understanding of the course of events leading from the initial absorption of radiation to the final chemical products was greatly improved.

The esr technique was introduced at Whiteshell about 1967 and continued very successfully thereafter. In 1969, the program at Chalk River was terminated in favour of research on isotope-separation processes. More information on the Whiteshell program is provided later in this section.

The first accelerator at Chalk River for pulse radiolysis research was commissioned in 1968. In this machine, each pulse was formed by charging a bank of capacitors in parallel and discharging it in series. The resultant pulse was applied to the field emission cathode of a vacuum tube. The resulting electrons were directed through a thin window at currents of about

4000 A and energies up to 1.6 MeV. The radiation dose rate delivered to the gas in a cell placed in front of the window was up to two-hundred billion times the dose rate from conventional gamma-ray sources. From a radiation chemistry viewpoint, there are several advantages to accelerators of this type. They can produce concentrations of free radicals approaching in strength those of the reagents commonly used in chemical research, and reactions between radicals that do not occur at low dose rates are frequently observed. Furthermore, reactions of ions, commonly encountered in the radiolysis of gases, are usually much simpler than at low dose rates. These factors simplify the interpretation of the results.

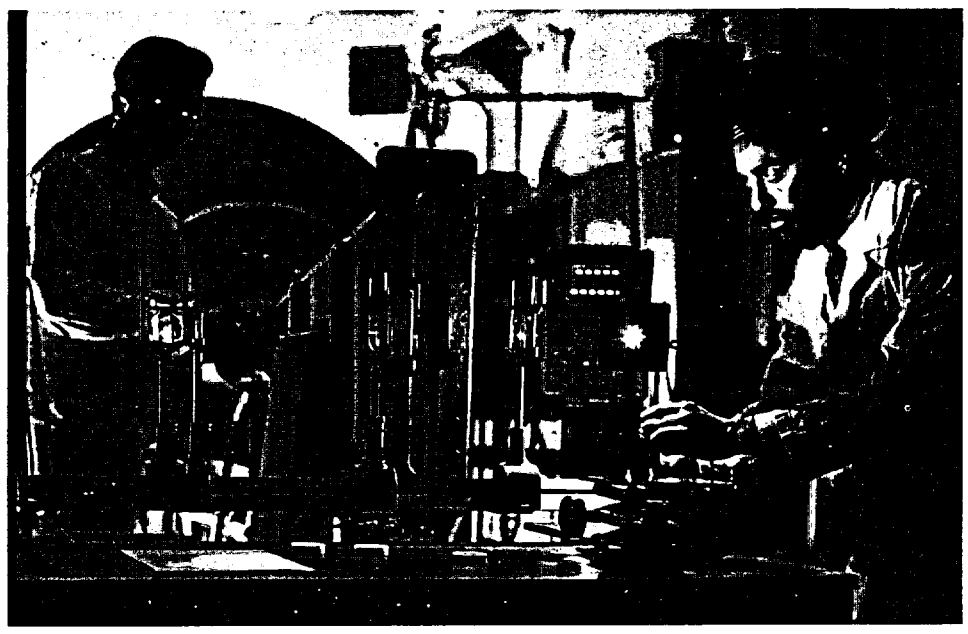
Chalk River was one of the few laboratories to use such an accelerator and the scientists made many contributions to the understanding of the radiation chemistry of di- and tri-atomic gases, including oxygen, nitrogen, water, carbon dioxide, carbon monoxide, hydrogen chloride, and others. In addition, they deduced the rate constants for many of the reactions of ions in which the electrons are in different states of excitation. These data are useful in other fields, such as upper-atmosphere chemistry, and pulse radiolysis is a fruitful method for obtaining them.

A pulse radiolysis technique of a more conventional type was introduced at Chalk River in 1969, and at Whiteshell in 1972, using electrons from accelerators of the Van de Graaff type (see figure 7.3). Although the pulses were less intense than those from accelerators in the capacitor-discharge class, the electron energies were higher, and with greater penetrating power, experiments with liquids could be made.

At Chalk River, the major interest was in the radiation chemistry of amine solutions of the alkali metals. Of special note was the chemistry of three remarkable species that exist in equilibrium in such solutions: the solvated electron, the aggregate molecule, and the metal anion, M^-, where M stands for lithium, sodium, potassium, or cesium. The aggregate molecule was thought to be a complex of the metal cation, M^+, and the solvated electron, and hence is electrically neutral, but the structure was controversial. The metal anion, it was generally agreed, is formed by the addition of a solvated electron to the aggregate molecule. The chemistry is similar to the solution chemistry of the hydrogen-amine process for heavy-water production, and the research enlarged the community of chemists at Chalk River familiar with the field. The advantage of radiolysis is that it produces higher concentrations of these species than occur in ordinary solutions and the work considerably expanded the basic understanding of their chemistry.

At Whiteshell, the focus was mainly on the chemistry of the free radicals and solvated electrons produced in radiolysis, and a wide variety of compounds was studied. Of particular note

FIGURE 7.3
The lifetimes of transient chemical species were measured at WL using optical spectroscopy synchronized with pulses from the Van de Graaff accelerator in the background.



were the direct observations of several of these species using a combination of esr and pulse-radiolysis techniques. The beam tube of the accelerator was extended through the massive pole piece of the electromagnet of the esr spectrometer, and a flow system was used to continuously refresh the liquid being irradiated and flush out the products. In this way, it was possible to characterize the radicals produced in various alcohols and aqueous solutions. Information about the solvated electron in water was also refined in experiments in which each electron lived only ten microseconds on average.

A fundamental aim in all these pulse radiolysis experiments was to push the understanding of the chemical processes closer and closer in time to the primary interaction of radiation with matter. Research at Whiteshell provided the culminating AECL example. The experiments were made to test a theoretical prediction of the effects of changes in magnet field strength on the light emitted by the molecular fragments in liquids during their brief existence – less than a microsecond – following irradiation pulses. The theoretician, from the University of Sheffield, and the experimentalists cooperated during a summer appointment of the former in 1974, and after refinements to both the theory and the experiments, the sought-for agreement was found. Further investigation of the effect and its applications have been made at Whiteshell and other laboratories around the world.

HIGH-TEMPERATURE SOLUTION CHEMISTRY

High-temperature water is an important constituent of CANDU reactors and heavy-water plants. Its presence is also invoked in the analysis of potential reactor accidents and in predictive studies of how radionuclides might migrate from geological disposal vaults. Fundamental data about aqueous solutions at high temperature are scarce, however, because the measurements are difficult to make. Not only are high pressures required to keep water liquid at high temperatures, hot water is very corrosive. Chemists at Whiteshell began to contribute to knowledge in this field in the early seventies, and have made many contributions during the period of this review.

Their program had two complementary parts: experimental on the one hand, and predictive or theoretical on the other. The approach was based on thermodynamics, the science of heat energy, and on chemical thermodynamics, in particular. This branch of the science deals with heats of chemical reactions, heats of solution, and other measures of the forces driving chemical changes.

The initial focus was on the thermodynamic variables of compounds of the metals present in process streams of reactors and heavy water plants; e.g., iron, cobalt, and nickel. Several methods for extrapolating room-temperature data to temperatures up to 300°C were advanced. Electrochemical measurements were used to determine some of the basic data and to confirm some of the extrapolated values. The compounds of interest dissociate into pairs of oppositely charged ions when they dissolve. From the experimental results, the researchers could deduce characteristic properties for both members of the pair in many of the compounds. These data are useful in their own right and for comparison with theory. Furthermore, the chemists used them to deduce the properties of additional single ions from conventional thermodynamic measurements.

As the waste-management program advanced, the techniques were used to predict the high-temperature properties of about one-hundred different species of the actinide elements in solids and high-temperature water solutions. The solubility of uranium dioxide was measured up to 300°C and the behaviour of typical glass-waste forms in high temperature water was studied. Chapter nineteen describes how such basic information contributes to the waste-management program.

The techniques were also used to determine the properties of compounds of some fission-product elements at elevated temperatures. The work on technetium compounds was of special scientific interest. This element, atomic number 43, does not occur naturally, but is produced in good yield by fission of uranium. Thus, the Whiteshell chemists were able to study compounds of an element not available to many others. Primarily, however, the fission products were studied for their relevance to waste management and reactor-safety analysis. In addition to technetium, these included iodine, selenium and palladium. In

reactor safety analysis, it is postulated that some reactor accidents may result in the release of fission products and high-temperature water into a reactor's containment building. Research on the fission-product elements, particularly iodine, which has complex chemistry, contributed to the knowledge necessary to anticipate the chemical consequences of such hypothetical accidents.

Thus the research results contributed to basic knowledge of a wide selection of chemical elements in high-temperature water, and were quickly applied in several AECL programs. They are also applicable to other Canadian industries, such as mining, and pulp and paper.

ISOTOPE CHEMISTRY

Chapter seventeen discusses the alternatives to the Girdler Sulfide (GS) process that were investigated for making heavy water. In parallel with the applied research-and-development work described there, some basic studies on two of the alternatives, the amine-hydrogen and the hydrogen-water processes, were undertaken beginning in the late sixties. For example, fundamental to the latter process is the deuterium-isotope separation factor between hydrogen and water. This factor was measured at Chalk River over a wider temperature range and with greater precision than reported by experimentalists in other laboratories. The results not only met AECL needs, but led to a refinement of the theory for the calculation of the equilibrium constant for the reaction from first principles that resolved a long-standing discrepancy with experimental determinations.

For practical application, both processes require a catalyst. In the amine-hydrogen process, the catalyst is dissolved in the amine and the chemistry is quite complex. The mechanism by which the catalyst acts, and the nature of unwanted side reactions that remove catalyst, were investigated. In the hydrogen-water process, the reactants pass over a solid catalyst, and both water and hydrogen molecules must be adsorbed on the surface of the catalyst for isotope exchange to occur. Other molecules compete for space on the surface and may deactivate the catalyst. Research done throughout the seventies, using some very advanced surface-chemistry techniques, showed how these reactions happen. Towards the end of the period and continuing to 1985, ion channeling was used to complement more conventional surface-science techniques. At Whiteshell, water-soluble catalysts for the hydrogen-water process were investigated from a fundamental point-of-view as alternatives to the surface catalysts developed at Chalk River.

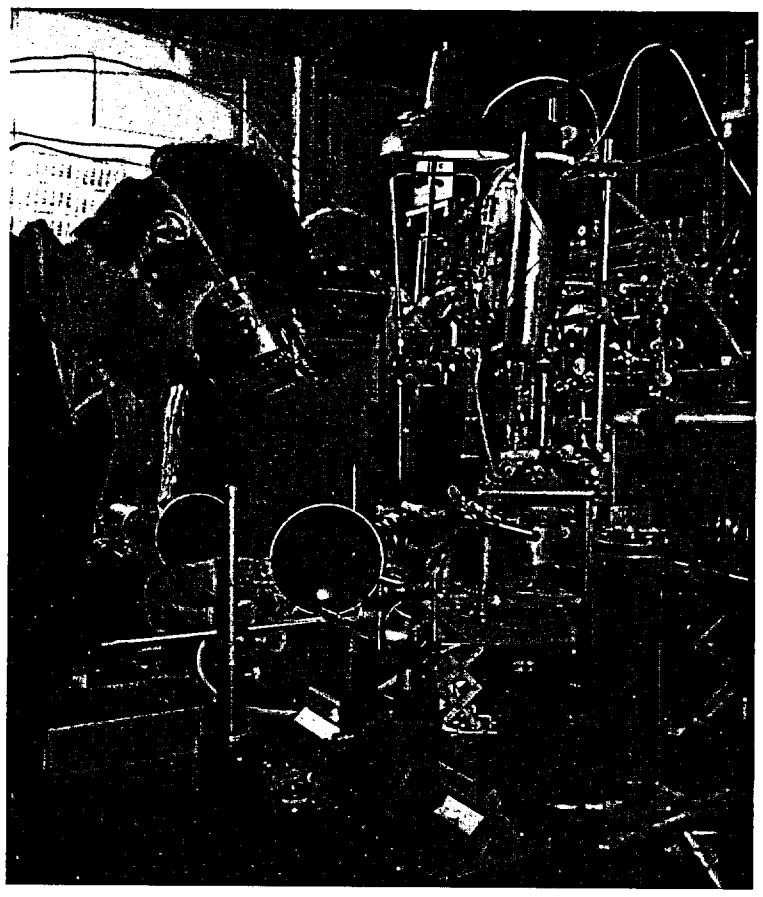
A heavy-water upgrading plant operated at Chalk River based on deuterium enrichment by water electrolysis. To provide scientific support for the process, the effects of electrochemical variables were studied in the laboratory. With the discovery of a promising surface catalyst for the hydrogen-water process, a natural extension was to combine the two processes, and this led to the Combined Electrolytic-Catalytic Exchange process.

By around 1975, worldwide advances in laser technology, and the understanding of the interaction of laser light with chemical compounds, had advanced to the stage that isotope separation using lasers might be considered possible. To stay abreast of these developments, a research program on laser isotope separation, initially centred on deuterium, was launched at Chalk River; it continued during the balance of this history.

High-powered lasers can pump many quanta of light into target molecules almost simultaneously (see figure 7.4). Because the light-absorption spectra of isotopes of elements in the target differ, the large amount of energy deposited by the laser may be associated preferentially with a particular isotope. This concentration of energy may, in turn, lead to dissociation of the excited molecule at the site of the selected isotope. Following separation, the isotope can be swept up by chemical reaction or other means.

Hydrogen fluoride and carbon-dioxide lasers were used with quite a variety of simple molecules in vapour form: formic acid, methanol, methylamine, fluoroform, and related compounds. The effects of collisions between the excited target molecule and unexcited molecules, which tend to deactivate the target molecule, were important issues. The work led to AECL

FIGURE 7.4
Research on isotope separation using high-powered lasers at CRL required the complex equipment shown here, and stringent safety measures.



research contracts with Canadian universities, and cooperative experiments in which expertise was shared with scientists at several laboratories in Canada and the United States. Possibilities for separating isotopes of other elements were assessed: e.g., removal from zirconium of the isotope that is the main contributor to the neutron cross-section of the natural element, for use in pressure tubes; uranium-235 for enriched fuel; and isotopes for medical applications. Although a laser-based heavy-water process remains an elusive economic goal, a laser was used to enrich the deuterium in a few drops of water nearly 2,000-fold (i.e., from 0.015 percent to 30 percent) in a laboratory tour de force.

ANALYTICAL METHODS — LATER PERIOD

As programs progressed and new ones arose, analytical methods advanced in parallel to meet new requirements at Whiteshell and Chalk River. With the growth of the fuel-development program, faster methods evolved to meet the increased demand for analyses to help determine the energy output from test specimens and, in mixed fuels, apportion shares to the appropriate isotopes of uranium, plutonium and thorium.

Water and organic coolant in the circuits of operating reactors were analyzed for the trace elements responsible for the generation of radioisotopes that are then carried outside the reactor shielding by the coolant. These methods were also used to analyze water in waste-management studies. Special attention was given to the determination of several elements simultaneously in the same sample or, where practicable, continuously in liquid or vapour streams. In a similar vein, the deuterium content of process streams at heavy-water plants and in the research laboratories was measured on-line.

Advanced techniques were used to analyze the surfaces and near-surface layers encountered in research on catalysts and corrosion. The solutions of the organic chemicals used to decontaminate reactor circuits required analyses to maintain them at efficient and economic operating levels. The amine-hydrogen process for heavy water raised unusual requirements for the analysis of organic-nitrogen compounds. Investigation of pressure-tube hydriding required the determination of low levels of deuterium in many highly radioactive specimens cut from tubes removed from the Pickering reactors.

Requirements for the analysis of one-of-a-kind samples from physics, engineering, or environmental programs were not uncommon and often required small research programs for their completion. An unusual example of this capability was the work done at Whiteshell on the fragments of the Russian

Cosmos nuclear-powered satellite, which fell to earth in northern Canada in 1978. The whole repertoire of chemical, isotope and surface analytical techniques was applied to determine the hazard from the reactor parts and help the authorities decide on the Canadian response.

EPILOGUE

The eighties was a period of transition and change in AECL. In materials science and chemistry, high-priority work for the Pickering reactors, waste management, and commercial activities demanded attention from some of the scientists normally engaged in basic research. Also, budgetary pressures increased, and funding for the sophisticated equipment for work at the frontiers of these fields became scarce. These factors combined to make it a difficult time for basic research in chemistry and materials science.

Several steps were taken to alleviate the difficulties. Although always a feature of the laboratories, special emphasis was placed on the development of cooperative programs with other institutions. Thus, visiting-scientist appointments at Chalk River and Whiteshell were advantageous, and part-time and sabbatical arrangements for AECL staff at other laboratories were also helpful. To meet the needs of materials-science experiments, irradiation facilities were rented in a British reactor. These measures helped to keep the programs thriving, but the squeeze on basic research continued.

In late 1985, management concluded that the pressures could be sustained no longer. It was decided that underlying research in chemistry and materials science should be discontinued as discrete programs. AECL made arrangements for the continuation of some research arising from the channeling phenomenon at a Canadian university. These included the transfer of staff and necessary accelerator equipment, and the provision of financial support for several years. The balance of the research staff transferred to related applied fields or to commercial activities.

Rapid change continued, and by the end of the decade conditions were such that some projects in the underlying research category could be launched. At the time of writing, they continue at both the Whiteshell and Chalk River laboratories.

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