

Literature Review of the Effects of Radiation and Temperature on the Aging of Concrete

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ABSTRACT

The open literature and accessible United States Department of Energy-sponsored reports were reviewed for the effects of radiation and temperature on concrete. No effects of radiation were found for exposures less than 10^{10} neutron/cm² or 10^{10} Gy gamma for periods less than 50 years. Reductions in compressive and tensile strength and a marked increase in volume are reported for exposures greater than 10^{20} neutron/cm² or 10^{10} rads of gamma. There are conflicting reports of damage for doses in the middle ranges.

SUMMARY

Concrete has been used in the construction of nuclear facilities because of two primary properties, its structural strength and its ability to shield radiation. Concrete structures have been known to last for hundreds of years, but they are also known to deteriorate in very short periods of time. The use of concrete in nuclear facilities for containment and shielding of radiation and radioactive materials has made its performance crucial for the safe operation of the facility. The accessible literature was searched for reports on the effects of radiation and temperature on concrete deterioration.

The effects of low doses, $<10^{10}$ neutron/cm² or $<10^{10}$ Gy gamma, of radiation over periods of less than 50 years do not seem to have a significant effect on the concrete. Longer-term exposure, over 100 years, has not been studied.

The effects of higher doses of radiation are not as clear, and some difference in reported results exists. For some concrete types, neutron exposures $>10^{10}$ neutron/cm², are reported to cause deterioration.

For high radiation exposure, $>10^{20}$ neutron/cm² or $>10^{10}$ rads of gamma, concrete has been reported to exhibit reduction in compressive and tensile strength and a marked increase in volume.

The effects of long-term exposure of concrete to elevated temperatures are a loss of water in the concrete leading to a decrease in compressive strength, changes in the modulus of elasticity, creep resistance, conductivity, and diffusivity. Generally speaking, the threshold of degradation is 95°C, and the effects increase with increasing temperature and time exposure.

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ACRONYMS

ANS	American Nuclear Society
ANSI	American National Standard Institute
ASME	American Society of Mechanical Engineers
CFR	Code of Federal Regulations
DOE	U.S. Department of Energy
GALL	Generic Aging Lessons Learned
Gy	Grey, a unit of radiation. Equal to 100 rad
INEEL	Idaho National Engineering and Environmental Laboratory
Rad	Radiation absorbed dose
SNF	spent nuclear fuel
SSC	Structures, systems, and components

Literature Review of the Effects of Radiation and Temperature on the Aging of Concrete

1. INTRODUCTION

Available literature was reviewed for information pertinent to the effects of radiation and temperature on the deterioration of concrete. The accessible literature was searched for the following data on concrete deterioration.

- Concrete degradation
- Radiation effects on concrete
- Temperature effects on concrete
- Concrete aging.

The review covered the available open literature such as accessible U.S. Department of Energy (DOE) reports, available reports from user organizations (i.e., the American Concrete Institute), standards organizations (i.e., the American Society of Mechanical Engineers [ASME]), and professional organizations (i.e., the American Nuclear Society [ANS]). In addition, personal contacts were made with appropriate researchers at the Idaho National Engineering and Environmental Laboratory (INEEL) and other DOE sites.

1.1 Background

Concrete has been used in the construction of nuclear facilities because of two primary properties, its structural strength and its ability to shield radiation. Concrete structures have been known to last for hundreds of years, but they are also known to deteriorate in very short periods of time when exposed to deteriorious physical and chemical environmental conditions.

The use of concrete in nuclear facilities for containment and shielding of radiation and radioactive materials has made its performance crucial for the safe operation of the facility. Spent nuclear fuel (SNF) is stored in concrete structures, casks, and vaults for planned storage up to 40 years. This concrete is exposed to several conditions that have been shown to cause the concrete to deteriorate. These conditions include: freeze/thaw, heat, cracking, acids, chlorides, sulfates, carbonation, calcium leaching, and radiation. These conditions are compounded by the aging of these concrete structures.

The effect of radiation on concrete was studied between 1944 and 1956, and a literature review report was published.¹ The data reported came from two types of tests, small exposed samples irradiated in high radiation fluxes and examinations of concrete cores removed from reactor shields, which had accumulated a significant amount of radiation (see Reference 1). Since that time, there have been additional studies published that have added data. No comprehensive studies have been published that incorporate all of the studies into one theory or conclusion. No studies could be located that addressed the performance of concrete in SNF storage facilities.

Several organizations have recognized the importance of understand the long-term performance of concrete and developing the ability to project lifetimes of concrete. These include the U.S. Nuclear

Regulatory Commission, the International Atomic Energy Association, the American Concrete Institute, and the Electric Power Research Institute.

1.2 Technical Work Scope

The Central Research Institute of Electric Power Institute requested that the INEEL perform a literature search to identify papers and data related to temperature and radiation effects on concrete. Particular interest was to be paid to SNF storage. INEEL researchers prepared an annotated list of sources and this report. They provided a limited analysis of this information and identified key references on information and data that evaluates concrete performance for an extended period of time.

2. RESULTS

2.1 Status of Existing Research

Long-term degradation of concrete structures under permanently humid environmental conditions is mainly influenced by interacting chemical and mechanical processes leading to the destruction of the microstructure by the dissolution of cement constituents and the propagation of micro-cracks. Mechanical damage accelerates the chemical degradation as a consequence of improving transport properties.² Many of the concrete degradation processes have several potential causes, and it is difficult to separate which causes are responsible for observed damage. Early reported research results, for example, were unable to distinguish between damage caused by heat and damage caused by radiation (see Reference 1).

Since the year 2000, there have been many reports published about the deterioration of concrete. Some of these have reported the results of experimental work, and others have proposed models that might be used to predict the deterioration of concrete over periods of time. Many of the predictive models have focused on the micro-scale processes. Few of these have addressed the effects of radiation on the concrete. None of them have addressed the effects on radiation on concrete used for the dry storage of SNF.

The INEEL reviewed several hundred reports as a result of the search. Forty documents were determined to be pertinent to the request. In the sections below, various physical and chemical factors are identified, and the results of the research are presented. Included in this review are also references to concrete degradation from mechanisms other than radiation and heat that might be pertinent to the study of long-term dry storage of SNF in concrete structures.

2.2 Physical Processes

2.2.1 High Temperature

Tests on the effects of long-term exposure to elevated temperatures showed that the compressive strength in general tended to decrease with increasing temperature and with length of exposure. Temperature affects the modulus of elasticity.³ The moisture content in the concrete near a hot surface decreases because of pressure-induced flow.⁴ Heating of refractory concrete causes physical and chemical changes mainly because of removal of water. The compressive strength is reduced after exposure to about 540°C.⁵ In conventional concrete, long-term exposure to high temperatures can cause changes in compressive strength, modulus of elasticity, creep resistance, conductivity, diffusivity, and shrinkage/expansion characteristics. Generally speaking, the threshold of degradation in the concrete is approximately 95°C (see Reference 5) In some of the investigations, radiation led to an increase in temperature of the concrete up to 250°C. Such a temperature increase may cause considerable damage to the concrete even if there is no radiation effect (see Figure 1).⁶ The effect of temperature is even more pronounced on the tensile strength of concrete. A temperature increase from 20 to 100°C may cause a reduction of the tensile strength of concrete by as much as 50%.

2.2.2 Cracking

The main distress of concrete components, which were identified where life extension of nuclear plants was being considered, were cracking. Cracking occurs virtually in all concrete structures.⁷ Seventy-five percent of all reported degradation age effects were caused by cracking (see Reference 3). Several papers have reported the development of models that could be used to predict the performance, i.e., fracture, cracking, crack growth, temperature effects, and stress of concrete.^{8,9} Most of

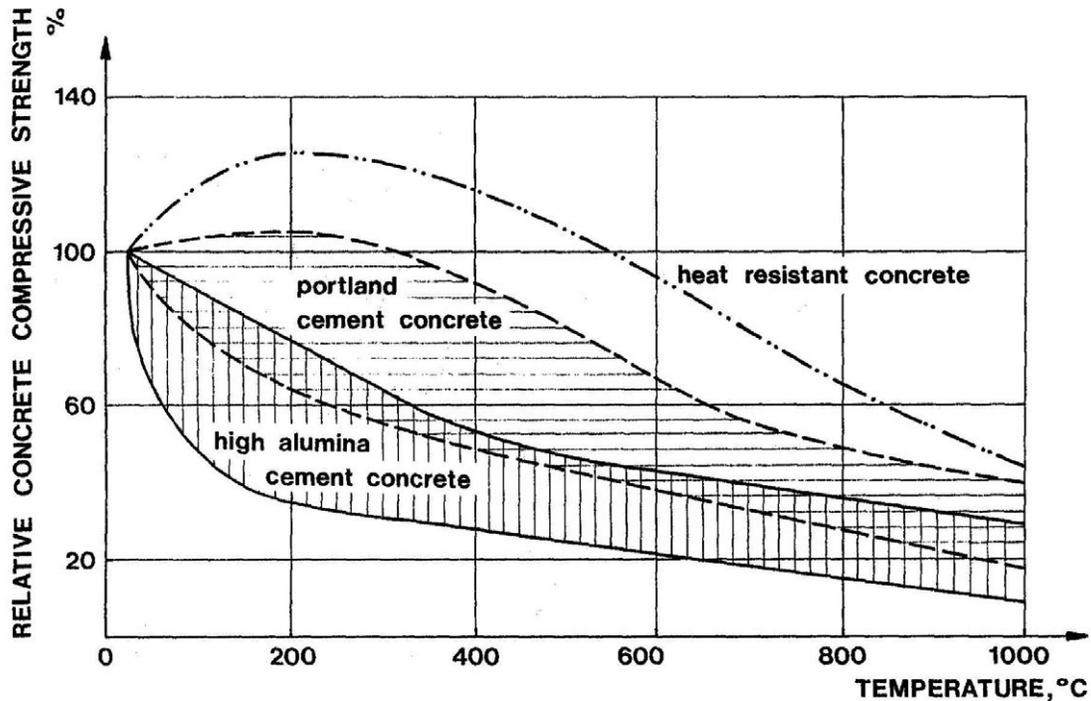


Figure 1. Compressive strength of concrete as a function of temperature (see Reference 6).

these papers also reported the values of the constants that were used in the models as functions of temperature and stress. In 2002, a 3-D lattice type fracture model for concrete was reported.¹⁰ This obtained more accurate representation than the results observed in actual physics experiments. The cracking of concrete can reduce its ability to shield radiation (see Reference 3). Cracks, irrespective of their origin, have a considerable influence on the moisture permeability of cementitious materials. As a consequence, the transport of aggressive substances is promoted, and the degradation process is further accelerated.¹¹

2.2.3 Radiation

There are two possible causes of damage to concrete by irradiation. One is the change in the material properties caused by the radiation interactions with the material. Such damage might result from the breaking of bonds in the material or embrittlement of the material. The second possible cause might be the localized heating of the concrete caused by the absorption of the radiation energy. Radiation can also cause the dewatering of the concrete by the dissociation of the water. In many cases, it has not been possible to distinguish between the two mechanisms when concrete degradation has been identified.

Early historical data show that the compressive strength of concrete relative to a control sample decreases with radiation, see Figure 2 (see Reference 6). For conditions of radiation flux up to 2×10^{19} nvt (thermal) and temperatures to 120°C, radiation damage to some concrete is apparently insignificant, while other types show considerable loss of strength. A similar effect is seen in tensile strength, see Figure 3 (see Reference 6). A similar effect is seen for pure gamma exposure up to 1,011 rad as shown in Figure 4 (see Reference 6). All effects on concrete due to radiation, per se, were too slight to reliably measure because of the gross effects from the increased temperature during exposure (see Reference 1).

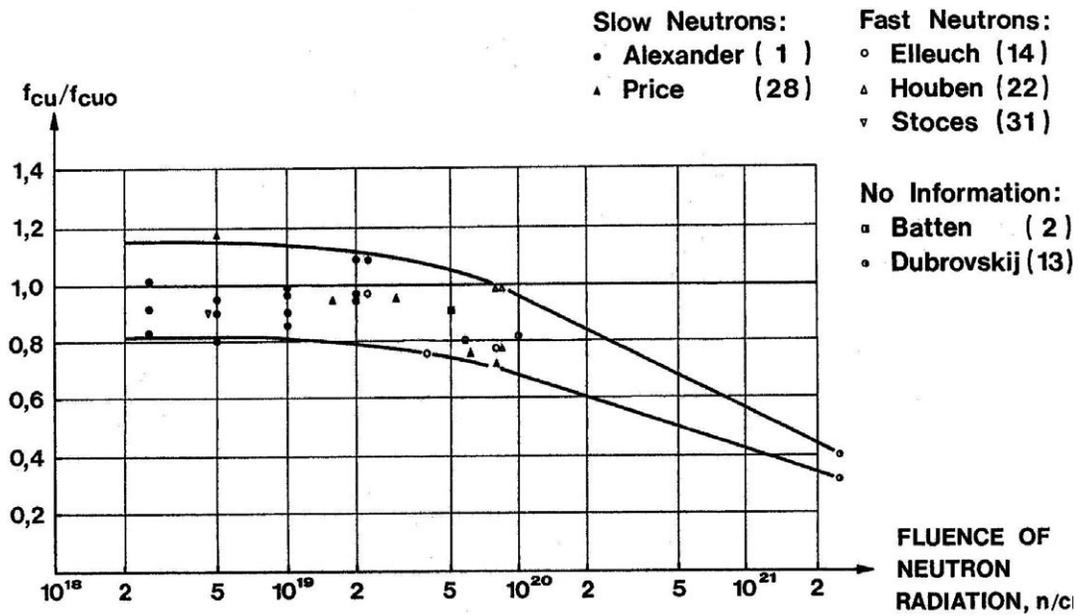


Figure 2. Compressive strength of concrete exposed to neutron radiation f_{cu} related to strength of untreated concrete f_{cuo} (see Reference 6).

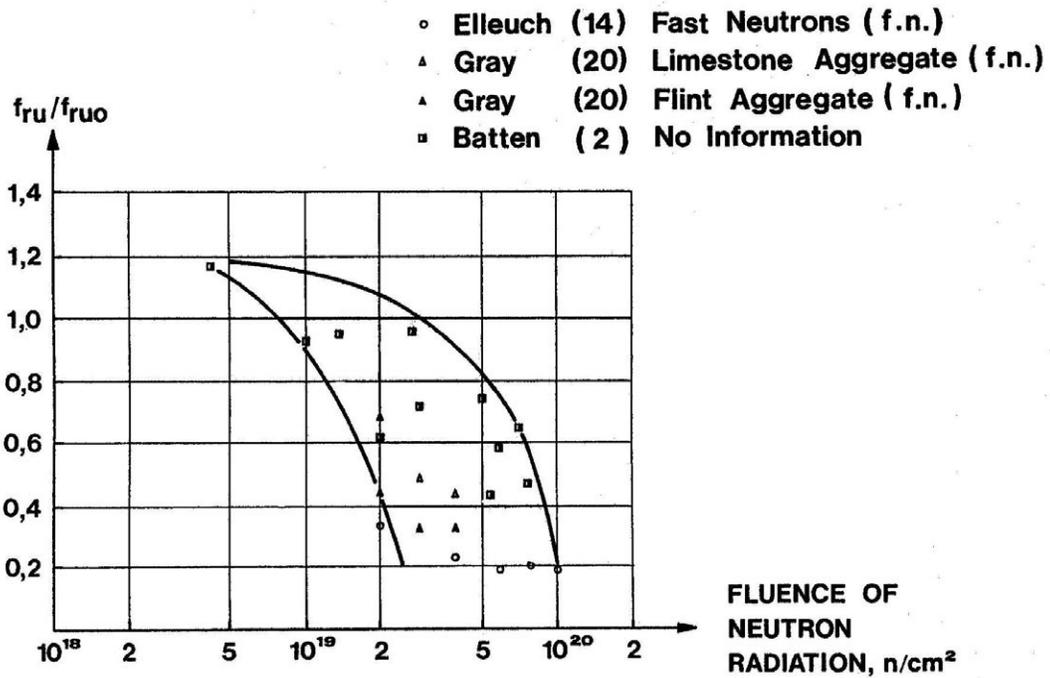


Figure 3. Tensile strength of concrete exposed to neutron radiation f_{ru} related to strength of untreated concrete f_{ruo} (see Reference 6).

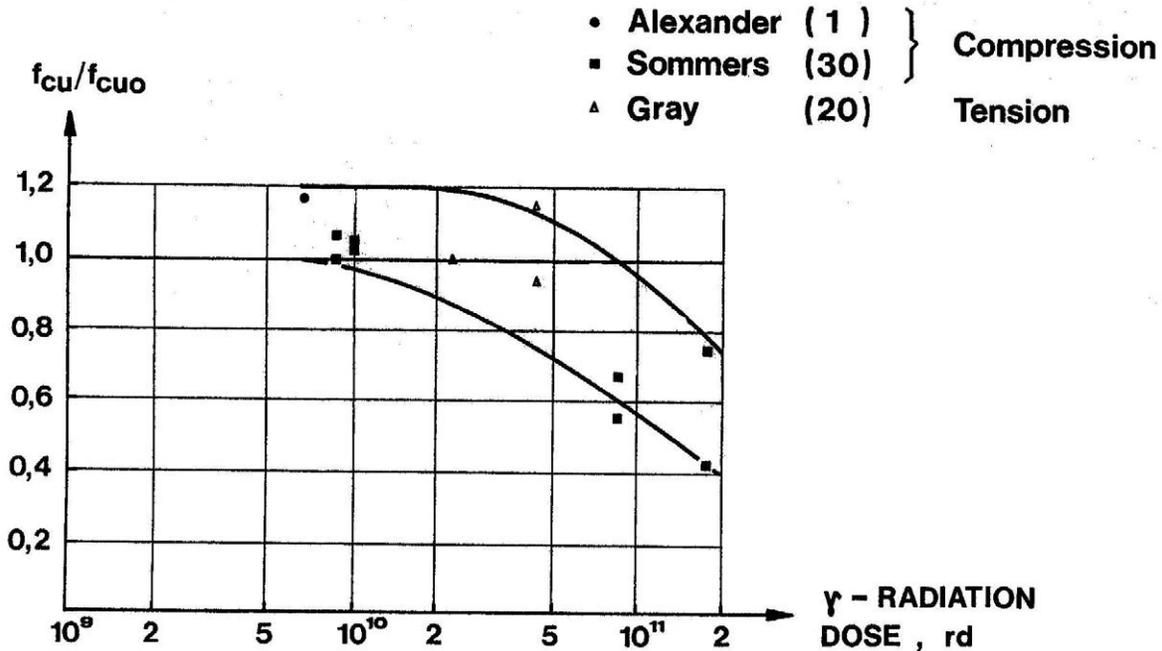


Figure 4. Compressive and tensile strength of concrete exposed to gamma radiation f_{cu} related to strength of untreated concrete f_{cu0} (see Reference 6).

Section III, Division 2 of the ASME *Boiler and Pressure Vessel Code* gives a radiation exposure level allowable to 1×10^{20} neutrons/cm². These criteria are based on a limited number of data. Quantifying the extent to which irradiation will change the properties of concrete is impossible. This is because the quantification is dependent on many factors, such as variation of material properties, material state of testing, neutron energy spectrum, and neutron dose rate (see Reference 3). There is a great deal of variability in the reported data of the effects of radiation on concrete, and the results can be misleading (see Reference 3). Based on the existing reports some conclusions might be drawn (see References 3 and 6):

1. For some concretes, neutron radiation of $>10^9$ neutron/cm² may cause some reduction in compressive and tensile strength. The amount of damage depends on the properties of the concrete mixture.
1. The decrease of tensile strength due to neutron radiation is more pronounced than the decrease of compressive strength.
2. Resistance of concrete to neutron radiation apparently depends on the energy spectrum.
3. Resistance of concrete to neutron radiation depends on mix proportions, type of cement, and type of aggregate.
4. The effect of gamma radiation on concrete's mechanical properties requires clarification.
5. The deterioration of concrete properties associated with temperature rise resulting from irradiation is relatively minor.

6. Coefficients of thermal expansion and conductivity of irradiated concrete differ little from those that would result from temperature-exposed concrete.
7. The modulus of elasticity of concrete when exposed to neutron irradiation decreases with increasing neutron fluence.
8. Creep of concrete is not affected by low-level radiation; but for high levels of exposure it is likely to increase.
9. For some concretes, neutron radiation with a fluence of $>10^{19}$ neutron/cm² can cause marked increase in volume.
10. In general, concrete's irradiation resistance increases as the irradiation resistance of aggregate increases.
11. Irradiation has little effect on shielding properties beyond the effect of moisture loss due to temperature increases.

There have been specific reports that both support and refute radiation damage to concrete. Examination of the concrete of the Materials Test Reactor fuel storage canal at the INEEL Test Reactor Area found that for gamma irradiation of 2^{11} R/hr in freely circulating water, there was a 50% loss of strength with significant surface damage.¹² On the other hand, analysis of the shielding of the graphite reactor shield at Oak Ridge National Laboratory showed that the chemical properties and density of the shield had not changed appreciably between investigations done 8 years apart. However, the compressive strength at the reflector shield interface had dropped as much as 40%. It was felt that damage from irradiation was less than that caused by relative temperature effects.¹³

According to the American National Standard Institute (ANSI) ANSI/ANS-6.4-1985, nuclear heating can be neglected if the incident energy fluxes are less than 10^{10} MeV/cm²-sec. Compressive strength and modulus of elasticity are degraded if the concrete is exposed to greater than 10^{19} n/cm² or to an integrated dose of gamma radiation exceeding 10^{10} rads.¹⁴

Concrete surrounding the pressure vessel of a nuclear reactor as a shielding material receives the highest radiation dose among concrete structures in a nuclear power plant. Assuming the lifetime of a commercial nuclear power plant to be 60 years, the integrated absorbed dose of the concrete is about 10^9 Gy for gamma rays, which is lower than the critical dose. However, the integrated dose of fast neutrons for the concrete of a conventional pressurized water reactor is close to the critical one. Because the kinetic energy of fast neutrons is quickly dissipated while traveling in concrete, the thickness of concrete that is possible to exceed the critical dose is less than 20 cm. The mechanical properties of the entire concrete structure may not be damaged by the radiation-induced degradation of such a thin layer. However, concrete pieces may be expected to fall from the surface.¹⁵

Because of the longevity requirements and importance of the concrete structures being considered, condition assessment and performance monitoring are considered to be of prime importance. Some guidance for general civil engineering structures is available through organizations such as the American Concrete Institute, the American Society of Civil Engineers, the Institute of Civil Engineers, and the International Union of Laboratories and Experts in Construction, Materials, Systems, and Structures. Unfortunately, little information is currently available that addresses condition assessment of concrete structures contained as part of a radioactive waste management facility.¹⁶

2.3 Chemical Processes

2.3.1 Chloride

Chlorides have been known to attack concrete for many years. The resistance of concrete to the penetration of chloride is one of the simplest measures to determine the durability of concrete.¹⁷ In marine environments, the reinforcing steel corrodes mainly because of the attack by chloride ions.¹⁸ Several reports have reported concrete mixtures that have resistance to chloride penetration.

2.3.2 Carbonation

Carbonation leads to a uniform corrosion of reinforcing steel that would accelerate crack formation and decrease structure life (see Reference 18). This is due to the reduction of the pH, destroying the steel reinforcing members passive layer and accelerating steel corrosion that leads to concrete cracking (see Reference 18). Carbonation occurs generally faster in dry conditions because of water blocking the pores in the concrete in wet conditions.^{19,20} Carbonation of concrete has been shown to decrease water absorption, reduce chloride ion diffusion into concrete and slow down sulphate attack.²¹ The sorptivity of concrete decreases with the age of the concrete. This is believed to be caused by carbonation of the concrete (see Reference 21). Some numerical models have been developed to describe the process.²²⁻²⁴ High-performance concrete has been proposed.^{25,26}

2.3.3 Calcium Leaching

Calcium leaching of cementitious materials has been identified as a severe long-term chemical degradation scenario of concrete structures.²⁷ Calcium leaching kinetics in water is very slow.²⁸ Aggressive environmental conditions such as ammonium nitrate can accelerate the process. Calcium leached in the pore fluid from the structural material is an equilibrium-based process (see Reference 28) and is diffusion limited (see Reference 2). The leaching is irreversible and reduces the stiffness and the limit load of structures, decreases brittleness, and changes the structural behavior (see Reference 2). It leads to a significant increase of the porosity of the material (see Reference 27).

2.4 Inspection Methods

Nuclear plant owners and operators are required to monitor the performance or conditions of structures, systems, and components (SSCs) against owner-established goals and to take appropriate corrective actions when the condition of a SSC does not conform to established goals.²⁹ In order to verify the implementation of 10 CFR.65, the Nuclear Regulatory Commission issued inspection procedure 62002. Subsequently, on May 8, 1995, the Nuclear Regulatory Commission published a final rule amending 10 CFR Part 54 that contained the requirements an applicant must meet to renew an operating license. The Nuclear Regulatory Commission concluded that passive, long-lived components should be subject to an aging management review. Inspection and timely remedial actions will ensure continued safety of the structures (see Reference 29).

2.5 Age Projection

Several models have been developed in the last 5 years to address the mechanisms of the concrete deterioration and projection of lifetimes for the structures.^{2,30} The interactions between the structural materials and the environment must be considered and modeled (see Reference 23). The modeling of carbonation in concrete is a complex problem which requires, for realistic simulation, a systematic and

robust finite element approach (see Reference 24). The presence of cracks has an important effect. One proposed model, CONDUR, is capable of handling geometry including cracks(see Reference 24).

The Structural Aging Program was initiated in 1988 and had the overall objectives of providing background data and information for identification and evaluation of the potential structural degradation processes as related to nuclear power plants.³¹ The NRC has studied the effects of aging on nuclear power plant and published Nuclear Power Plant Generic Aging Lessons Learned (GALL) Report³² where they document all of the known failures.

3. CONCLUSIONS

Concrete has been used in the construction of nuclear facilities because of two primary properties, its structural strength and its ability to shield radiation. This use has made its long-term performance crucial for the safe operation of the facilities. Understanding the conditions that lead to concrete deterioration is therefore important. An open literature, and DOE document search found that research in this area has occurred since the late 1940s and has accelerated in the last 5 years.

Radiation can affect concrete in two mechanisms. One is the change in the material properties caused by the radiation interactions with the material. Such damage might be the breaking of bonds in the material or embrittlement of the material. The second possible cause might be the localized heating of the concrete caused by the absorption of the radiation energy. In many cases, it has not been possible to distinguish between the two mechanisms when concrete degradation has been identified.

The effects of low doses, $<10^{10}$ neutron/cm² or 10^{10} Gy gamma, of radiation over periods of less than 50 years do not seem to have a significant effect on the concrete. This is supported by existing research and standards published by ASME and ANSI. Longer-term exposure, over 100 years, has not been studied.

The effects of higher doses of radiation are not as clear, and some differences in reported results exist. For some concretes, neutron exposures $>10^{10}$ neutron/cm² may cause reduction in compressive and tensile strength and may depend on the neutron energy spectrum and the composition of the concrete.

For high radiation exposure, 10^{20} neutron/cm² or 10^{10} rads of gamma, concrete has been reported to exhibit reduction in compressive and tensile strength and a marked increase in volume, due to expansion of the aggregate and the shrinkage of cement paste. It was not possible to determine if these effects were caused by the heating and loss of water or by direct radiation effects. Either way, there are deleterious effects reported.

The effects of long-term exposure of concrete to elevated temperatures are a loss of water in the concrete leading to a decrease in compressive strength, changes in the modulus of elasticity, creep resistance, conductivity, and diffusivity. Generally speaking, the threshold of degradation is 95°C, and the effects increase with increasing temperature and time exposure. Rapid temperature increase or thermal cycling can lead to significant concrete degradation.

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