

IAEA-TECDOC-693

Use of fast reactors for actinide transmutation

*Proceedings of a Specialists Meeting
held in Obninsk, Russian Federation, 22–24 September 1992*



INTERNATIONAL ATOMIC ENERGY AGENCY

IAEA

March 1993

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USE OF FAST REACTORS FOR ACTINIDE TRANSMUTATION
IAEA, VIENNA, 1993
IAEA-TECDOC-693
ISSN 1011-4289

Printed by the IAEA in Austria
March 1993

FOREWORD

An important feature of the activities of the International Atomic Energy Agency is to assist Member States in the safe handling and management of nuclear power and spent nuclear fuel and radioactive wastes that arise from the use of nuclear energy.

The management of radioactive waste is one of the key issues in today's political and public discussions on nuclear energy, especially the long term disposal of high level radioactive wastes. Rather than waiting for their radioactive decay, it is principally possible to reduce the period of toxicity of the actinides and long lived fission products through transmutation of these isotopes in fission reactors or accelerators.

The recycling of plutonium in liquid metal fast breeder reactors (LMFBRs) would allow 'burning' of the associated extremely long life transuranic waste, particularly actinides, thus reducing the required isolation time for high level waste from tens of thousands of years to hundreds of years for fission products only. This additional important mission for the LMFBR is gaining worldwide interest.

In the past years, an increasing number of studies have been carried out on the advanced waste management strategy (i.e. actinide separation and elimination) in various countries and at an international level. Recognizing this, the International Working Group on Fast Reactors (IWGFR) decided in 1991 to include the topic of actinide transmutation in liquid metal fast breeder reactors in its programme of specialists meetings. It was apparent that due to the expansion of supporting research programmes and the fragmented nature of information arising from these various activities it would be beneficial to co-ordinate this topic at an international level.

Support for this approach was provided by Member States and, against this background, the IAEA organized the Specialists Meeting on Use of Fast Breeder Reactors for Actinide Transmutation in Obninsk, Russian Federation, from 22 to 24 September 1992.

The Specialists Meeting made clear the increasing interest in the issue concerning long lived radioactive nuclides and toxicity aspects in nuclear waste management and the desirability to work out a co-ordinated approach to its solution.

The potential of fast reactors for reducing the long term radiotoxicity of spent fuel justifies an active programme of conceiving and developing technologies for recovering transuranics for in-core transmutation.

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SUMMARY OF THE SPECIALISTS MEETING

The Specialists Meeting on Use of Fast Breeder Reactors for Actinide Transmutation was held in Obninsk, Russian Federation, from 22 to 24 September 1992.

The meeting was sponsored by the International Atomic Energy Agency on the recommendation of the International Working Group on Fast Reactors and was hosted by the Institute of Physics and Power Engineering (IPPE) in Obninsk. The meeting was attended by 23 participants from eight countries (Belgium, France, Germany, Japan, Russia, Switzerland, United Kingdom and USA), two International organizations (IAEA and CEC) and 12 observers from Russia.

The meeting was opened by Mr. L.A. Kochetkov, Deputy Director of IPPE, who welcomed the participants on behalf of the Ministry of the Russian Federation on Atomic Energy and the IPPE. Referring to the subject of this Specialists Meeting, he emphasized the importance of the advanced waste management strategy (i.e. actinide separation and elimination). Mr. V. Arkhipov of the IAEA presented a review of IAEA activities in the field of minor actinide transmutation. The meeting was chaired by Mr. L.A. Kochetkov.

Mr. L. Koch presented a background paper on the status of transmutation, prepared at the request of the IAEA. The paper discusses the objectives and the proposed schemes for partitioning and transmutation of long living radiotoxic nuclides. In particular, the constraints for the efficiency of a P&T process guiding R&D work for a fast reactor fuel cycle are described.

Session 1: National programmes on transmutation of minor actinides and long lived fission products in FBRs
Chairmen: L.A. Kochetkov, U. Wehmann

This Session gave an overview of the national programmes followed in France, the USA, Japan and Switzerland. The programmes reflect the large effort which is being undertaken worldwide with the goal to reduce the quantities of the toxic wastes and the long term risk of geological repositories. In the technical presentations, the potential of fast reactors of standard MOX fuel design as well as of special minor actinide burner cores was illustrated.

Session 2: Physics aspects of transmutation of minor actinides and long lived fission products in FBRs
Chairmen: L.A. Kochetkov, L. Koch

The papers presented in this Session cover a variety of topics related to transmutation of actinides and fission products, ranging from heavy water blankets for a LANL type accelerator-driven reactor to the adaptation of fast reactors to transmute plutonium and radiotoxic waste from LWRs together with self-generated waste. Here the European EFR as well as the GE designed ALMR are considered and the possibilities of the Russian BN-800 are discussed.

The advantage of high thermal fluxes in fissioning Am-241 and Np-237 by their neutron capture products, Am-242 and Np-238, is pointed out. These nuclides with odd neutron and proton numbers possess very high fission cross-sections. In order to reduce the competing decay reaction,

high thermal neutron fluxes in the area of 10^{16} n/s cm^2 are needed, which cause secondary problems in the reactor itself, e.g. the buildup of tritium. One possibility to circumvent these problems is to use a stationary liquid blanket.

Session 3: Engineering aspects of transmutation using fast reactors
Chairmen: L.A. Kotchetkov, H. Sztark

Topics covered in this Session relate to transmutation of minor actinides and fission products, mainly with reactors devoted to this purpose, with specific core concepts and adapted fuels and fuel cycles.

Theoretical studies, mainly on the Russian side, review the physical parameters which govern the burning capabilities of the core while improving the safety parameters, with special concern for the voiding coefficient. The need to increase the number of free neutrons available for transmutation of minor actinides and eventually fission products is emphasized, leading to very new arrangements for core concepts, fuel and also coolant. These new ideas are in a very preliminary stage, the feasibility of such systems has to be confirmed.

In the Japanese OMEGA programme, the part concerning fast reactors was also developed, with specific studies of reactors devoted to burn actinides; M-ABR with harder neutron spectrum, P-ABR with higher neutron flux, SLLC (Super Long Life Core) with no need for refuelling. In this case also, only the neutronic feasibility has been assessed, further studies being necessary to confirm these concepts.

When comparing the "figure of merit" of all the proposed solutions, it appears again that there is a lack of a common model to normalize the hazard reduction, particularly for what concerns the reprocessing process and the related reprocessing losses. This is apparent in this Session, but also when comparing this Session to previous ones, for example for the same EFR core model which has been treated in all three sessions. The potential of fast reactors for hazard reduction seems obvious to everybody, but the way to express this potential is not the same: transmuted mass per fast reactor unit, ratio between the number of fast reactor and the number of light water reactors, long term radiotoxicity, etc. Also, the need to recycle the fission products was discussed, some of the participants being in favour of controlled deep disposal.

The separation techniques for the different wastes, especially the separation efficiency and the related losses in the residual wastes, were also discussed, while this topic was outside the scope of this meeting (separation of rare earth and curium). Nevertheless, it appears that if further progress in hazard reduction has to be achieved, improvements in separation techniques must be obtained since the amount of losses during fuel operations (reprocessing, fabrication) becomes the major part of these hazards.

The experimental background and future programme was explained by Russian papers, as well for the nuclear data as for some fuel process. Experiments performed in Japan (FCA) and in Russia (BFS) showed quite similar results, with some discrepancies in actual calculations. Further experiments are planned in BFS involving some 50 to 100 kg of Np-237. Concerning fuel, experience has been accumulated on BOR60 with oxide fuel obtained by vibropacking, irradiated up to 7.7% burnup for uranium oxide and 4.5% for mixed oxide. This type of fuel would allow simpler fabrication and reprocessing technology, and could be demonstrated on BOR60 provided fundings are available.

In summing up this session, one can observe that advanced fast reactor concepts can improve significantly the transmutation capability of classical fast reactors, provided that similar improvements are obtained in reprocessing efficiency.

Conclusions

The IAEA Specialists Meeting made clear the increasing interest in the issue concerning long lived radioactive nuclides and toxicity aspects in nuclear waste management and the desirability to work out a co-ordinated approach to its solution.

The potential of fast reactors for reducing the long term radiotoxicity of spent fuel justifies an active programme for conceiving and developing technologies for recovering transuranics for in-core transmutation.

The specialists agree that future progress in solving transmutation problems could be achieved by improvements in:

- Radiochemical partitioning and extraction of the actinides from the spent fuel (at least 98% for Np and Cm and 99.9% for Pu and Am isotopes);
- Technological research and development on the design, fabrication and irradiation of the minor actinides (MAs) containing fuels;
- Nuclear constants measurement and evaluation (selective cross-sections, fission fragments yields, delayed neutron parameters) especially for MA burners;
- Demonstration of the feasibility of the safe and economic MA burner cores;
- Knowledge of the impact of maximum tolerable amount of rare earths in americium containing fuels.

BACKGROUND PAPER

STATUS OF TRANSMUTATION

L. KOCH

Institute for Transuranium Elements,
Commission of the European Communities,
Joint Research Centre,
Karlsruhe

Abstract

The objectives and the proposed schemes for partitioning and transmutation of long-living radiotoxic nuclides are discussed. In particular the constraints for the efficiency of a P & T process guiding R & D work for a fast reactor fuel cycle are described.

1. INTRODUCTION

During nuclear energy generation highly radiotoxic by-products such as the minor actinides are formed, which accumulate in spent fuels or in reprocessing waste streams on the ton scale (Table I).

Because of the present low prices for uranium there are no or little incentives to recycle plutonium in LWR. Therefore several states (USA, Spain, Sweden, and others) have opted for direct disposal of the spent fuel, whereas some states still hang on with the reprocessing of spent fuels in order to recover plutonium (F, GB, J, CIS). Other states consider both options e.g. FRG. Initially the recovered plutonium was foreseen to fuel fast reactors, but because of the same reason (low uranium price) the deployment of this reactor type will be delayed at least for another 20 to 40 years. Hence - as an alternative - the recycling of Pu in LWR is considered, where the reprocessing technology is established instead of its use in fast reactors. This, however, will further increase the formation of minor actinides (Table I) [1].

The toxicity potential of the spent fuel is known to diminish with time (Fig. 1). The most toxic nuclides are ^{241}Am and its daughter nuclide ^{237}Np (which is also independently formed during nuclear energy generation by other processes). If the transuranium elements are partitioned from the spent fuel and transmuted in a fast reactor to 0.5% of their initial value then the radiotoxic hazard is reduced by a factor of 200 compared to the initial values in general [2]. ^{238}Pu is the only nuclide where this factor is not achieved due to build-up during the transmutation processes. Although the radiotoxicity of fission products decays rapidly during the first 600 years the hazard posed by the remaining nuclides is considerable, because some of these nuclides are readily soluble in water and could migrate back to the biosphere.

During the last 20 years the feasibility of partitioning and transmutation schemes have been studied. The results were presented at several topical meetings over the last few years [3 - 13].

2. OBJECTIVES

In order to develop 'Partitioning & Transmutation' schemes for radiotoxic nuclides one should find answers to the following questions in order to define clearly the scope of the R & D work needed:

- Which nuclides and to what extent should be transmuted?
- Which of the proposed P & T schemes is the most promising one?
- How efficient can the P & T process be?

2.1. Nuclides to be transmuted

During the meetings mentioned above several candidate radionuclides for transmutations were identified, but not all of the listed radionuclides (Table II) pose the same radiological hazard to the public when they are eventually released from a geological repository. If one assumes that such a repository will remain intact for several thousand years (as it is the experience for human made structures on earth) then nuclides with short half-lives like ^{137}Cs , ^{90}Sr , ^{137}Cs , ^{121}Sb , ^{126}Sn would decay. From the remaining only those which form monovalent ions could be transported back by water to the biosphere: ^{14}C , ^{36}Cl , ^{99}Tc , ^{129}I , ^{135}Cs and ^{237}Np . Since the latter nuclide has three parent nuclides: ^{241}Pu , ^{241}Am , ^{245}Cm all actinides have to be considered as potentially hazardous. During the migration through the geosphere the nuclides will be diluted with natural stable isotopes, which could reduce the specific activity to insignificant levels depending on the geological formation. To support this argument the annual average uptake of the natural element by the "CORP-man" [14] is compared to the annual limits of uptake for each radionuclide [15]. From this follows the tolerable isotopic abundance for each of the considered radionuclides in the uptaken natural element (Table II). To what extent such an isotope dilution will occur during the release from the repository is presently being investigated [3]. This concerns all nuclides except ^{99}Tc and the actinides. Therefore, as a first priority, the artificial elements should be transmuted: ^{99}Tc , ^{237}Np and its parent nuclides ^{241}Pu , ^{241}Am , ^{245}Cm .

2.2. Proposed partitioning and transmutation schemes

There are several nuclear processes under investigation which could transmute a radionuclide into a less toxic one (Fig. 2). Photons in the range of 10 MeV produced as Bremsstrahlung from accelerated electrons induce transmutation (by (γ, n)) and fission in actinides and can be employed for transmutation purposes [16 - 18]. The direct spallation of radionuclides by high energetic charged particles has been proposed [19] as well as the use of an inactive

TABLE I. FORMATION OF CERTAIN LONG-LIVING
RADIOTOXIC NUCLIDES IN ^{235}U AND MOX
FUELED LWR (AMOUNT PER GW_{e,a})

Initial Fuel	^{235}U (W ₀)	3.2	0.7
$^{239} + ^{241}\text{Pu}$ (W ₀)	—	—	3.2
Burn-up (GW _{th} d/t)	33	33	33
spent fuel			
^{237}Np (kg)	14.1	4.2	4.2
^{241}Am (kg)	2.2	22.9	22.9
^{243}Am (kg)	2.8	54.1	54.1
^{242}Cm (kg)	0.2	2.0	2.0
^{244}Cm (kg)	0.8	28.5	28.5
^{99}Tc (kg)	26.0	26.0	26.0

TABLE II. CANDIDATE RADIONUCLIDES FOR TRANSMUTATION

Yearly uptake of elements [g]	Nuclide	T _{1/2} [a]	ALI [Bq]	Isotopic abundance [%]
1.1 E5	114C1	5.7 E31	1.5 E61	1 E-81
1.9 E31	36Cl1	3.0 E51	1.3 E41	5 E-71
7.3 E-21	129I1	1.6 E71	3.3 E21	7 E-21
3.6 E-31	135Cs1	20 E61	5.1 E41	3 E11
	79Se2	6.5 E42		
	93Zr2	1.5 E62		
	90Sr3	28.53		
	121Sn3	503		
	126Sn3	10.53		
	137Cs3	30.173		
	99Te4	21 E54		
	237Np4	2.1 E64		
	238Pu4	87.74		
	239Pu4	2.4 E44		
	240Pu4	6.5 E34		
	241Pu4	14.44		
	242Pu4	3.7 E54		
	241Am4	4324		
	242Am4	1414		
	243Am4	7.3 E34		
	245Cm4	8.5 E34		
	246Cm4	4.7 E34		
	247Cm4	1.5 E74		
	248Cm4	3.4 E54		

1 Nuclides forming monovalent ions with natural isotopic diluent.

2 Nuclides forming no monovalent ions.

3 Nuclides with half-lives below 100a decaying into stable nuclides.

4 Nuclides and parents forming monovalent ions with no natural diluent.

spallation source (e.g. Pb-Bi) to produce intense neutron fluxes [20 - 24]. The latter concepts are not very different from nuclear reactors - discussed below -- except that they can use subcritical arrangements with thermal [20, 22] or fast neutrons [21, 24] driven by accelerators. Included in the installations are reprocessing facilities which separate the remaining nuclides from the transmutation products (if needed). The radionuclides are introduced either continuously as aqueous slurries or

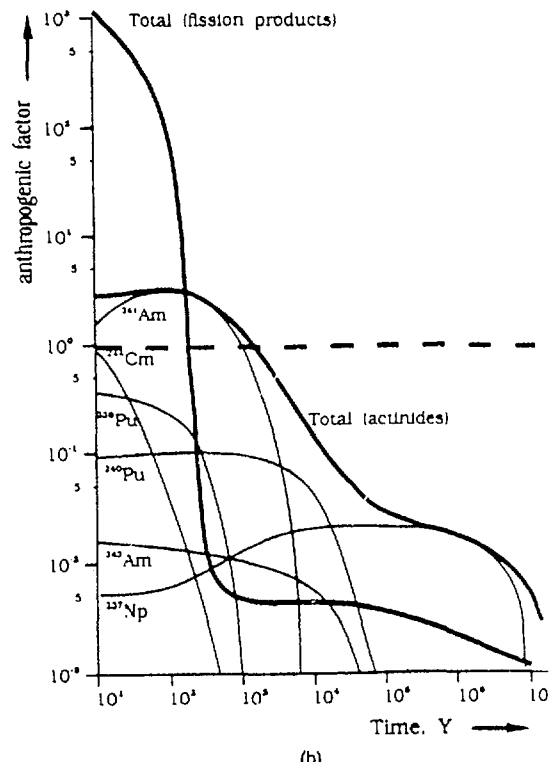
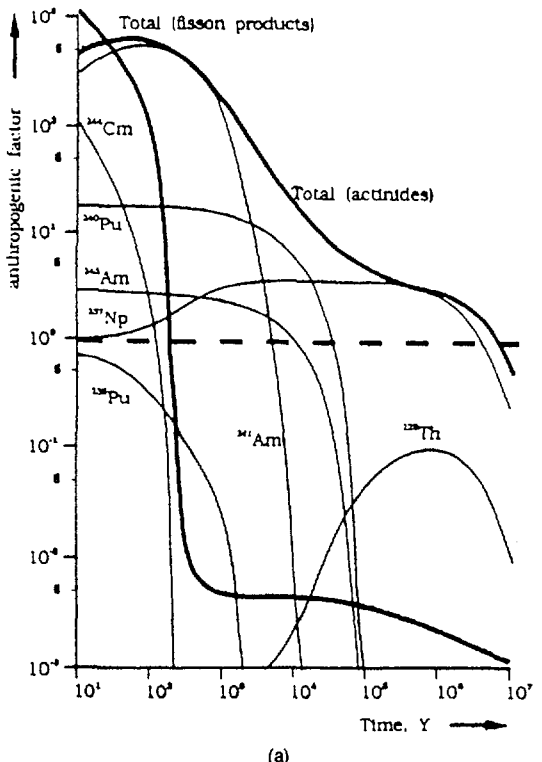


Fig. 1. Time-dependent radiotoxicity of a PWR fuel without reprocessing (a) and with MA recycling in a FR (b). (Here the radiotoxicity of a nuclide mass in terms of fatal cancer doses is normalised to the radiotoxicity of uranium ore to produce a PWR fuel [2].)

TABLE III. MINOR ACTINIDE - CONTAINING FUELS IRRADIATED IN FAST REACTORS

$(^{241}\text{Am})_{0.5}(\text{U})_{0.5}\text{O}_{1.95}$	PFR
$^{241}\text{Am}_2\text{O}_3$	PFR
$^{244}\text{Cm}_2\text{O}_3$	PFR
$(^{241}\text{Am})_{2.44}(\text{Cm})_{0.73}\text{O}_3$	PFR
NpO_2	KNKII
$(^{241}\text{Am})_{0.5}(\text{U})_{0.5}\text{O}_{1.92}$	KNK II
$(\text{Np})_{0.45}(\text{U})_{0.55}\text{O}_{2.00}$	PHENIX
$(\text{Np})_{0.2}(\text{Am})_{0.2}(\text{U})_{0.6}\text{O}_{1.95}$	PHENIX
$(\text{U})_{0.73}(\text{Pu})_{0.25}(\text{Np})_{0.02}\text{O}_{1.98}$	PHENIX
$(\text{U})_{0.73}(\text{Pu})_{0.25}(\text{Am})_{0.02}\text{O}_{1.98}$	PHENIX

solutions [20] or as molten salts [24] to the reactor or are added to fuel assemblies consisting of alloys [21] similar to the MA burner concept being studied by JAERI [25].

Other concepts propose using existing LWR [26] and FR [2, 27, 28] or HWR [22] for transmutation. Not only Pu produced in the LWR fuel cycle but also the minor actinides could fuel fast reactors. Several years ago MA-containing mixed oxide fuels were developed and tested [2] (Table III). The MA partitioned from the spent LWR amount to about 10% of the Pu in that fuel. They can be either homogeneously mixed to the FR MOX fuel or concentrated in a few assemblies of the reactor. Their concentration would be about 2% of the fuel, well below the limit that would influence the Na void coefficient [29]. For higher MA concentrations the geometry of the core needs to be altered [30]. The partitioning of minor actinides (and selected fission products) from the spent fuel is not yet established. R & D work is under way to improve the Am recovery by electro-refining of Zr-based alloys [31] and the coextraction of Am, Cm is achieved by the TRUEX process [32]. Due to the similar chemical behaviour of lanthanides and Am, Cm especially in the latter case the products contain too much of the lanthanides to be directly used as fuels. Studies are under way to develop new processes to separate the two groups of elements [31, 33].

Since this meeting is dedicated to the use of fast reactors for transmutation purposes I will briefly compare this concept with the above mentioned ones. The present strategies to cope with the rising anthropogenic waste streams of all kinds are to develop processes which

- generate less waste,
- recycle (and reuse) waste,
- incinerate waste in special installations,
- condition waste for geological disposal [34, 35].

The options are listed according to public acceptance giving the first the highest and the last the lowest ranking. If these criteria are applied to the proposed concepts of P & T one should add four more: nuclear material safeguards, transportation of nuclear materials between different sites, technical feasibility and last but not least, impact on nuclear energy generating cost. The advantage of the

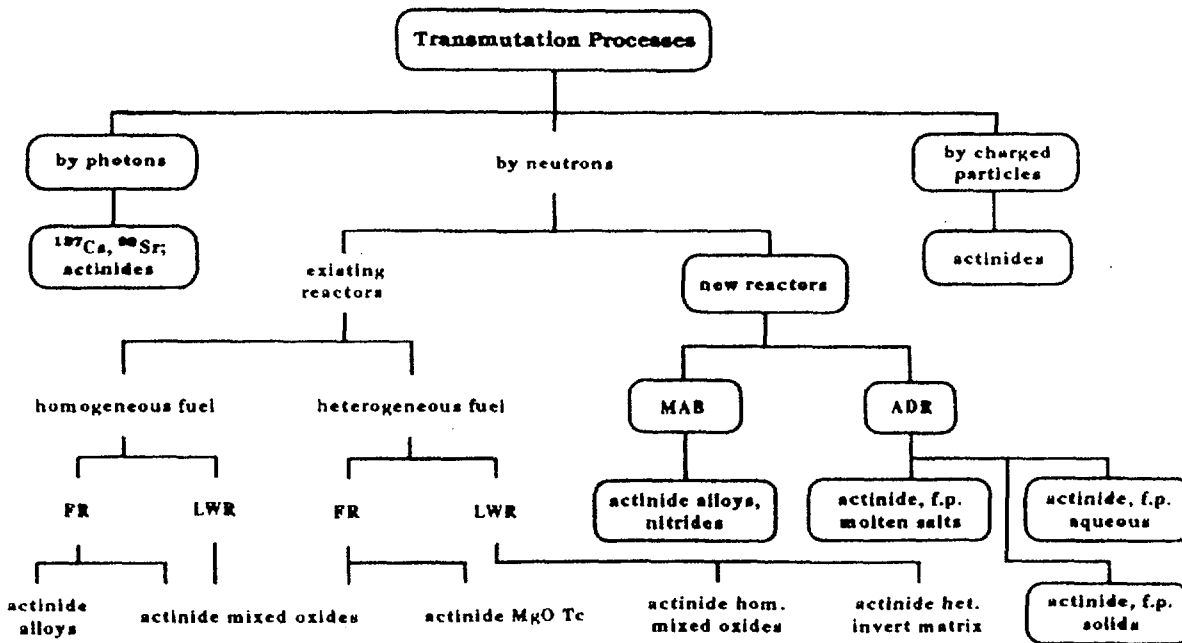


Fig. 2. Scheme of proposed transmutation processes.

accelerator-driven reactor concepts as well as the MA burner is that they can be used in symbiosis with any nuclear fuel cycle or transmuted the remaining nuclear waste after a "stop of nuclear energy generation" (Ausstieg scenario) or destroy nuclear weapons material quickly. The technical feasibility of such systems, however, is not yet developed to a stage comparable to the existing nuclear fuel cycle, by which one also can achieve the same goals (with lower efficiency in respect to transmutation rates but certainly less cost). Moreover in the public opinion the recycle of waste is preferred to its incineration in special installations. Hence, if the present nuclear reactors can cope with the posed transmutation problems, they would be preferred to any other concept.

MA's together with Pu can be recycled in a self-generated mode in existing LWR and FR [2]. The differences between the two reactor types are: The LWR in an equilibrium fuel cycle contains more than 10 times transplutonium nuclides compared to a fast reactor fuel in equilibrium [36-39]. The neutron flux of present LWR is too low to transmute ^{99}Tc and eventually ^{129}I efficiently. This is not the case for the FR where in the blanket region a thermalised neutron flux of between 0.5 - 1 · 10¹⁵n/sec cm² can be produced [40]. Inherent with the transmutation of MA is the build-up of ^{238}Pu which further denatures Pu in the sense of nuclear material safeguards especially in case of FR with its low ^{240}Pu content. However it would be a nuisance to the fabricator of mixed oxide fuels - not of metallic fuel for the IFR where already remote fuel fabrication is foreseen. The latter concept eliminates the need of fuel transportation, which in case of oxide FR - and even more the LWR - fuels will be difficult, mainly due to the higher neutron radiation of ^{244}Cm in fresh and of ^{242}Cm , ^{252}Cf in spent fuels.

MA actinides are a fuel for fast reactors, so that the additional energy generation and breeding rate would compensate for the extra costs in fuel make-up [2]. Since the energy generating costs of a LWR are considerably lower than that of a FR this argument is presently not valid. The development of a P & T scheme (according to the present R & D programmes) will take more than 15 years, until then the uranium prices may well favour the FR over the LWR.

The FR with on-site reprocessing seems to be a viable concept for transmutation and presently the only existing reactor that can reduce its waste efficiently by recycling. It should not be forgotten, however, that an alternative fuel cycle based on the thorium molten salt reactor [35] could prove to be even more advantageous, since the MA production there is considerably lower and MA together with fission products can be transmuted as well. Today the technical feasibility (despite extensive efforts at ORNL in the past) is not yet proven.

2.3. Efficiency of P & T schemes

If one has agreed on the kind of nuclides to be transmuted, the question arises: to what extent should a P & T scheme reduce the radiotoxicity of a particular nuclide? Earlier answers to this question proposed "hazard reduction factors" of several orders of magnitude [41] which aimed at elimination of nuclides down to the "maximal permissible concentrations". Any realistic approach, however, has to consider losses during the P & T processes, which have still to be buried in a geological repository. How large these losses are, depends on the available technology, therefore one should retreat to the ALARA principle and reduce the radiotoxicity of a certain nuclide as low as reasonably achievable. For the existing fuel cycle - when extended to P & T - one could attain a reduction factor of 100 under the following constraints [34]:

Since one expects a reasonable reduction of the radiotoxic nuclides during the life-time of a reactor one can determine the lower limit of a cross-section for the neutron reaction causing the transmutation. The half-life, T_{1/2}, of this process is given by the equation

$$T_{1/2} = \frac{\ln 2}{\sigma \phi} \quad (\sigma \text{ being the absorption cross section in the neutron flux } \phi).$$

If one assumes that the time of the minor-actinide-containing fuel in the core of the nuclear reactor is limited by the mechanical stability of its cladding (which is presently about 3 years in a fast reactor) and that during this period at least half of the minor actinides or fission products have to be transmuted, then the cross section should be

$$\sigma > 1.5 \text{ b } (5 \cdot 10^{15} \text{ n/sec cm}^2 \text{ fast}) \\ \sigma > 18 \text{ b } (5 \cdot 10^{14} \text{ n/sec cm}^2 \text{ thermal}).$$

The need to recycle the minor actinides due to the limitations of present claddings leads often to a wrong perception. If one expects a reduction of the radiotoxicity to 99.6% (which is equivalent to about 8 refueling cycles and which also corresponds to the life-time of the nuclear power station) then one easily can see that in total only twice the mass of the radionuclides have to be processed.

With N being the mass which is transmuted in a reactor of a refueling cycle length, t, assuming that t is about the same as T_{1/2} (transmutation half-life of the nuclide), then the decrease of the initial mass N₀ as a function of the number of cycles (n), can be described as:

$$N(n) = N_0 \cdot 2^{-n/T} \\ = N_0 \cdot 1/2^n \quad (\text{no feedback due to } \alpha\text{-decay considered})$$

with

N(t), the mass to be reworked for each cycle.

$\Sigma N(t)$, the total mass to be recycled is only 2 times the initial mass N₀

$$\Sigma N(t) = N_0(1 + \Sigma 1/2^n) \\ = 2N_0.$$

Under such circumstances one can estimate the reduction factor of the radiotoxicity. The present reprocessing technology and subsequent fuel make-up has losses of minor actinides to the waste streams on the order of 0.3% per cycle. Hence the accumulated losses during all reprocessing stages would be 0.6% which together with the remaining 0.4% of the radionuclide after 8 cycles will sum up to 1% of the original radiotoxicity, i.e. the overall reduction of the initial radiotoxicity will be hundred times. This is still a significant reduction of an environmental hazard if compared to that of other noxes like NO_x, SO₂ (in the exhaust gases of automobiles and fossil fuel power stations) having a reduction by only a factor of 3.

3. OUTLOOK

If nuclear energy generation is to be used by future generations then the limited supply of ^{235}U will renew the interest in the fast breeder concept. Regardless of what future fast reactors will look like, they will have the inherent potential to burn most radiotoxic waste. Therefore the development of the minor-actinide-containing fuel cycle for fast reactors is an essential task in keeping the nuclear energy option open.

Apart from basic research of more effective partitioning processes the basic physico-chemical properties of MA-containing fuels have to be studied and their irradiation behaviour tested.

For the IFR concept the electro-refining process has to be extended to extract all minor actinides from the spent fuel with a sufficient decontamination from lanthanides and to isolate technetium which accumulates with other elements in the cadmium cathode. Investigations in these directions are under way, where the

development of an appropriate fuel is also included [31, 33]. This fuel is being developed in a joint effort between the European Institute for Transuranium Elements and CRIEPI and will be tested in a joint irradiation experiment with CEA in PHENIX.

For the oxide-fueled fast reactor the reprocessing covering minor actinides and technetium has not yet been developed to a comparable stage as it is for the IFR. Nevertheless promising extraction schemes are under test which would not only extract these nuclides from the LWR spent fuel but, also later could be used to reprocess the FR fuel [33, 42]. Minor-actinide-containing fuels have been tested in the PHENIX reactor (Table III) [43]. The post-irradiation examinations revealed no unexpected behaviour under irradiation. A follow-up irradiation experiment is presently under preparation which should lead to higher burn-ups. These fuels are developed jointly with the CEA, France and the European Institute for Transuranium Elements and have been or will be irradiated in PHENIX.

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NATIONAL PROGRAMMES ON
TRANSMUTATION OF MINOR ACTINIDES
AND LONG LIVED FISSION PRODUCTS IN FBRs

(Session 1)

Chairmen

L.A. KOCHETKOV
Russian Federation

U. WEHMANN
Germany

UNITED STATES NATIONAL PROGRAM ON ACTINIDE RECYCLE

(Summary)

H. KHALIL

Argonne National Laboratory,
Argonne, Illinois,
United States of America

The current U.S. National Energy Strategy includes four key goals for nuclear policy: maintain safety and design standards, reduce economic risk, reduce regulatory risk, and establish an effective high-level nuclear waste program. A potentially effective means of reducing the long-term radiological toxicity of high-level wastes destined for geologic disposal (primarily spent LWR fuel) is to extract the transuranic irradiation products and to destroy them by transmutation in reactors or accelerator concepts. Actinide recycle was considered previously in the United States and rejected because of proliferation concerns and little perceived benefit to the viability of light water reactor (LWR) deployment or to disposal. The current U.S. program, using new fuel processing and waste management technologies and a modular, passively safe advanced liquid metal reactor (ALMR) concept, offers the prospect of overcoming these concerns.

The United States national program on Actinide Recycle is directed by the U.S. Department of Energy (DOE), who is responsible for development of energy policy and R&D. The actinide recycle program is coordinated with the Integral Fast Reactor (IFR) metal fuel development program and the ALMR program, all sponsored by DOE.* The actinide recycle approach comprises technology for the recovery and conversion of the transuranic content of LWR spent fuel to a metallic fuel form by pyrometallurgical means and its subsequent consumption in an ALMR fuel cycle.

Actinide Recycle from LWR Fuel

The extension of the IFR closed fuel cycle pyroprocess technology for the extraction of transuranics from LWR spent fuel for consumption in ALMRs is being undertaken in the actinide recycle program at Argonne National Laboratory. Pyrochemical processes are being developed for the purpose of economically extracting the transuranic species from LWR spent oxide fuel and for concentrating these transuranics in a metallic form suitable for introduction as feed material into the IFR fuel cycle. The goal of the process development is to recover at least 99.9% of the LWR discharged transuranics for use in ALMRs.

* S. Rosen and C.E. Weber, "The ALMR as a Future Energy Source for the United States," *Proceedings of International Conference on Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, (1991) Paper 1-1

The processes being studied include the decladding of the spent LWR fuel pins by suitable mechanical and/or chemical means and the subsequent pyrochemical decomposition of the spent fuel to provide (a) a transuranic product containing the transuranics plus a portion of the lanthanide fission products, and designed for introduction to the ALMR cycle at the electrorefining step; (b) a uranium-rich component (composing the bulk of the LWR spent fuel mass) suitable for storage and potential future use as the source of LWR or ALMR fuel; and (c) waste streams that can be converted and packaged into forms acceptable for geologic disposal.

In the current, early phase of the actinide recycle program, three different pyrochemical separation concepts, referred to as the "salt transport," "magnesium extraction," and "zinc-magnesium" processes, are being investigated using small-scale experiments to establish chemical feasibility and to identify the most attractive process for further development. In the subsequent phase, the scale will be increased to examine the feasibility of engineering aspects of the selected process, such as materials compatibility, equipment scaleup, and process integration. The program goal is to provide sufficient information to support a technical feasibility decision in 1995. Large-scale demonstration of the process with LWR spent fuel is planned pending a favorable decision at that time.

To address fuel fabrication and in-reactor performance issues, work is also underway to demonstrate fuel fabrication by injection casting and to achieve acceptable burnup levels in fuel containing minor actinides (Np and Am) and lanthanide fission product impurities. Initial fuel casting studies have been concerned with verifying adequate control of fabricated fuel compositions. Upon establishment of adequate fuel casting parameters, fuel elements will be cast for irradiation, and their in-reactor performance (e.g., fuel-clad compatibility) will be established by periodic examinations as a function of exposure.

To help assess the ability of current nuclear data and computational procedures to predict the relevant actinide transmutation rates, actinide samples will be irradiated in EBR-II for the purpose of measuring their reaction rates and comparing these measurements with calculational estimates.

Neutronics studies are being performed to evaluate the performance and safety characteristics of core concepts in which the actinide management objective ranges from efficient breeding to net transuranic consumption. Related system evaluations will be conducted of LWR/IFR synergistic fuel cycle scenarios and of the radiological hazard implications of these scenarios, in order to optimize the management of transuranic inventories.

Advanced Liquid Metal Reactor (ALMR) Development

The reactor design is a sodium-cooled, pool type reactor based on the Power Reactor, Innovative Small Module (PRISM) concept originated by General Electric. The design is being performed by a GE-led industry team. The current reference design is a 471 MWt modular reactor (nine modules constitute a 1440 MWe plant).

ALMR passive safety features assure accommodation of anticipated transients without scram. A passive reactor vessel auxiliary cooling system assures safety-grade decay heat removal. The reference core design is a breeder core utilizing ternary metal fuel (U-Pu-Zr). Actinide recycle is incorporated into the reference core design by designing cores to study the possibilities of transmuted actinides in both breeder and burner configurations which use the current reactor design. The Nuclear Regulatory Commission (NRC) is reviewing the pre-application Preliminary Safety Information Document (PSID) and preparing a Safety Evaluation Report (SER). Key components, including electromagnetic pump stators and seismic isolators, are in the testing phase. Future work includes commercial participation in building a prototype test module. Safety tests in the prototype will be the bases for final plant design certification by the NRC.

Overview of Technical Papers

Two technical papers from the United States will be presented at this meeting. The first paper is "Physics Considerations in the Design of Liquid Metal Reactors for Transuranium Element Consumption." This paper is by H. Khalil, R. Hill, E. Fujita, and D. Wade (all ANL) and will be presented by H. Khalil. This paper addresses concepts which exploit key features of the metal fuel cycle and the hard spectrum characteristics of ALMRs. Physics aspects of actinide transmutation are discussed for a range of transuranic management options. The second paper is "Actinide Transmutation in the Advanced Liquid Metal Reactor (ALMR)" by Christine Cockey (GE). This paper addresses the possibilities of actinide transmutation in the reference metal-fueled ALMR. Actinide transmutation core designs are presented as well as lifetime actinide mass consumption values for different configurations.

LONG LIVED WASTES TRANSMUTATION STUDIES IN FRANCE

M. SALVATORES, C. PRUNIER,
P. BERGEONNEAU, A. ZAETTA
Commissariat à l'énergie atomique,
Centre d'études nucléaires de Cadarache,
Saint-Paul-lez-Durance

H. SZTARK
Framatome,
Lyon

G. VAMBENEPE, J. VERGNES
Electricité de France,
Lyon
France

Abstract

In this paper, the French activities for LMFBR, in the field of basic data validation, reactor and fuel studies are reviewed.

1. INTRODUCTION

Reactor incineration of transactinides and long-lived fission products is considered as a possible complementary strategy to the deep storage of high-level radioactive wastes.

The main objective is to envisage the reduction of the quantities of toxic wastes to be stored, the longterm safety (and then the risk reduction) of geological repositories being a separate issue.

In France, CEA, FRAMATOME and EDF make studies on this topic. The French utility EDF, since it is the main producer of HLW, is much concerned by the management of these wastes : in this respect, EDF takes a financial part in the programme developed in CEA. FRAMATOME, as reactor constructor and fuel manufacturer, participates also to this programme.

Concerning transmutation potential in LWR and accelerators, studies are also performed but will not be described in this note as it is out of the scope of this seminar.

2. INTEGRAL EXPERIMENTS FOR DATA VALIDATION

In France, a large experimental data base is available to validate the basic nuclear data. Both irradiated fuel experiments and integral experiments have been performed, which can give informations on MINAC and fission product data.

2.1. IRRADIATIONS IN FAST NEUTRON SPECTRA :

Irradiations have been performed in PHENIX of :

- special fuel pins with variable Pu vectors (TRAPU experiment),
- separated isotopes samples (PROFIL-1 and PROFIL-2 experiments),
- special fuel pins with different contents of Np and Am (SUPERFACT experiment), (Figure 1).

The PROFIL-1, PROFIL-2 and TRAPU experiments have been analyzed in terms of JEF-1 data [1]. Indications have been obtained on integral capture and fission cross-section, on some (n,2n) reactions (in particular for Np-237), and on some branching ratios in the decay schemes.

The analysis of the SUPERFACT experiment is still underway.

2.2. LWR AND HCLWR NEUTRON SPECTRA

As these reactor types are out of the scope of the present meeting, we will not detail the corresponding work programme.

Note only, that for these reactor types, both irradiation experiments (pool reactor MELUSINE at GRENoble) and experimental analysis (JEF-2 library) are performed.

3. REACTOR STUDIES

Reactor studies are devoted to actinide burners (LWR, FBR) with homogeneous and heterogeneous recycling.

Previous studies have shown the order of magnitude of the potential incineration in a large fast reactor.

Progressively, the FBR destroys the actinides initially loaded (as well as those produced by its own operation), and after approximately 6 to 7 cycles, the actinide amounts reach an equilibrium, with a reduction by a factor of 10 of the initial quantity of Np-237.

In the PHENIX reactor, a first demonstration of this potential has been made (the SUPERFACT experiment [2]). Two types of pins have been irradiated for 380 FPD, each type being representative of the two recycling modes :

- homogeneous recycling, i.e. Np-237 and Am-241 (2 %) mixed to the fuel of standard UO_2/PuO_2 pin ;

SUPERFACT 1

19 PINS PHENIX CLUSTER

pins location

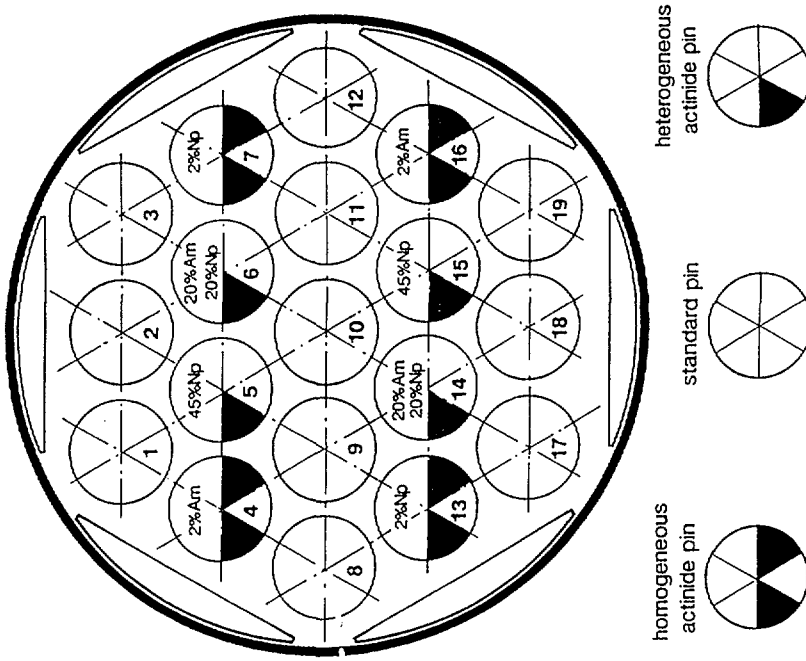


Figure 1

- heterogeneous recycling, i.e. the Np-237 and the Am-241 are added in large quantities (45 % and 20 %) to a UO_2 pin.

Present studies are focused :

- 1] on homogeneous recycling in LMFBRs with different fuel types (e.g. oxide and nitride), and of different sizes, and on heterogeneous recycling in special subassemblies (e.g. in the periphery of a standard core, or in the decoupling regions of a modular

core) where the actinides/fission products can be irradiated during all the life of the power plant. Preliminary results indicate a reduction of the Np-237 quantity of at least 20 %, when irradiated in the external radial blanket of a SUPER-PHENIX type reactor ;

2] on homogeneous recycling in LWR with different moderating ratios. Heterogeneous recycling is also investigated.

A special attention is given to the reactivity coefficients, which can potentially be worsened by the addition of minor actinides, and to the optimization of the spatial power shapes.

For long-lived fission products (such as Tc-99 and I-129), for which the epithermal capture process is the main mechanism of transmutation, it is explored the feasibility to transmute them at the periphery of a FBR core in special subassemblies containing a moderating material.

This technique was successfully proved in PHENIX for the production of Co-60 from Co-59 targets surrounded by a calcium hybride moderator.

Complementary studies are performed on more advanced burner systems (accelerator, hybride systems, etc...), as well as studies on fuel cycle consequences of the introduction of long-lived wastes burner reactors.

4. FUEL STUDIES

The SUPERFACT experiment has demonstrated the good performance of the (UNp)₂O₃ fuel and some problems (e.g. the production thermal conductivity) for the (U, Am, Np)₂O₃ fuel. Present studies are devoted to nitride fuels (for the homogeneous recycling) and to different types of inert matrices for the heterogeneous recycling. A matrix NpO₂ + MgO has been already experienced successfully in the past, but other matrices (ZrO₂, Al₂O₃) are investigated.

Also, a joint experiment with ITU/Karlsruhe and CRIEPI/Japan, is planned for irradiation of metallic fuel pins (actinides and some rare earths in UPuZr matrix), in PHENIX.

Finally, a reflexion is being made, about the future of SUPER-PHENIX. In this context and as expressed very recently by French Officials, the SUPER-PHENIX plant could be used as a waste incinerator. In that respect, NERSA (owner of the plant) must propose some specific experiments : this work is just at the early beginning.

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"Int. Conf. on Fast Reactors and Related Fuel Cycles", KYOTO, Oct 28-Nov.1, 1991

MINOR ACTINIDES RECYCLING IN AN EFR TYPE FAST NEUTRON REACTOR

H. SZTARK
Framatome,
Lyon

G. VAMBENEPE, J. VERGNE
Electricité de France,
Lyon

A. ZAETTA
Commissariat à l'énergie atomique,
Centre d'études nucléaires de Cadarache,
Saint-Paul-lez-Durance

France

Abstract

In the framework of the french nuclear programme, based on PWR type of reactors associated with reprocessing of spent fuel elements, the management of the nuclear wastes and particularly the Minor Actinides has to be defined.

Among the possible solutions, the transmutation of these long lived elements using Fast Neutron Reactors such as SPX and EFR is envisaged. This paper aims at presenting the studies which are being performed on an EFR type of core, 1500 MWe, oxide fuel with very high burn-up, in the case of minor actinides homogeneously distributed inside the fuel. The efficiency of the Minor Actinides transmutation as a function of the initial content, the consequences of the insertion of these Actinides on the major core parameters, mainly the safety parameters are presented. Finally, the consequences of this Minor Actinides recycling on the long term radiotoxicity are given.

1. INTRODUCTION

In the framework of the french nuclear programme, based on PWR type of reactors associated with reprocessing of spent fuel elements, the management of the nuclear wastes and particularly the Minor Actinides has to be defined.

Among the possible solutions, the transmutation of these long lived elements using Fast Neutron Reactors such as EFR is envisaged. The present study aims at presenting the capability of a EFR type of core, 1500 MWe, oxide fuel with very high burn-up, to transmute Minor Actinides homogeneously distributed inside the fuel. The efficiency of the Minor Actinides transmutation as a function of the initial content, the consequences of the insertion of these Actinides on the major core parameters, mainly the safety parameters, are presented. Also, the consequences of this Minor Actinides recycling on the long term radiotoxicity is discussed.

2. CONTEXT OF THE STUDIES

In order to work with well established material quantities, the present studies will be centered around the french nuclear programme.

Today, the french nuclear electricity generation rate is somewhat higher than 300 TWh electrical per year ; it is expected to increase slightly and to reach a rate of about 450 TWh per year around 2010. This nuclear electricity is mainly produced by PWR type reactors, using uranium oxide fuelled assemblies.

With such type of fuel, the heavy nuclei production rates at the reactor output are given here below, per each TWh electrical produced :

- (a) 2900 kg of " Reprocessed Uranium " containing :
98.6 % U238 ; 0.9 % U235 ; 0.5 % U236,
- (b) 35 kg of plutonium containing :
2.9 % Pu238 ; 54.3 % Pu239 ; 22.9 % Pu240 ; 14.3 % Pu241 ; 5.7 % Pu242,
- (c) 2.5 kg of Minor Actinides with :
64 % Np237 ; 8 % Am241 ; 20 % Am243 ; 6 % Cm244,
- (d) 135 kg of fission products, with in particular :
- 3.3 kg of Te99
- 0.8 kg of I129.

All these quantities result from fuel burn-up calculations, which are then compared with results coming out from the analysis of samples from some well known irradiated fuel subassemblies.

For an averaged electricity generation rate of 400 TWh per year, we will then have the following heavy nuclei production every year, assuming that only uranium oxide is used as fuel :

- 1160 tons of " Reprocessed Uranium "
- 14 tons of plutonium,
- 1 ton of Minor Actinides.

Presently, only a few part of the PWR's are recycling plutonium, and only partially in the core (i.e. only 30 % of MOX fuel assemblies per reload) : in 1991, it was the case for 5 of the 900 MW power plants, and it is expected that 16 plants will recycle (partially) plutonium by the end of the century.

In these conditions, it is foreseen that the amount of unused plutonium and Minor Actinides will approach 300 tons before 2010, only for France, the Minor Actinides themselves representing some tens of tons.

3. STUDIES OF MINOR ACTINIDES TRANSFORMATION IN AN EFR TYPE OF CORE

3.1. CALCULATION MODEL

For the purpose of the present studies, the EFR core [1] was taken as the reference, with the following characteristics :

- total thermal power : 3600 MWth,
- homogeneous core concept with two radial enrichments,
- 388 fuel subassemblies, distributed respectively into 208 and 180 between inner and outer zones,
- 1 meter fissile height,
- maximum fuel burn-up : 20 at %,
- fuel management in 5 batches, with a 320 efpd cycle length and a total fuel residence time of 1600 efpd.

The fuel itself is made of mixed oxide UO₂-PuO₂, with the following compositions for the reference case :

- depleted uranium : 0.25 % U235 ; 99.75 % U238,
- plutonium coming out from a PWR :
2.8 % Pu238 ; 54.4 % Pu239 ; 22.8 % Pu240 ; 11.8 % Pu 241 ; 7 % Pu 242, with a 1.2 % of Am 241.

With this fuel composition, the fuel enrichments in plutonium oxide are respectively 16.8 % and 21.9 % for both radial zones.

When adding Minor Actinides in the core, these new isotopes are homogeneously distributed all over the fuel, in replacement of heavy atoms of uranium and plutonium. The fuel enrichment in PuO₂ is adjusted in such a way to get the same service as for the reference fuel :

- same burn-up, which means same cycle length and overall fuel residence time,
- same reactivity at end of equilibrium cycle.

The core calculations are performed in a 2 dimension model in RZ geometry, using the ERANOS code system and the CARNIVAL IV cross section data set, with the standard 25 energy groups. This data set is completed by the JEF1 cross sections for the Minor Actinides.

The 5 batches fuel subassembly management is not simulated explicitly in the calculations. The whole core is burnt from beginning of life up to end of life (1600 efpd), the equilibrium core cycle being represented respectively at 640 efpd and 960 efpd. With respect to the fuel itself, one lifetime in the core will represent one fuel cycle.

3.2. MINOR ACTINIDES AUTO-RECYCLING

In a first step, the auto-recycling of Minor Actinides (M.A.) was studied in order to determine the burning capability of the EFR core. For this purpose, the following assumptions were made :

- for the first fuel cycle, a given amount of M.A. is inserted inside the fresh fuel, homogeneously distributed all over the core : 2 %, 5 % and 10 % first of Np237 and then of Am241. Indeed, these two isotopes are the main contributors in the PWR spent fuel,
- the complete core is burnt for one complete fuel cycle (1600 efpd),
- the burnt fuel is supposed to be reprocessed, the fission products being extracted and replaced by a mixture of heavy atoms (U, Pu) without any new M.A., in such a way to

get the correct reactivity, and the same heavy metal mass as for the previous beginning of fuel cycle,

- the out of pile isotopic evolution is simulated by taking 1 year for fuel fabrication and 1 year for fuel cooling and reprocessing,
- the same process is being repeated for each new fuel cycle.

With these assumptions, one can establish the burning rate and the burning speed for each of the two isotopes individually, and also for the total amount of M.A. taking into account those which are produced in the fuel.

The results of these evolutions are given on Tables I and II respectively for Np237 and Am241, in terms of masses of the involved isotope, and in terms of the sum of the 3 isotopes leading to Np237 ; Np 237 + Am241 + Cm245. On Fig. 1 and 2 are plotted the cases of 2 % Np237 and 2 % of Am241.

TABLE I. EVOLUTION OF THE M.A. MASSES FOR THE CASE OF Np237 INSERTION. AUTO-RECYCLING

Number of cycles	Masses in kg					
	2 % Np237		5 % Np237		10 % Np237	
	Np7	Σ (3 iso)*	Np7	Σ (3 iso)*	Np7	Σ (3 iso)*
0	902	950	2257	2306	4518	4569
2	253	408	610	766	1251	1409
4	93	251	184	338	358	508
6	52	219	75	237	120	277
8	41	213	47	216	58	223

* Σ (3 iso) = sum of Np237 + Am241 + Cm245

TABLE II. EVOLUTION OF THE M.A. MASSES FOR THE CASE OF Am241 INSERTION. AUTO-RECYCLING

Number of cycles	Masses in kg					
	2 % Am241		5 % Am241		10 % Am241	
	Am1	Σ (3 iso)*	Am1	Σ (3 iso)*	Am1	Σ (3 iso)*
0	1027	1027	2336	2336	4625	4625
2	339	337	672	714	1328	1383
4	182	234	245	301	428	492
6	155	211	164	222	210	273
8	155	211	154	211	163	222

* Σ (3 iso) = sum of Np237 + Am241 + Cm245

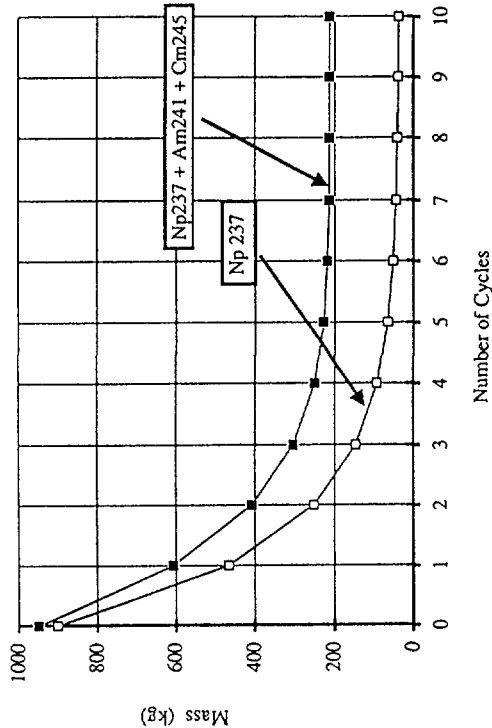


FIG. 1. Evolution of the M. A. masses for the case of 2 % Np237 insertion.

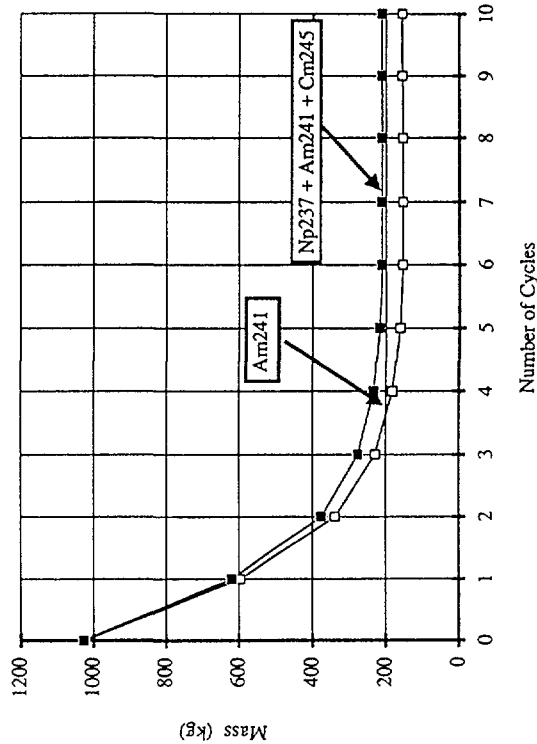


FIG. 2. Evolution of the M. A. masses for the case of 2 % Am241 insertion.

One can see that whatever is the initial actinide and its amount in the fresh fuel, the final composition of the fuel is the same after about 5 to 6 fuel cycles, this final composition depending only on the core characteristics. Indeed, this final composition is constant even if there are no M.A. inside the fuel at the beginning. This equilibrium state is obtained when the production of M.A. inside the core is equal to their destruction.

The burning rate of this type of core can be established to about :

- 95 % to 99 % for the Np237 alone,
- 85 % to 96 % for the Am 241 alone,
- 75 % to 95 % for the sum Np237 + Am 241 + Cm245.

Concerning the burning speed, whatever is the initial amount of M.A. inside the fuel, almost half of this amount is burnt after one single fuel cycle. For the sum of the three actinides leading to Np237, this ratio is of the order of 40 to 50 %.

3.3. INFLUENCE OF THE MAIN CORE CHARACTERISTICS ON THE BURNING CAPABILITY

In order to check the influence of the core characteristics on the burning capabilities and also on the equilibrium composition of the fuel, comparative calculations were performed with the reference EFR core as defined previously, and with a core of same total power but with the following modifications :

- core diameter decreased by about 20 %, corresponding to much smaller fuel pellet diameter, with the same core height,
- in correspondance, the fuel volume fraction is decreased by about 10 % relative, and the fuel enrichment increased by about the same amount,
- for the same peak burn-up, the fuel lifetime goes down to about 980 efpd.

Due to the increased fuel enrichment, the neutron spectrum is harder, but this has no influence on the M.A. burning capability. For the two cases corresponding to the insertion of 2 % of Np237 and 2 % of Am241, the transmuted masses during the first fuel cycle in the core are respectively, for the same energy produced and for the fuel at the reactor output :

- for the reference core : 8.3 kg/TWh for Np237,
9.0 kg/TWh for Am241,
- for the higher enrichment core : 8.1 kg/TWh for Np237,
9.1 kg/TWh for Am241.

So, the transmuted masses of M.A. are identical in both cores ; indeed, in the core with the higher initial enrichment, the M.A. production coming from the plutonium isotopes is slightly higher, which compensates the increased mass fissioned with the harder neutron spectrum.

On the other hand, the fuel composition at equilibrium, after 5 to 6 fuel cycles, can be slightly modified due to the different core characteristics, mainly because of the different fuel enrichments. The equilibrium compositions after 6 fuel cycles are respectively :

Reference core :

3.7 % Pu238 ; 48.7 % Pu239 ; 36.3 % Pu 240 ; 5.5 % Pu241 ; 5.8 % Pu242

Higher enrichment core :

3.9 % Pu238 ; 45.2 % Pu239 ; 37.6 % Pu240 ; 6.2 % Pu241 ; 7.1 % Pu242.

With the higher enrichment core, the plutonium quality in terms of equivalent plutonium 239 is slightly lower, but in both cases the isotopic composition is stabilized after 5 to 6 fuel cycles. Also, for both cores, as the plutonium quality is decreasing when recycling it, the fuel enrichment has to be increased at each new fuel cycle : this increase in enrichment is of the order of 10 % relative between the first cycle and the last one where the fuel composition is stabilized, and so is the fuel enrichment.

This shows that whatever is the type of core, with or without M.A. recycling, the Fast Neutron Reactor allows indefinite recycling of plutonium.

4. CONSEQUENCES OF M.A. RECYCLING ON THE CORE PERFORMANCES

4.1. M.A. MASS LIMITATION

For the reference EFR core as defined on § 3 - 1, the influence of the insertion of different amounts of M.A. on the main core performances is shown on Table III.

TABLE III. INFLUENCE OF M.A. INSERTION ON THE MAIN CORE PERFORMANCES

M.A. content	$\Delta\rho(0)^a$ (pcm)	$\Delta\rho(BU)^b$ (pcm)	$\Delta\rho(\text{rods})^c$ (%)	$\Delta\rho(\text{Na})^d$ (%)	$\Delta\rho(\text{Dop})^e$ (%)
2 % Np237	- 2300	+ 1400	- 4	+ 20	- 20
5 % Np237	- 4800	+ 3500	- 9	+ 40	- 40
10 % Np237	- 10000	+ 6100	- 17	+ 50	- 60
2 % Am241	- 2600	+ 2300	- 4	+ 20	- 20
5 % Am241	- 6200	+ 5400	- 10	+ 40	- 40
10 % Am241	- 11200	+ 9300	- 19	+ 50	- 70
2.5 % Np237 + 2.5 % Am241	- 5800	+ 4300	- 10	+ 40	- 40

^a $\Delta\rho(0)$: initial reactivity (fresh core),

^b $\Delta\rho(BU)$: reactivity swing with burn-up,

^c $\Delta\rho(\text{rods})$: rods efficiency,

^d $\Delta\rho(\text{Na})$: sodium void reactivity at beginning of life,

^e $\Delta\rho(\text{Dop})$: doppler effect at beginning of life.

In this comparison, the fuel enrichment has been adjusted in such a way to get the same core reactivity at beginning of life, instead of end of equilibrium cycle.

As one can see on Table III, the effects related to the insertion of a given amount of Np237 or Am241 are very similar, and variations are almost linearly dependant on the initial amount. The main tendencies are as follows :

- for a given plutonium enrichment, the initial core reactivity is significantly reduced, Np237 and Am241 having large neutron capture rates. As the present comparison is made at same initial reactivity, this would lead to an increased enrichment by 5 % to 15 % depending on the M.A. amount,
- the reactivity swing over the fuel lifetime is strongly decreased, due to the production of highly efficient isotopes, in terms of equivalent plutonium 239. In fact, if one compares the different effects with the same reactivity at end of equilibrium cycle instead of beginning of life, this would lead to a decrease of the initial core enrichment instead of the increase quoted just before,
- the control rod efficiency is decreased by about 5 % to 15 %, due to the harder neutron spectrum,
- the sodium void reactivity for the fissile zone is strongly increased, by about 20 % to 50 % depending on the initial M.A. amount, with correspondingly a reduction of the Doppler coefficient by about the same amount.

It is essentially this later effect which limitates the amount of M.A. that could be inserted in such an EFR core, homogeneously distributed over the whole core. This limitation is of the order of 2 % to 2.5 % of the total heavy metal mass of the fissile zone, which means about 0.7 to 1 ton of M.A. in the EFR core. The corresponding increase on sodium void reactivity is of the order of 20 % relative, which would need anyway specific improvements on the core itself but such improvements seem possible.

4.2. BURNING POTENTIAL OF AN EFR CORE

The mass of about 1 ton of M.A. (Np237 + Am241) corresponds approximately to the annual production of the french PWR park (400 TWh annual), fed with uranium oxide. For such a PWR park, and with an auto recycling of the type described on § 3 - 2, a new EFR plant of 1.5 GWe every year would be necessary. This, after 5 to 6 fuel cycles, which corresponds roughly to the plant lifetime, would allow to reduce the total amount of M.A. (Np237 + Am241 + Cm245) by about 80 % as compared to the production of the LWR's. Anyway, this type of autorecycling is not very realistic, since it is always the same fuel which feeds the reactor, with only a complement of plutonium.

A more realistic mode of recycling M.A. would be to maintain a constant percentage of M.A. in the fuel, by adding M.A. at each new fuel cycle to complete the amount to the level of 2 to 2.5 % : this is the case which is described in the next chapter.

5. CONSEQUENCES OF M.A. RECYCLING ON THE RADIOTOXICITY OF WASTES

A management strategy based on wastes (Np + Am) produced by a nuclear park seems important to analyse in order to compare the ability of the different systems to reduce the potential radiotoxicity source of wastes.

5.1. REFERENCE PARK

The reference park is always the french one, consisting on PWR's fed with uranium oxide fuel at 3.25 % enrichment, and generating 400 TWh per year.

At the fuel end of life, we consider that :

- the fuel is reprocessed, the separated plutonium being used once more in PWR or in FBR ; only the Pu losses estimated to 0.3 % during reprocessing operations join the wastes,
- all the Minor Actinides (Np + Am + Cm) are considered as wastes.

5.2. MINOR ACTINIDES TRANSMUTATION

The park is now a mixed park composed with a proportion of some standard PWR (the same as previously described) and of some amount of EFR which make the transmutation working. The total power (PWR + EFR) settled stays at the same level : generation of 400 TWh per year. The EFR fuel contains a 2.5 % amount of Minor Actinides (Np + Am), homogeneously distributed in the whole core.

The self-recycling is simulated in the following manner :

- core burnt in one fuel cycle of 1600 efpd,
- cooling time of 1 year,
- reprocessing simulation in :
 - * removing from the fuel all the fission products,
 - * adding a Np + Am mass to re-obtain the maximum allowed content,
 - * completing with a U-Pu mixture to keep the total mass, and the correct reactivity level,
- a period of 1 year for fuel manufacturing, and so on till an equilibrium situation is reached.

The needed number of EFR plants is determined so that the FBR park yearly consumption in Np + Am equals the PWR park yearly production.

In this management strategy, wastes find their origin in :

- those occurring when partitioning the Np and Am from the PWR and FBR fuels : the partitioning quality for Np and Am was taken equal to 90 %,
- Pu losses estimated to 0.3 % when reprocessing the PWR and FBR fuels,
- the total Cm produced by both PWR and FBR parks.

5.3. RESULTS

With the previous hypothesis, the proportion of the incinerator FBR park was found to be about 20 % of the total settled power.

The comparison between the wastes annual mass for the reference and the incinerator park shows that :

- the Np mass is reduced by about 80 %,
- the Am mass is reduced by about 75 %,
- the Cm mass, mainly the Cm244, is increased by a factor of 4.

The consequences in terms of radiotoxicity are given in the following table, expressed in terms of radiotoxicity reduction factors as a function of time :

Time (years)	10 ²	10 ³	10 ⁴	10 ⁵	10 ⁶	10 ⁷
Radiotoxicity reduction factor	2.0	2.3	1.1	3.0	4.5	4.3

Note that a strategy of Np + Am incineration is able to reduce the wastes radiotoxicity as compared to the reference strategy (fuel closed cycle) by a factor 2 to 5 on the whole time scale, excepted around 10⁴ years where the gain is almost zero. This is due to the Cm244 contribution which represents about 60 % of the total radiotoxicity. This higher contribution is understandable by the large mass generated by Am transmutation and by the fact that at this time, its elementary radiotoxicity is highly superior by a factor of 100 as compared to the elementary radiotoxicity of the other Minor Actinides.

More generally, the analysis of each isotope contribution on the whole radiotoxicity shows that :

- at short times (10² to 10³ years), the Am241 and the Cm244 represent 80 % of the total radiotoxicity,
- at 10⁴ years, the Cm244 is higher (about 60 %),
- at 10⁵ years, the Am combined with the Np amounts to 60 % of the entire radiotoxicity, but with a noticeable contribution of the Pu239 for about 25 %,
- for very long times (10⁶ to 10⁷ years), the Np predominance (about 50 %) combined with the Am241 (about 35 %) is settled up.
(note : the radiotoxicity contribution is associated to the original isotope).

These results show that the M.A. are the major contributors to the whole toxicity of wastes, as compared to plutonium and also to fission products for long times.

To reduce this toxicity, the Np and Am partitioning quality has to be improved and the Cm whose contribution is important at 10⁴ years but also at short time has to be recycled. With a

different hypothesis concerning the partitioning quality of M.A. (99 %) and by recycling Cm as Np and Am, we observe a reduction of the radiotoxicity on the entire time scale by a factor of about 7 to 30, as shown in the following table and on Fig. 3 :

Time (years)	10 ²	10 ³	10 ⁴	10 ⁵	10 ⁶	10 ⁷
Radiotoxicity reduction factor with a partitioning quality of 99 % EFR (Np + Am)	3.6	4.5	1.4	7	20	14
EFR (Np + Am + Cm)	14	16	7	8	33	25

Then the analysis of each isotope contribution shows that Pu represents 60 % to 80 % of the whole radiotoxicity till 10⁵ years and around 50 % after. So to improve again the radiotoxicity reduction factor, it is necessary to improve now the separation of Pu more than that of the M.A.

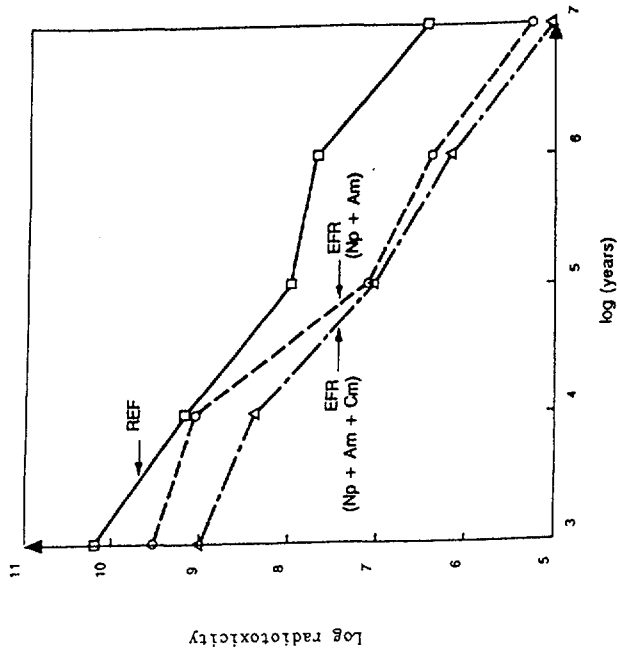


FIG. 3. Evolution of the radiotoxicity as a function of time.

6. CONCLUSION

These preliminary studies have shown that the use of a Fast Neutron Reactor such as EFR to recycle the Minor Actinides coming out of the reprocessing of LWR spent fuel can be an intermediate solution to reduce the long term radiotoxicity of long lived wastes. However, a significant reduction of the radiotoxicity needs also improvements on the reprocessing process, mainly concerning the partitioning quality for both Minor Actinides and plutonium.

As regards the core, the reinsertion of Minor Actinides inside the fuel has some benefits on the core physics, for example a strong reduction of the reactivity swing, but also drawbacks on the main safety parameters such as an increased sodium void coefficient. Due to these drawbacks, the Minor Actinides amount that can be inserted has to be limited to about 2 to 2.5 % of the total heavy metal mass, with as a consequence a limited transmutation capability : the percentage of FBR in the nuclear park must be of the order of 20 %, in order to compensate the LWR production in Minor Actinides.

In a further step, it is foreseen to study an heterogeneous mode of recycling the M.A. in a FBR, in specific target subassemblies located either in central core positions or in peripheral positions. Indeed, this would simplify the manufacturing process by separating the plutonium subassemblies from these high activity targets, but the burning potential as well as the influence of these specific subassemblies has to be established.

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PARTITIONING AND TRANSMUTATION RESEARCH AND DEVELOPMENT PROGRAM (OMEGA) IN JAPAN

T. MUKAIYAMA

Japan Atomic Energy Research Institute,
Tokai-mura, Naka-gun, Ibaraki-ken,
Japan

Abstract

In the Japan Atomic Energy Industry Forum report on the waste management of long-lived nuclides the importance of research and development for partitioning and transmutation (P-T) of long-lived nuclides was pointed out as long term efforts in developing a complete system for radioactive waste management. Based on this, the interested Japanese organizations proposed to initiate a major R&D program on P-T. The development of P-T technology was deemed to be quite an interesting subject for ongoing investigation from the perspectives of potential utilization of resources and possible long term advances in radioactive waste management.

The proposed project was called "OMEGA". The Japanese Government proposed an international cooperation for information exchange to cover the areas of nuclear physics, reactor physics, advanced technologies and physico-chemistry. In addition, advancement of technologies such as laser and accelerator technology, will provide spinoffs for other fields of science and technology.

1. INTRODUCTION

In 1973, the Japan Atomic Energy Industry Forum published the report on the waste management of long-lived nuclides after the two years assessment studies by the groups of scientists and engineers in Japan. In that report entitled "A closed system for radioactivity", the importance of research and development for partitioning and transmutation(P-T) of long-lived nuclides was pointed out as long term efforts in developing a complete system for radioactive waste management.

Even after the pessimistic conclusions for P-T of high level radioactive waste(HLW) drawn by Oak Ridge National Laboratory(1980), IAEA(1982), etc., small number of groups in Japan, as well as Europe, the United States etc. continued their studies in P-T as basic research.

Based on their studies, the interested Japanese organizations proposed to initiate a major R&D program on P-T. The development of P-T technology was deemed to be quite an interesting subject for ongoing investigation from the perspectives of potential utilization of resources and possible long term advances in radioactive waste management.

In 1987, Japan's Atomic Energy Commission(AEC) concluded that the potential benefits from the use of some elements in fission products and from recycling minor actinides for power

generation could be achieved provided that a well planned, efficient and effective R&D program could be formulated. The AEC then submitted in October 1988 a report entitled "Long Term Program for Research and Development on Nuclide Partitioning and Transmutation Technology", which plots a course for technological development up to the year 2,000.

The program is called "OMEGA" which is the acronym derived from Options Making Extra Gains from Actinides and fission products.

The R&D programs were jointly stimulated by the collaborative efforts of the Japan Atomic Energy Research Institute(JAERI) and Power Reactor and Nuclear Fuel Development Corporation (PNC). In the public sector, the Central Research Institute of Electric Power Industry(CRIEPI) also has been carrying out R&D on this subject.

The Japanese government (represented by Science and Technology Agency, STA) proposed an international cooperation for information exchange to cover the areas of nuclear physics, reactor physics, advanced process technologies and physico-chemical characterization relevant to P-T technology under the framework of the OECD Nuclear Energy Agency in January 1989. The first information exchange meeting on this subject was held in Japan in November 1990. Eleven OECD member countries and two international organizations, namely IAEA and Joint Research Center of CEC, participated in the meeting.

The program is conceived as a research effort to pursue benefits for future generations through long term basic R&D, and is not to seek a short term alternative for established or planned fuel cycle back-end policies. The program is expected to serve to revitalize nuclear R&D in general, and also to attract capable young researchers dedicated to bringing the nuclear option into the 21st century in a healthy state. In addition, advancement of technologies such as laser and accelerator technology, as advocated in this program, will provide spinoffs for other fields of science and technology.

2. OMEGA PROGRAM IN JAPAN

The program is to be proceeded in two steps: the phase-I and II. The phase-I covers a period up to about 1996, and the phase-II covers a period from about 1997 to about 2000. In general, the basic studies and testing are to be conducted in the phase-I to evaluate various concepts and to develop required technologies. In the phase-II, engineering tests of technologies or demonstration of concepts are planned. After 2000, pilot facilities will be built to demonstrate the P-T technology.

The program covers the following fields.

2.1 PHYSICAL AND CHEMICAL PROPERTIES OF MINOR ACTINIDES AND FISSION PRODUCTS

Reliable data base of minor actinides and long-lived fission products is indispensable to proceed the program. Underlying studies will improve understanding of the science and technology for separation and recovery of these nuclides from HLW, for fabrication of actinide fuels for recycling to reactors or accelerator driven systems for transmutation, and for utilization of these nuclides. Nuclear data and thermodynamic data of these nuclides are measured, compiled and evaluated for reactor physics and materials development. Following items are to be studied;

- Chemical properties and behavior of actinide species in aqueous and organic solutions
- Analytical techniques and methods
- Physical and chemical properties of actinide compound
- Collection and evaluation of nuclear and thermodynamic data

2.2 PARTITIONING

Advanced separation technology of minor actinides and fission products is to be developed on the basis of current trends in the chemical separation processes. Both wet and dry processes are to be developed. This would include application of new extracting solvents, laser induced separation, and sublimation and volatilization processes. Studies for utilization of separated nuclides will also be included. The following items are to be studied;

- Partitioning for HLW
- Partitioning in the reprocessing process
- Recovery of platinum group metals
- Cost estimation of a process

2.3 TRANSMUTATION

Recently new approaches such as minor actinide transmutation using actinide burner reactors, metal fuel FBRs(LMR) with dry reprocessing, intense beam proton accelerator as neutron source have been studied. Optimization of minor actinide recycling into Pu-LWR and MOX-FBR is to be studied. Studies of transmutation of Sr-90, Cs-137 with an electron accelerator are also proposed. Fields to be studied are the followings;

- Nuclear data and fuel property data of minor actinides
- Development of computer code
- System design study
- Reactor fuel and accelerator target development
- Fabrication technology of the fuel and target materials
- Development of high power accelerator for transmutation
- Cost estimation of R&D, construction, and fuel cycle

The areas covered by the program and R&D activities are illustrated in Fig.1.

3. RESEARCH AND DEVELOPMENT ACTIVITIES

Under the framework of the OMEGA program, the following R&D activities are under way at JAERI, PNC and CRIEPI.

3.1 PHYSICAL AND CHEMICAL PROPERTIES OF MINOR ACTINIDES AND FISSION PRODUCTS

As the basis of R&D for P-T, the following studies were carried out in the field of chemistry and nuclear data at JAERI.

a) *Destructive analysis of PWR spent fuel*[1]:
 PWR spent fuels with burnup between 6.9 and 34.1 GWd/MTU were analyzed by mass spectrometry, alpha-ray and gamma-ray spectrometry. The results were compared with the calculation using nuclear data library JENDL-2 and two burnup codes, ORIGEN-2 and JAERI's SRAC-FPGS. The intercomparison of analysis of nuclides generated from minor actinide isotopes irradiated in a fast reactor is to be carried out under the cooperative program with ORNL.

b) *Flow-coulometry of U, Np, Pu ions*[2]:
 The oxidation-reduction behavior of U, Np and Pu ions in acidic aqueous solutions was studied by flow-coulometry with multi-step column electrodes. The rapid preparation and determination of the ion of desired oxidation state were developed.

c) *Photo-acoustic spectroscopy induced by Fourier transformed laser beam*[3]:
 Fourier transform laser-induced photo-acoustic spectroscopy (FT-LPAS) has been developed for speciation of actinide elements in aqueous solutions, and ultraviolet, visible and near-infrared photo-acoustic spectroscopy (UV-VIS-NIR PAS) and Fourier transform infrared photo-acoustic spectroscopy (FT-IR PAS) for those in solid phases. These methods were applied to the speciation of U(VI) in $\text{NaHCO}_3/\text{NaClO}_4$ solution/precipitate systems with satisfactory result. These methods are also applicable to speciation of solid phases, especially of amorphous materials.

d) *Electrolytic deposition of actinides in molten salt*:
 The electrolytic deposition study for TRU separation using chloride salts is under way with emphasis on reaction kinetics and equilibrium measurements.

e) *Nuclear data*:
 The latest version of Japanese Evaluated Nuclear Data Library 'JENDL-3'[4] contains the nuclear data of actinides up to Fr-255. Since the accuracies of these nuclide data are uncertain, the revision of these data are planned under NEACRP/NEANDC International Evaluation Cooperation. The data of Np-237 and Am-241 are being reevaluated using the available data of integral experiments as the first step of the cooperation. For neutrons which are emitted in the charged particle induced reactions, evaluation of the nuclear data for neutron energy between 20MeV and 50MeV are under way. For neutron above 100MeV, validity of various theoretical calculation codes are examined using the experimental data. For the future evaluation high energy nuclear data, the neutron emission data are being collected both for thin and thick target experiments.

3.2 PARTITIONING

Wet processes of partitioning are developed at JAERI and PNC. Dry processes are developed at CRIEPI and PNC.

A. Studies at JAERI

a) *Three group partitioning and cost estimation*:
 From 1973 to 1984, a partitioning process was developed for separating elements in HLW into three groups; TRU, Sr-Cs and others. The process consists of three steps; the first is solvent extraction of U and Pu with tributylphosphate (TBP) and the second is solvent extraction of Am

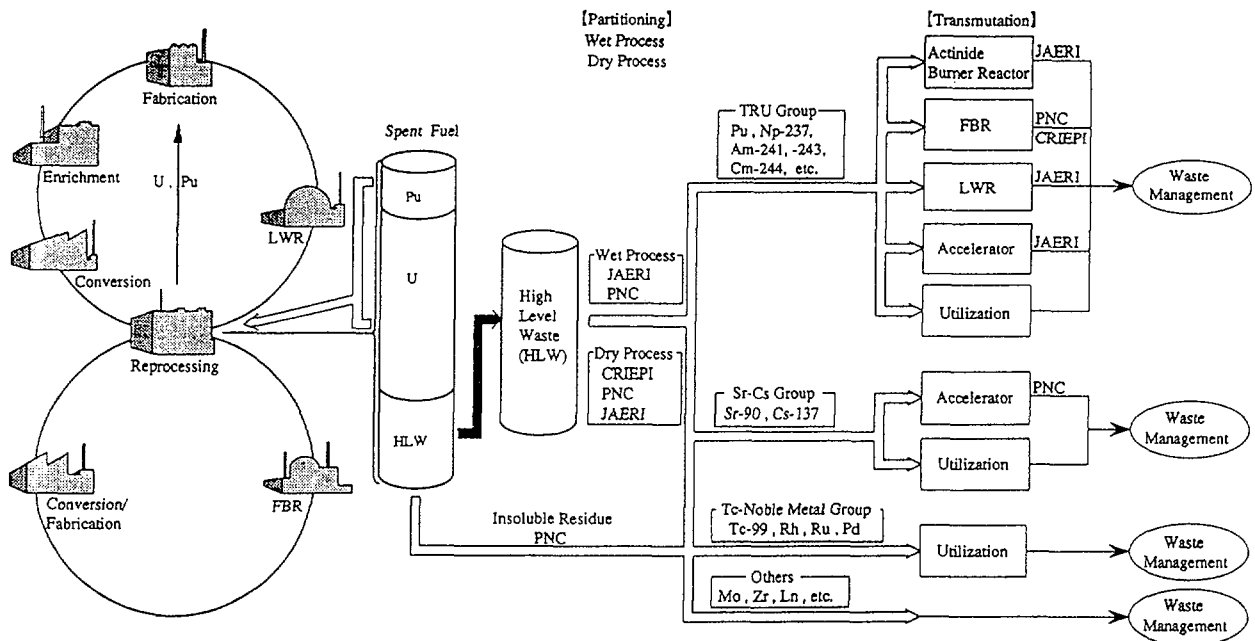


Fig. 1 P-T R/D ACTIVITIES UNDER OMEGA PROGRAM

and Cm with diisododecylphosphoric acid (DIDPA), and the third is adsorptions of Sr and Cs with inorganic ion exchangers. The process was demonstrated by using actual HLW solution. More than 99.99% of Am and Cm were extracted with DIDPA[5]. The preliminary assessment study indicated that total volume of solid material is reduced to less than half of that of vitrified waste, 12wt% of which is fission products oxide from HLW, and that the cost for construction and operation of a partitioning plant is less than 5% of that of a Purex plant, provided that a partitioning plant is operated in connection with the Purex plant[6].

b) Development of four group partitioning:

After 1985, a four group partitioning process has been developed in which a step for separating Tc-platinum group was developed in addition to the three group separation. An effective method for separating TRU, especially Np, and Tc, was developed[7].

c) Separation of Np:

Experiments on a counter-current continuous extraction using a mixer-settler were carried out to determine the conditions for extracting Np(V) with DIDPA. The addition of hydrogen peroxide accelerates the extraction of Np. More than 99% of Np was extracted when hydrogen peroxide was added so as to compensate for its decomposition[8].

The effect of radiolysis of DIDPA on extraction of Np(V) was simulated by irradiating DIDPA with Co-60 γ -ray. Little change in extraction rates of Np was observed when the solvent was irradiated with the dose up to 1 MGy[9].

d) Separation of Am and Cm from rare earths:

Rare earths as well as minor actinides are extracted with DIDPA. Recently it was found that Am and Cm are preferentially back-extracted by diethylenetriaminepentaacetic acid as a complexing agent in aqueous phase while rare earths remain in the DIDPA solvent and that all of rare earths and TRU such as Np and Pu remaining in the solvent are back-extracted with 3M nitric acid and an oxalic acid solution, respectively.

e) Separation of Tc:

Two methods has been developed to separate Tc; precipitation by denitrating HLW and adsorption with active carbon. More than 95% of Tc in a simulated HLW was recovered as precipitate by denitration of HLW by adding formic acid to make the solution pH above 2.0. The use of the active carbon column resulted in the quantitative adsorption of Tc from a simulated HLW by adjusting the solution to a 0.5M nitric acid and the elution of Tc from the column was achieved quantitatively by the use of an alkaline thiocyanate solution as eluant[10].

f) Demonstration of four group partitioning:

The four group partitioning process is to be tested with actual HLW at NUCEF (Nuclear Fuel Cycle Safety Engineering Research Facility) which is under construction. A cold operation of the facility is scheduled in 1993.

g) Integration of partitioning process into Purex process[12]:

To integrate these partitioning processes into Purex process, a reductive stripping with iso- and normal-butylaldehyde was successfully applied for selective separation of Np and Pu from the codecontamination product stream[11]. A study to develop a system for automatic measurement in TRU extraction process was commenced in 1990 under the co-operative program with AEA-Harwell. To support these works, a TRU valency monitor based on the photoacoustic spectroscopy is under development.

B. Studies at PNC

B.1 Wet Process

a) Extraction tests based on the "TRUEx" flow sheet:

Extraction tests have been conducted to recover minor actinides from HLW, based on the "TRUEx" flow sheet. Fundamental batchwise tests were carried out to determine the distribution ratios of minor actinides and fission products. Based on the above results, counter-current flow sheet tests with small scale mixer-settlers were also conducted using the actual HLW derived from FBR fuel reprocessing tests[13]. Future studies are planned to improve the process for back-extraction of Pu, to develop a separation process of minor actinides/rare-earth elements, to improve extraction efficiency of Np, to clarify the conditions leading to third phase formation and so on.

b) Fuel cycle including minor actinides separation process:

Fuel cycle including minor actinides separation process is going to be designed as follows; spent fuels from both LWRs and LMFBRs will be reprocessed by the advanced PUREX process, in which minor actinides can be separated from other elements. MOX fuels including minor actinides will be loaded to LMFBR and minor actinides can be reduced in the core.

c) Recovery of noble metals from insoluble residue:

Recovery tests have been conducted by Pb extraction process to separate noble metals from actual insoluble residue produced by dissolution of spent fuel. It was confirmed that noble metals, such as Ru, Pd and Rh, can be separated efficiently by this process. Mutual separation of noble metals will be conducted.

B.2 Dry Process

a) Separation by fluoride volatilization process:

Vapor pressure measurements of fluorides and basic separation tests are in progress as a feasibility study of fluoride volatilization process.

b) Ultra-high temperature separation:

Cs and Sr can be separated by heating HLW at extremely high temperatures(1800°C-2500°C). Basic tests have been conducted using a simulated HLW. It was shown that Cs can be easily separated and the residue is stable and compact.

C. Studies at CRIEPI

a) Pyrometallurgical process[14]:

Separation of TRU from HLW by pyrometallurgical processing and transmutation in a commercial metallic fuel FBR. In this scheme, TRU produced in LWR are confined in FBR cycle. This process consists of the following four steps. Both denitration and chlorination steps are pre-processing for partitioning.

(1) Denitration of HLW by microwave heating: This method is preferable for converting HLW to oxide because of the small amount of secondary wastes and the safety in operation. Technical feasibility of this method was examined experimentally. Based on the tests, a design study was carried out for the denitration step.

(2) Chlorination of the oxides to chlorides: Chlorine gas together with carbon was used for chlorination of the oxide, where carbon serves as reductant for chlorination. Chlorine gas can be recycled. Nearly 100% chlorination of HLW oxide was demonstrated.

(3) Reductive extraction to separate TRU from molten chlorides using liquid Cd-Li: TRU with some amount of lanthanides are separated from molten chlorides by reductive extraction using liquid Cd-Li. Li serves as the reducing agent and Cd as the solvent for recovery of reduced metals. In order to evaluate the purity of TRU recovered in the reductive extraction step, it is necessary to obtain the distribution coefficients between salt and Cd phases, and related thermodynamic data, especially activity coefficients for TRU and lanthanides. The distribution coefficients, defined as a mole fraction in Cd phase divided by mole fraction in salt phase, are being measured for lanthanides such as La, Ce, Pr, Nd and Y [15], and actinides, U and Np, by irradiating U using Kyoto University Research Reactor.

(4) Electrorefining of recovered TRU: The electrorefining with liquid Cd anode from the preceding reductive extraction step, solid cathode and molten salt electrolyte (KCl-LiCl eutectic salt) has been applied. The anode dissolution and cathode deposition study using actinide and lanthanide mixture will start from the end of this year in the TRUMP-S Program [16]. The development of process technology including the reductive extraction and electrorefining steps are to be started in 1992.

b) Measurement of activity coefficient:

The activity coefficients and free energy for formation of chlorides in salt and activity coefficients of metals in Cd have been obtained by EMF measurements for La, Ce, Pr, Nd, Sm, Eu, Gd and Y and for U, Np, Pu and Am by the cooperation with Rockwell International Corp., University of Missouri-Columbia, US DOE and Kawasaki Heavy Industry under the TRUMP-S Program started in 1989 [16,17]. The data can afford to make the modeling processes for separating lanthanides and actinides.

A series of the experiments starting 1992 is planning to develop the process technology through denitration step to electrorefining step with the simulated waste. This separation technology will be applied not only for HLW but also for solvent scrubbing liquid waste in Purex process, undissolved residues and hulls after dissolution by nitric acid.

3.3 TRANSMUTATION WITH FISSION REACTORS

Optimization of minor actinide recycling into MOX-FBR is studied at PNC. Studies of minor actinide recycling into metallic fuel FBR are conducted at CRIEPI. Actinide burner reactor design study and the integral experiments to improve cross section data of minor actinides are carried out at JAERI.

3.3.1. MOX-FBR and metallic fuel FBR

A. Studies on transmutation in MOX-LMFBRs at PNC

a) Characteristics of minor actinides transmutation [18]:

Parameter survey calculations have been performed to investigate basic characteristics of minor actinides transmutation in a 1,000MWe LMFBFR. The homogeneous loading of minor actinides has no serious penalties to the reactor core performance, provided that minor actinides in

fuel is less than 5wt%. The minor actinides transmutation rate is about 11% per cycle. The amount of minor actinides transmuted is about six times of that of minor actinides generated in a 1,000MWe LWR. Minor actinide loading of 5wt% reduces the burnup reactivity loss by about 50%.

b) Minor actinides material balance:

To estimate the effect of transmutation in LMFBFRs, the total material balance of minor actinides in Japan was calculated. The nuclear generating capacity was estimated based on the Long-Term Program for Development and Utilization of Nuclear Energy in Japan (1987) and the introduction of the LMFBFR on a commercial scale was supposed to start from 2020. This study implies that minor actinides transmutation in LMFBFRs suppresses the quantities of stored minor actinides after reprocessing.

c) Irradiation test in JOYO and development of fuel:

To evaluate neutron cross section data, small amounts of minor actinides will be irradiated in the experimental FBR "JOYO". Irradiation tests of fuel pins containing a small amount of Am-241 will be conducted. The followings are the items for future studies: fuel fabrication technology, physical and mechanical properties of minor actinides loaded fuel, and additional irradiation tests.

B. Studies on transmutation in metallic fuel FBR at CRIEPI

Transmutation of TRU recovered by pyrometallurgical process is studied in the following areas of FBR with metallic fuel.

a) Calculation of transmutation rate and design study of fuel element and core:

CITATION-TRU code was developed by modifying the burnup calculation code CITATION. When 5, 10 or 15 wt% of minor actinides are mixed with FBR fuel, transmutation rate per year is 11 to 12 wt% in oxide fuel, and 13 to 14 wt% in metal fuel. One unit of 1000MWe FBR with metallic fuel which contains 5 wt% of minor actinides can transmute minor actinides from 5 units of 1000MWe LWR [19].

Based on the chemical analysis of minor actinides oxide samples irradiated in KNK-2 fast reactor, the bench mark calculations for transmutation rate are being carried out in cooperation with KIK, ITU and CRIEPI.

b) Characterization study of U-Pu-Zr alloy with minor actinide and irradiation study:

In order to evaluate the characteristics of metal fuel with minor actinides, the following items are being measured in the cooperation with ITU [20]:

- mutual miscibility of actinides,
- allowable content of minor actinides and lanthanides in fuel,
- other fuel properties

Fabrication and irradiation studies of metallic fuel with minor actinide are in preparation. Studies have been proceeding in cooperation with ITU.

3.3.2. Actinide Burner Reactor studies at JAERI

a) Actinide Burner Reactor (ABR) design study [21,22]:

ABR design study was carried out to obtain a technically feasible ABR model. An ABR is a fast reactor designed to burn minor actinides (MA) efficiently. Its major component of fuel is minor

actinides and neutron energy spectrum is very hard to fission MA nuclides which have fission thresholds at neutron energy of about 700keV. Two types of ABR design were obtained.

Na cooled MA metallic fuel ABR(M-ABR): The advantages of metallic fuel are the hard neutron spectrum and the compact fuel cycle facilities when pyrochemical reprocessing is used.

He cooled MA nitride particle fuel ABR(P-ABR): A particle fuel reactor has very high power density since heat removal in a core is very efficient because of a large heat transfer surface per volume of particle fuel. Coated fuel particles are directly cooled with helium. A particle fuel consists of actinide nitride microspheres and thin TiN coating. When fuel operating temperature is lower than one thirds of their melting points, the fuel would perform as a so-called "cold fuel" where the atomic migration is suppressed to result in a low swelling and a little fission-gas release.

b) Study on ABR fuel and fuel cycle[23]:

The thermophysical data base of MA metals is insufficient. Phase diagram and thermodynamic studies of MA alloys are started in cooperation with ORNL. Fabricability and performance particularly during transients of particle fuel are the areas of future study. Forced cooling of fuel, even of metal ingots, is necessary because of large alpha decay heat. Applicability of pyrochemical processes to the nitride particle fuel has yet to be studied. With a suitable chlorinating agent such as $ZnCl_2$, actinide nitrides could be selectively incorporated into a fused chloride, while TiN coating fragment is left unreacted.

c) Integral experiments for minor actinide cross section evaluation [24 25]:

The integral experiment was carried out in the fast critical facility (FCA) of JAERI to evaluate and modify minor actinide cross sections. Fission rate ratios and small sample reactivity worths of separated isotope samples were measured in seven cores where neutron energy spectra were systematically shifted. Using the measured data, JENDL-2 cross section data were adjusted.

3.4 TRANSMUTATION WITH ACCELERATORS

A transmutation system driven by a proton accelerator is studied and the development of an intense proton accelerator is under way at JAERI. At PNC, accelerator assisted or fusion driven systems for Sr-90 and Cs-137 transmutation are evaluated and an intense electron accelerator is under development.

A. Studies of a transmutation system driven by an intense proton accelerator at JAERI

a) Development of simulation codes for a spallation process[26]:

The simulation code NMTC of ORNL for high energy nuclear reaction processes in the energy range from 15 MeV to 3 GeV was modified as NMTC/JAERI to calculate high energy fission reaction above 15MeV and spallation reaction of minor actinides. The simulation code 'NUCLEUS' for spallation reaction of one nucleus was developed to evaluate a computational model for spallation reaction and analyze the data measured in thin foil experiments. These codes will be improved to include pre-equilibrium neutron emission and fragmentation process in spallation reaction. The intercomparison of the routines for high energy fission calculation of NMTC/JAERI and HETC/KFA is planned. The code SPCHAIN has been developed for calculating generation and decay of spallation products. To analyze nuclear reactions in the entire energy region lower than 3 GeV, two code systems were developed by combining NMTC/JAERI with the neutron transport codes, one with MORSE-DD and the other with TWOTRAN-II.

b) Experiments for spallation in a heavy metal bulk target[27]:

The integral experiment with a lead bulky target and 500 MeV protons from the booster facility at the Institute of High Energy Physics, KEK, was started in 1990 to evaluate the reliability of the code NMTC/JAERI. The neutron energy spectrum, spatial distribution and the yields of spallation products were measured. In future, the experiments using a tungsten and a depleted uranium targets are planned.

c) Design studies of minor actinide transmutation system[28]:

The conceptual design study for an accelerator driven transmutation system has been performed. The conceptual design study of a system of chloride molten salt core has been started to transmute minor actinides and long-lived fission products.

d) Development of an intense proton accelerator[29]:

The construction of Engineering Test Accelerator (ETA) with a proton energy of 1.5 GeV and a current of 10 mA is planned. Various engineering tests will be performed using this accelerator for an accelerator-driven transmutation system. ETA is a significantly large system. In particular, an average proton current of 10 mA is 10 -50 times larger than that of existing accelerators used for nuclear physics experiments. To build ETA, therefore, considerable efforts and resources are required.

As a first step, Basic Technology Accelerator (BTA) with a proton energy of 10 MeV and a current of 10 mA is to be built to study a low energy portion of ETA. A study of high energy portion of the accelerator (high β structure) is also planned. As a hot model test, a RF power source will be built and the electric magnetic characteristics of ETA will be measured.

Various computer design codes are to be checked for their reliability. The conceptual design study and various optimization (trade-off) study for the performance parameters of ETA is being carried out.

B. Studies of transmutation by use of accelerators at PNC

a) Assessment study of accelerator assisted system [30]:

The transmutation rates and energy balance of Cs-137 transmutation by accelerator assisted systems have been calculated by using Monte Carlo codes. It seems difficult to achieve required transmutation rates and energy balance by such reactions proposed as photoneuclear reaction, spallation reaction, neutron reaction by secondary neutrons in spallation, and $(n,2n)$ reaction by neutron from muon catalyzed fusion. Further efforts will be made to select the most efficient transmutation method, including new methods such as the moving target method and the inertial fusion transmutation method.

b) Development of an intense electron linear accelerator:

A high current electron linear accelerator with an electron energy of 10MeV and a current of 20mA is under development. PNC selected an electron accelerator for development of an intense accelerator because they are widely used in the industries and the basic technology that will be obtained during the development is applicable to any type of accelerators. With this energy and current, the accelerator under development is fairly larger than existing electron linear accelerators. A basic design study of the accelerator has been completed and trial fabrication of main components such as an accelerating tube and a klystron is in progress. Reflecting the results obtained during the development of these components, the design will be modified. The first operation is scheduled in 1995.

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STATUS OF MINOR ACTINIDE TRANSMUTATION STUDY AT CRIEPI

A. SASAHARA, M. KURATA

Central Research Institute of Electric Power Industry,
Tokyo, Japan

Abstract

The study of the partitioning and transmutation of minor-actinides (MA) are of great value on future nuclear fuel cycle. We have been studying neutronic characteristics of MA transmutation in a metallic fuel FBR and material characteristics of MA contained fuel. In the present study, we analyzed 1) the reactivity temperature coefficient and the reactivity feedback behavior under the anticipated transient without scram (ATWS) with simple asymptotic method, and we measured 2) the properties of MA contained fuel such as phase stability under the cooperation of CRIEPI and JRC-Karlsruhe.

1. INTRODUCTION

In the institute, we are studying a partitioning of MA by a dry processing method from high level waste (HLW) and a transmutation of MA by a metallic fuel FBR¹⁾. A metallic fuel FBR has some advantages as; 1) Neutron energy spectrum is harder than that in a MOX fuel FBR. Therefore, transmutation rates of MA are higher than that in a MOX fuel FBR. 2) Studies at Argonne National Laboratory and others on the dry processing method (i.e. pyrometallurgical partitioning) of alloy fuel suggest the simplicity and the compactness of the method and the dry processing is suitable for a metallic fuel.

Metallic fuel has several advantages as previously stated, but there is a disadvantage that the dry processing has low separation efficiency between RARE-EARTH (RE) and MA. Thus, some amount of RE are loaded with MA in a core simultaneously. When 90% of RE are removed in the partitioning process, it is predicted that the same amount of RE as that of MA will be partitioned from HLW. From the previous study for MA recycling in the metallic fuel FBR, MA transmutation rate was 14-15%/year²⁾ even if RE are contained in the metallic fuel.

For the fuel contained MA with RE, it is has to be analyzed the core safety parameters (reactivity temperature coefficient) and to be examined fuel characteristics.

In the present study, we analyzed 1) the reactivity temperature coefficient and the reactivity feedback behavior under the anticipated transient without scram (ATWS) events with simple asymptotic method, and we measured 2) the properties of fuel containing MA and RE such as phase stability under the cooperation of CRIEPI and JRC-Karlsruhe.

2. NEUTRONIC SAFETY CALCULATION

2.1. CALCULATION GEOMETRY

A reactor type is the 1000MWe metallic fuel FBR core designed in CRIEPI³⁾. Figure 2.1 shows the core geometry design. The core is composed of two homogeneous regions: Inner and outer core region. Refuel interval is 1 year and a fuel is loaded for 3 years. MA and RE were homogeneously loaded in inner and outer core regions. Reference designed fuel was U-Pu-Zr ternary alloy which contains 10% of Zr. We considered to exchange a part of U and Pu in the ternary alloy for MA and RE. Loading ratio of MA and RE was 2% and 5% of heavy metal (excluding Zr) in loading weight.

2.2. COMPOSITION OF MA AND RE

The composition of MA and RE is that in equilibrium recycle. Composition of MA and RE in feed stream is coming from high level waste of LWR spent fuel. We considered actinides as U-234 to Cm-246 and RE as Pr-141 to Eu-155 (16 nuclides) which affect on reactivity of the core.

2.3. CALCULATION METHOD

Nuclear library is ENDF/B-V for MA nuclear data and JENDL-2 for RE nuclear data. Delayed neutron data and group parameters for fuel are given by reference 4.

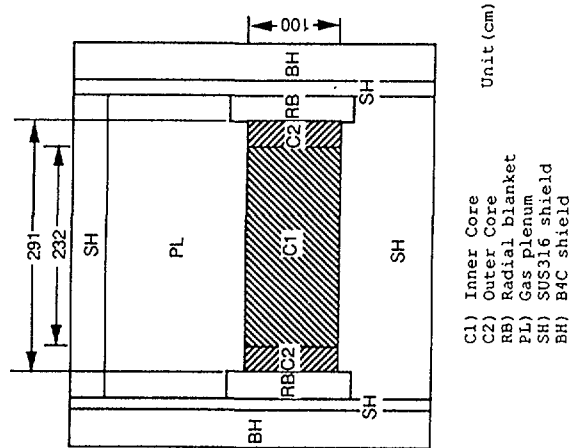


Fig.2.1 1000MWe Metallic Fuel FBR Design

We calculated the reactivity temperature coefficient taking account of the core structure material thermal expansion and nuclides density change based on perturbation calculation.

2.4. ANALYSIS CASE

In this study, we analyzed the following cases,
 1) Reference case : Loading fuel in metallic fuel FBR core is mainly composed of U and Pu. However self generated MA and RE are also loaded. In this case, loading ratios of MA and RE are 0.9% and 1.35% respectively and RE separation efficiency in discharged fuel is assumed to be 67%.

2) 2% MA with RE case : Each of MA and RE loading ratio is 2% of heavy metal respectively. Loading fuel is composed of MA, RE and Pu partitioned from HLM of LWR and from discharged fuel of metallic fuel FBR, and depleted U from the enrichment plant.

3) 5% MA with RE case : Similarly in above case, each of MA and RE loading ratio is 5% of heavy metal respectively.

2.5. REACTIVITY TEMPERATURE COEFFICIENT

The reactivity temperature coefficients are given in table. The absolute value of fuel (without doppler constant), sodium, cladding, duct and coolant temperature coefficient increase as an increase of MA and RE loading ratio because of the neutron energy spectrum hardening and material density decrease. The absolute value of the temperature coefficients for geometry thermal expansion increases by hardening of neutron energy spectrum enhancing neutron leakage.

The absolute value of the doppler constant and the delayed neutron fraction decrease as an increase of MA and RE loading ratio. Decrease of the doppler constants are mainly due to the decrease of U-238 loading

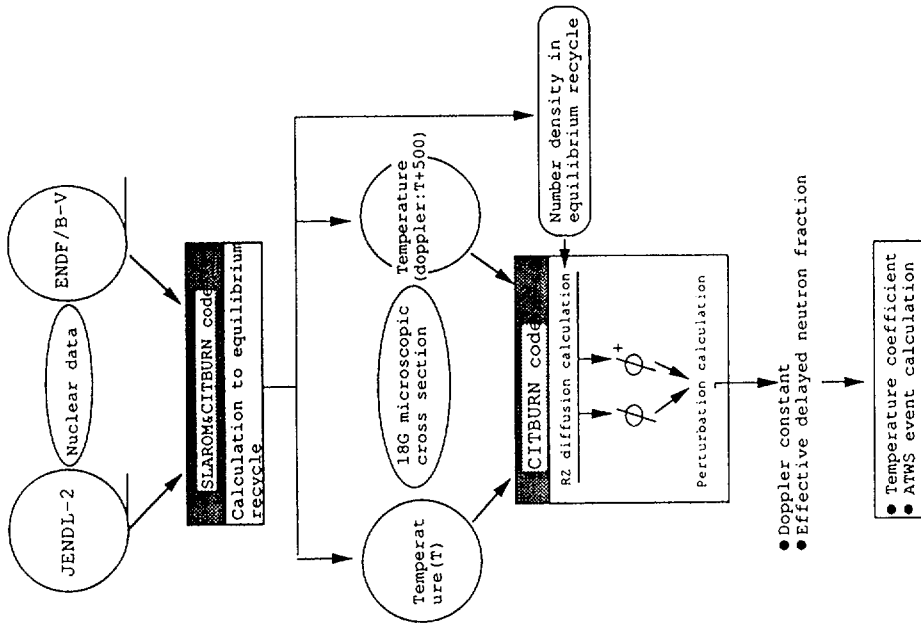


Fig.2.2 Flow Diagram of Calculation

The perturbation calculation was executed with CITURN code. In CITURN code based on CITATION code, the burnup, input and output module are developed in CRIEPI. Fig.2.2 shows calculation flow. Input data are the nuclide composition at equilibrium recycle and average effective microscopic cross sections of each core region. The average effective microscopic cross sections have 18 energy groups structure and are values in two different temperature conditions : One is at normal operating temperature condition and the other is at 500 degree higher than the normal operating temperature for perturbation calculation.

TABLE RESULTS OF TEMPERATURE COEFFICIENT CALCULATION

Temperature coefficient(ε /C)	Reference	MA2%,RE2%	MA5%,RE5%
Fuel*	-0.086	-0.09(4.7%)	-0.096(11.6%)
Sodium bond	0.030	0.032(6.7%)	0.038(26.7%)
Cladding	0.044	0.047(6.8%)	0.055(25.0%)
Duct	0.016	0.016(0%)	0.019(18.8%)
Coolant	0.264	0.282(6.8%)	0.339(28.4%)
Radial expansion	-0.5	-0.52(4.0%)	-0.565(13%)
Doppler constant (TdK/DT)	-4.94E-3	-4.48E-3(-9.3%)	-3.23E-3(-34.6%)
Delayed neutron	3.35E-3	3.29E-3(-1.8%)	3.10E-3(-7.5%)

* This value does not contain doppler effect

amount, because U-238 has large resonance capture cross sections between 10eV and 3keV. MA nuclides have smaller delayed neutron fraction values than that of U-238. Therefore, increase of MA and RE loading ratio caused decrease of core effective delayed neutron fraction.

2.6. REACTIVITY FEEDBACK UNDER ATWS

We analyzed anticipated transient without scram (ATWS) events such as unprotected loss of flow (ULOF) and unprotected loss of heat sink (ULOHS) and unprotected transient over power (UTOP) by simple asymptotic method⁵⁾.

Fig.2-3-2.5 shows the simple estimated outlet temperature under ATWS events. The figure shows that the trend of the outlet temperature rises as the increase of loading amount of MA with RE.

3. PROPERTIES FOR U-PU-ZR ALLOY FUEL CONTAINING MA AND RE

3.1. ITEMS OF MEASUREMENT

U-Pu-Zr based alloy is one of the candidate fuels for transmutation, not only because the high efficient transmutation is attained in the metallic fuel FBR, but also because the chemical and physical properties of U-Pu-Zr-MA-RE could not be so different from those of U-Pu-Zr ternary alloy.

The properties of U-Pu-Zr-MA-RE, such as phase stability, thermal conductivity, compatibility with cladding and so on, are measuring in the cooperation study of CRIEPI and JRC-Karlsruhe. And those of U-Pu-Zr are also measured as the reference.

Concerning the study for phase stability, specimens of U-Pu-Zr, containing 2% of MA and 2% of RE and those containing 5% of MA and 5% of RE, respectively, were annealed at various temperatures for about 50 hours and quenched to room temperature. These samples were analyzed by alpha-autoradiography, metallography and electromicroprobe.

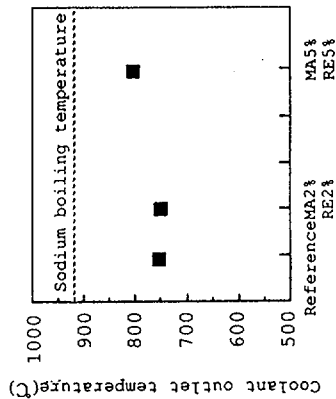


Fig.2.3 Asymptotic Outlet Temperature by Quasi-Static Method under ULOF event

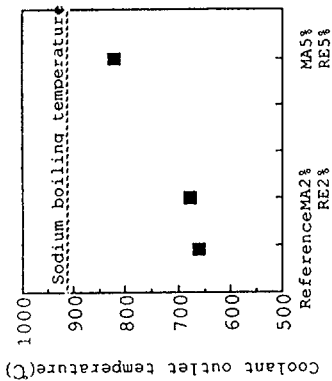


Fig.2.4 Asymptotic Outlet Temperature by Quasi-Static Method under ULOHS event

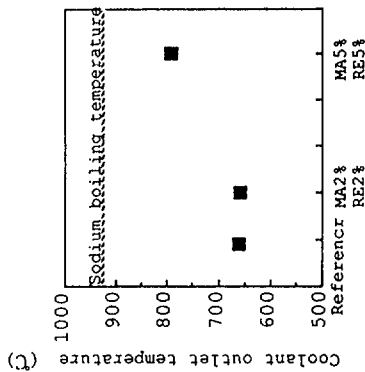


Fig.2.5 Asymptotic Outlet Temperature by Quasi-Static Method under UTOP event

3.2. ALPHA-AUTORADIOGRAPH

Fig.3.1 and Fig.3.2 show the alpha-autoradiographs of 2%MA-2%RE alloy and 5%MA-5%RE alloy after the heat treatment. In these samples, major alpha activity emitter is americium, therefore, small white spots are correspond to the positions of americium. Americium is homogeneously dispersed in 2%MA-2%RE alloy. In 5%MA-5%RE alloy, however, larger spots are observed. According to EPMA, these spots consist of 30%Am-10%Pu-60%RE (wt%).

3.3. METALLOGRAPHS

Fig.3.3 shows the microstructure of 2%MA-2%RE alloy annealed at about 500, 600, 700 and 800 C. The specimens annealed at temperature about 500 and 600 C show three phases. According to EPMA, these are a U-rich phase, a

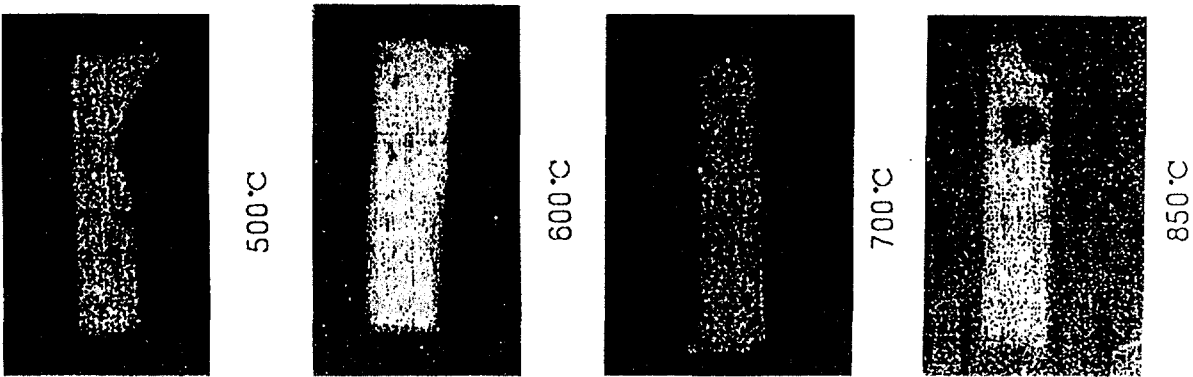
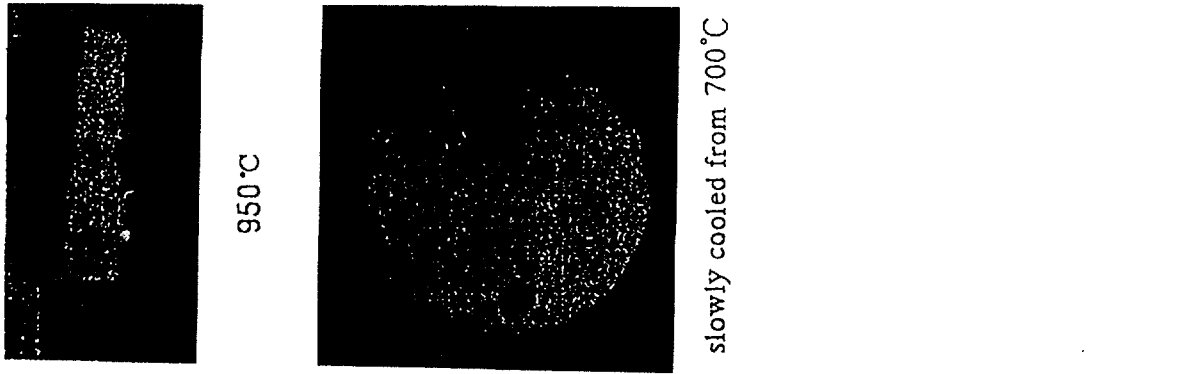
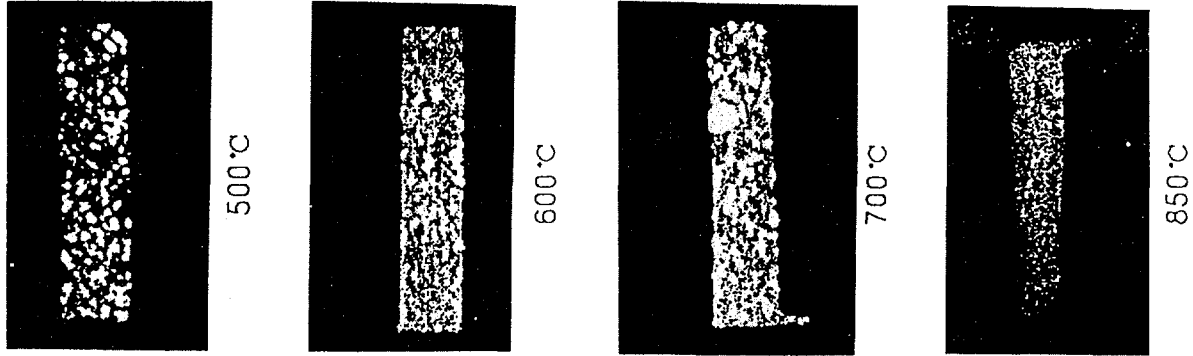
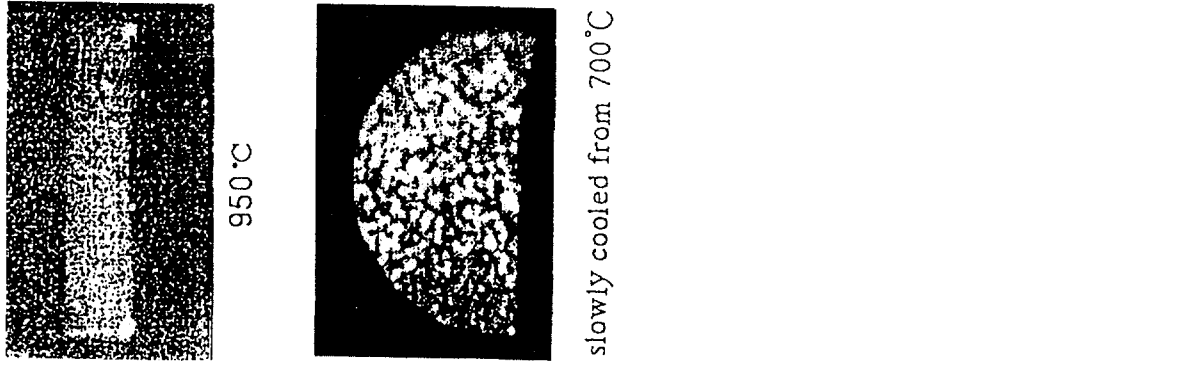


Fig. 3.2 alpha - autoradiograph of U-Pu-Zr based specimens, containing 5% of MA and 5% of RE, which were quenched from various temperature ($\times 20$)

Fig. 3.1 alpha - autoradiograph of U-Pu-Zr based specimens, containing 2% of MA and 2% of RE, which were quenched from various temperature ($\times 20$)

Zr-rich phase and an americium-rare earth phase. The U-rich and the Zr-rich phase correspond to zeta and delta phase in the U-Pu-Zr ternary system. Above 700 C, the U-rich and the Zr-rich phase are unified to one phase and it corresponds to gamma phase in the ternary system.

3.4. OTHER PROPERTIES

Thermal conductivity, density, mechanical properties, solidus-liquidus temperature and another properties of U-Pu-Zr and U-Pu-Zr-MA-RE are measuring. However, no significant differences are observed, because almost all of MA and RE are consisted as the inclusions and these inclusions do not seriously affect the various properties of the alloy. Therefore, U-Pu-Zr-MA-RE type alloy can be selected as the fuel for transmutation.

4. SUMMARY AND CONCLUSION

We analyzed a) the reactivity temperature coefficient with CITBURN code and the reactivity feedback behavior under the anticipated transient without scram (ATWS) events with simple asymptotic method, and measured b) the properties of fuel containing MA and RE such as phase stability under the cooperation of CRIEPI and JRC-Karlsruhe.

In the present study, we obtained following results,
 (1) The absolute value of fuel (without doppler constant), sodium, cladding, duct and coolant temperature coefficient increase as an increase of MA and RE loading ratio by the effect of the neutron energy spectrum hardening. The absolute value of the temperature coefficients for geometric thermal expansion increases by hardening of neutron energy spectrum enhancing neutron leakage.

(2) In the case of 5% loading ratio of MA and RE, simple estimated coolant outlet temperature increases near to the coolant boiling temperature during ATWS events. It should be noticed that present ATWS event analyses are in very simple asymptotic method and a metallic fuel properties such as fuel thermal conductivity are not clear now. Therefore, these analyses show only trend of change caused by MA and RE loading. However, these ATWS events analyses indicate the necessity of core design considering ATWS detailed analysis. From all results, the results of the simple asymptotic analysis in ATWS events show that the metallic fuel FBR has safety potential even if MA and RE are loaded. It is important that the reactivity feedback of thermal expansion increases slightly with MA and RE loading, but doppler constant and other parameters decrease with MA and RE loading.

(3) By the characterization study of the fuel containing MA and RE, americium and RE inclusions were detected in the grainboundaries. However, there are no significant difference between U-Pu-Zr-MA-RE and U-Pu-Zr alloy on thermal conductivity, density, mechanical properties, solidus-liquidus temperature and other properties.

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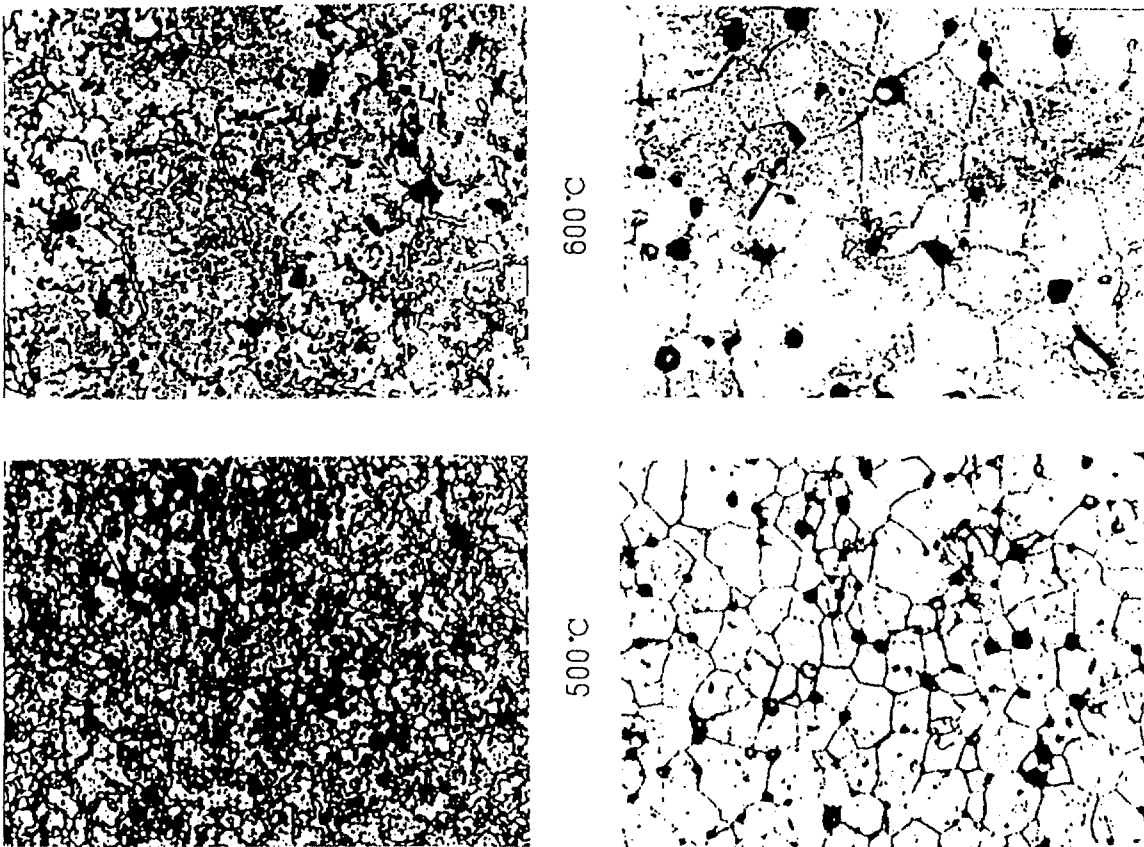


Fig. 3.3 micrograph of U-Pu-Zr based specimens, containing 2% of MA and 2% of RE, which were quenched from various temperature (after etching. X1000)

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STATUS OF WORK ON TRANSMUTATION IN SWITZERLAND

(Summary)

G. LEDERGERBER, P. WYDLER, R. W. STRATTON
Paul Scherrer Institute,
Villigen, Switzerland

Studies on the transmutation of long lived radionuclides are concentrated at the Paul Scherrer Institute (PSI) in the department "Nuclear Energy". Merging out of existing activities on Nuclear Fuel and Reactor Physics two general subjects have been identified for experimental work.

- PREPARATION of TARGET MATERIALS for fast reactor application utilizing wet chemistry processes for the direct conversion of nitrate solutions into a fuel form (dense spheres) or in an intermediate product to be pelletized (porous spheres).

The aim is to prepare uranium-plutonium-neptunium nitride and uranium-plutonium nitride microspheres and to compare sphere pac fuel and hybrid sphere-pellet with the dry route pellets in a fast flux reactor. This cooperative work is being shared with the Département d'Etudes des Combustibles at Cadarache of the French Commissariat à l'énergie atomique (CEA) and PSI.

In parallel, an evaluation of possible matrix materials in the form of oxides or nitrides to be prepared by wet chemistry processing has started. The oxide of magnesium and the nitride of zirconium are in the center of interest. Preliminary tests in the zirconium-cerium oxide and the zirconium-uranium nitride systems have been initiated in collaboration with Japan Atomic Energy Research Institute (JAERI).

- NUCLEAR DATA AND CALCULATIONAL METHODS PROBLEMS related to the accelerator-based transmutation of actinides.

High energy nucleon-meson transport computer codes used in the design of accelerator based transmutation systems have to be capable of correctly predicting, among other things, the yield and the mass distribution of spallation and fission products generated in the target. Simple code comparisons for the irradiation of thin samples of actinides have revealed considerable differences in the prediction of these quantities. To resolve the discrepancies and to confirm the high potential of the high-energy fission reactions for the transmutation of actinides, an experimental programme, ATHENA (Actinide Transmutation using High Energy Accelerators), has been initiated. In a first phase of the programme, thin samples of uranium, neptunium and americium encapsulated in aluminium will be irradiated with 590 MeV protons from the PSI ring accelerator using the PIREX irradiation facility. The distribution of the reaction products will be measured using different methods such as the ICP-MS and the total reflection X-ray fluorescence technique and compared with results of model calculations.

For accelerator-based systems in which protons are used directly to transmute actinides, integral information on the adequacy of neutronic design methods is scarce. In a second phase of the ATHENA programme, it is therefore proposed to study neutronic behaviour of such systems with the help of zero-power experiments at a separate beam of the accelerator.

The physics related studies are carried out in the framework of a collaboration with the Département d'Etudes des Réacteurs of the CEA at Cadarache.

The activities make use of existing knowledge and capabilities and utilize an almost unique combination of facilities available at PSI. The two topics, accelerator transmutation and target fabrication, are regarded by the PSI as long-term basic research and help to preserve and further develop competence in the area of advanced fuels and reactor physics.

SCIENTIFIC RESEARCH PROGRAM ON ACTINIDE TRANSMUTATION BY USE OF FAST REACTORS

L.A. KOCHETKOV, A.G. ZCYKUNOV
Institute of Physics and Power Engineering,
Obninsk, Russian Federation

Abstract

For the recent years in the Ministry of Atomic Energy of Russian Federation the national program on RAW management has been under development. The program of actinides transmutation (as the most hazardous part of RAW) was treated as its component. According to this program various investigations within this field have been supported for the last two years. The program includes five sections dealing with actinide transmutation with the use of fast reactors.

1. The problem of radioactive waste (RAW) management has recently become of immense importance in nuclear power of all the countries involved in its solution. Evidently in the nearest future it will be an issue of the main priority in the IAEA activity along with the safeguards and NPP's safety. The nuclear power progress could hardly be anticipated without its adequate solution. It doesn't mean that the authorities and experts absolutely neglect the problem. The time has just come when the attention to this problem is considered insufficient, when the problems of finding the sound solution of RAW management are more obvious. Nowadays we are quite aware of the international aspect of the problem and the scale of the financial support necessary. The nuclear power development as it is makes it necessary to work out:

- certain concepts of RAW management ;
 - national and international standards, regulations and rules;
 - research and development programs;
 - practical recommendations based on scientific and research works data and on the assimilated expertise.
- When working out the concept of RAW management we should proceed from the assumption of at least long-term preservation of the Earth natural radiotoxicity level. If we leave alone low-level

and middle-level RAW, and consider only high level wastes produced mainly (98%) in nuclear reactor cores the problem of their management could be reduced to at least two independent aspects to be solved quite differently:

- isolation of the main part of fission products from the environment for the period of 300-500 years;
- elimination (conversion, spatial ejection) of a number of transuranians, actinides, in particular the Pu-isotopes, and the so-called minor actinides-Np, Am, Cm whose radiological hazard is conserved for million years. The majority of investigators consider a compact multibarrier isolation of fission fragments storage under surveillance to be the most natural and efficient way of solving the first problem.

The most reasonable way of solving the second problem is evidently the fission fragments utilization by means of fast reactors. The advantage is testified by the following;

- the accumulated positive experience of the design and operation of fast reactors (fast reactors technology is a present day reality);
- specific physical features of fast reactors which can really utilize (burn or transmute) not only Pu-isotopes but all the minor actinides without any additional cost or energy consumption for their transmutation;
- their transmutation in fast reactor shows the best ecological effect.

For the recent years in the Ministry of Atomic Energy of Russian Federation the national program on RAW management has been under development. The program of actinides transmutation (as the most hazardous part of RAW) is treated as its component. According to this program various investigations within this field have been financially supported for the last two years. The program includes the following five sections dealing with actinide transmutation with the use of fast reactors

- physical and technical grounds, ecological and safety problems;
- constants, calculational and theoretical justification;

- conventional fast reactor core design meant for actinides transmutation (Pu, Am, Np, Cm);
- design of a specialized fast (fast-thermal) reactor, meant for transmutation of actinides and probably the most hazardous fission fragments;
- associated problems, such as
 - * radio-chemical
 - * fuel fabrication technology
 - * transport
 - * biophysical
 - * feasibility

In addition the logistics are envisaged for

- physical studies;
- technological investigations;
- development of core designs.

Section One (190-1994) stipulates the definition of requirements, elucidation of restrictions, conceiving the principles, concept and engineering proposals. The determination of requirements to the depth of most hazardous nuclides extracts from spent fuel seems essential.

Section Two (1990-1994) nuclides calculational and experimental investigations aimed for the provision of adequate accuracy of physical characteristics responsible for safety and economy. The fragments yield, delayed neutrons characteristics, nuclear reaction constants at a microlevel, group constants are to be refined. Concurrently calculational codes are to be up-dated.

Section Three (1992-2000) stipulates the elaboration of engineering design for the BN-800 (BN-600) core, with minor actinides doped into the its conventional fuel in the amount that does not noticeably alter the physical characteristics of the BN-800 core developed nowadays. Traditionally in this case the detailed parametric investigations of physical and thermophysical characteristics should be performed, the optimum solutions on the core composition arrangement and handling of fuel and absorbing materials in it should be found, that will provide for the required burn-up, stability of power density self-regulation.

Section Four (1992-2005) stipulates the development of an absolutely new core with an increased content of actinides and free from U-238.

These developments should be based on technological achievements that would form the ground for the next fifth section. The need for Section Four may emerge in the case of more intensive actinide burning occurrence.

4. The current status of the program features the following studies:

- in the field of constants;
- conventional BN-800-type cores with an addition of minor actinides;
- on irradiation of minor actinides oxide samples in the BN-350 reactor.

Preparation is being conducted for experimental fuel elements exposure in the BOR-60 reactor. Investigations are being held to support the concept of management of long-lived (most hazardous) components of nuclear wastes.

Fast neutron reactors may be considered to be able to attain a supplementary vital incentive for confirmation and development, for they will be able to solve the problem of management of the most long-lived and hazardous radwastes of nuclear power in the most rational way. In this context the joint efforts of the IAEA member countries and primarily of those committed to fast breeder reactor development aimed for the solution of the actinides transmutation problem appears desirable and beneficial.

Summary

The program on actinides transmutation with the use of fast reactors has been elaborated. It covers the period of 1990-2005 and is financed by the Ministry of Atomic Energy of Russian Federation. The program comprises the following five R&D aspects:

- physical and engineering grounds, ecological and safety problems;
- constants, calculational codes;

- design of conventional fast reactor cores, meant for actinides burning (transmutation);
- design of a special fast reactor with a high amount of actinides;
- associated problems (radio-chemical, fuel production, transport).

PHYSICS ASPECTS OF TRANSMUTATION OF MINOR ACTINIDES
AND LONG LIVED FISSION PRODUCTS IN FBRs

(Session 2)

Chairmen

L.A. KOCHETKOV
Russian Federation

L. KOCH
Commission of the European Communities

REACTOR ASPECTS OF ELECTRONUCLEAR TRANSMUTATION OF ACTINIDES IN THE HEAVY WATER HIGH FLUX BLANKETS. ANALYSIS OF PROBLEMS

E.V. GAI, A.V. IGNATYUK, N.S. RABOTNOV,
A.A. SEREGIN, Yu.N. SHUBIN
Institute of Physics and Power Engineering,
Obninsk, Russian Federation

Abstract

The following problems in reactor and neutron physics of high flux heavy water accelerator driven blankets are considered and shown to be very serious at the fluxes of 10^{16} n/cm²: the difficulty of attaining needed neutron flux; high radioactivity and power release in the primary circuit; tritium accumulation; heavy water radiolysis and hydrogen release; positive flux-reactivity feedback; poisoning. At more moderate fluxes fission rates of the minor actinides become unsatisfactory which leads to the accumulation of heavier hazardous long-lived isotopes. The desirability of investigation of static liquid blankets which may solve some of those problems is pointed out.

1. INTRODUCTION

Existing transmutation projects may be split into following main categories:

- high flux reactors with thermal or slightly more hard (epithermal, resonance) spectra;
- fast reactors (power plants or specialized actinide burners);
- subcritical assemblies and blankets both thermal and fast driven by high current accelerator neutron sources.

The interest to transmutation potential of electronuclear plants was growing fast in the last few years. During the last fifteen months at least three international scientific meetings on transmutation were held: in Obninsk (Russia), July 1991; in Vienna (IAEA, Austria), October, 1991; in Villigen, Switzerland, March, 1992. The last meeting was devoted completely to electronuclear transmutation. The papers presented to the meetings are the main part of the information base for present report. The complete comparative analyses of reactor and electronuclear options should, in principle, be done along two

lines - different schemes are to be compared between themselves and to alternative reactor schemes. But it's too wide a task and we shall narrow it to the limit and consider only what may be called polar opposite to fast breeder transmutation - heavy water super high flux electronuclear blanket burners. Other reports to present meeting will, no doubt, provide ample opportunity for comparison.

Although accelerator driven neutron multiplying transmutation devices are formally called targets or blankets due to their subcriticality, in the case of large power producing industrial systems they would be reactors all right with some of the problems characteristic for standard reactors and some new ones not less real and serious. First proposals on neutron transmutation were based on reactor options (see [1]). Minor actinides (MA) are in fact threshold fission elements. At neutron energies above 1 MeV capture cross sections are only a few per cent of fission cross sections, so fast breeder reactors (FBR) were the first to be considered. But it was immediately understood that averaging over real fast reactor neutron spectrum results in fission to capture ratios 10-20 times larger than for PWR spectrum but still considerably less than unity, closer to 0.2. Np and Am are relatively poor fuel for fast breeders and any considerable hardening of the spectrum is welcome. So specialized fast breeders-actinide burners (FBAB) were natural option to propose but resulting improvement was not qualitative, about threefold. Hence the interest to the transmutation possibilities of electronuclear facilities.

Four main types of actinide waste may be singled out and ordered by the complexity of the isotope composition:

- a. Military waste left after effective plutonium and uranium extraction from low burn fuels with Np-237 as a main component by a large margin.
- b. "Minor actinides" (MA) waste left after radiochemical reprocessing of commercial NPP spent, well burned fuel; Np-237 comprises about a half of this waste, the rest being mainly Am and Cm isotopes.
- c. Commercial spent fuel with only fission products and most of uranium removed by reprocessing; some 90% of it is plutonium, MA the rest.
- d. Spent fuel proper as a product of open nuclear fuel cycle.

Each of the groups a-c is a potential raw material for actinide transmutation and needs, in principal, individual approach. Key isotope Np-237 is most "stubborn" of all, and the overall effectiveness of actinide transmutation schemes must be measured by its success in burning neptunium.

2. ATW PROJECTS

LANL scientists have proposed what is probably the most radical concept of accelerator driven transmutation [2]. Its basic idea is very appealing. Minor actinides nuclei are effective absorbers of thermal neutrons with low fission probability. But in very high fluxes of thermal neutrons planned in LANL scheme the fission of the isotopes formed after the

capture of neutrons by long-lived MA radionuclides plays the main role (see Table I). Their significant equilibrium concentrations may be reached only if the flux is close to 10^{16} neutrons/cm² (relatively long-lived Am-242m is an exception). Thus the absorbers turn into superfuel.

TABLE I. COMPOSITION AND MAJOR NUCLEAR PARAMETERS OF PWRs MA DISCHARGE (THE CROSS SECTION VALUES TAKEN FROM [3])

Nuclide	Minor actinides			Capture products			
	ρ_1/ρ_{Np}	σ_γ, b	σ_f, b	Nuclide	σ_γ, b	σ_f, b	$T_{1/2}$
²³⁷ Np	1.	0.573	176	²³⁸ Np	203	2027	2.12 d
²⁴¹ Am	0.46	0.264	587	²⁴² mAm(533b)	?	2100	16.1 h
				²⁴² mAm(54b)	1344	6950	152 a
²⁴³ Am	0.224	0.128	75.1	²⁴⁴ Am(3,8b)	?	2300	10.1 h
				²⁴⁴ mAm(71,3b)	?	1600	26 min
²⁴⁴ Cm	0.049	0.028	15.2	²⁴⁵ Cm	369	2145	8500 a
²⁴⁵ Cm	9.4-3	5.4-3	369	²⁴⁶ Cm	1,22	0.14	4700 a
²⁴⁵ Cm	8.1-4	4.6-4	130	²⁴⁴ Cm	15.2	1.04	18.1 a
²⁴² Cm	3.1-4	1.8-4	16	²⁴³ Cm	130	617	28.5 a
²⁴⁶ Cm	2.4-4	1.4-4	1.22	²⁴⁷ Cm	57	81	1.6+7 a

The scheme is oriented on the use of liquid fuel-coolant circulating between blanket and heat exchanger, part of it going to a radiochemical bypass for continuous reprocessing. The scheme allows to avoid two major difficulties: remote controlled production of fuel pins of highly radioactive materials and additional high level waste from construction materials of the fuel assemblies.

Prospects of the method are estimated optimistically and the research activity is considerable. In later versions [4-7] the evolving concept was in fact split roughly according to selected target waste group. The common feature of all schemes besides a 1-1.6 GeV, \approx 100 mA linac is a heavy water blanket containing solutions or slurries of actinides and/or technetium compounds. High fluxes and cross sections allow in principle to use very low concentrations of the nuclides to be transmuted still getting good burning rates. Heavy water fuel-moderator-coolant (FMC) circulates in the primary circuit of most probably three-circuit unit. This general concept was accepted as most promising one by the majority of electronuclear experts in our country too, the preferred versions and estimates being close to American ones [8-10]. All the authors point out general advantages of liquid fuel schemes: the absence of nonuniform power density and of radiation endurance problems characteristic for fuel pin systems and the possibility to adjust the composition of the core continuously through a chemical bypass without frequent reloading of fast burning solid fuel.

Considerably less attention was paid so far to very serious problems the analogs of which plagued, for example, the molten salt reactors and prevented their practical use in spite of considerable long time efforts of ORNL where two experimental reactors were built and operated for a few years each. The problems are: extremely high radioactivity and decay heat in the primary circuit; complex, ever varying composition of the circulating liquid; possible sedimentation of radioactive and fissile materials in various parts of the circuit; coolant's radiolysis, especially dangerous in the case of profuse hydrogen production. These difficulties may be qualitatively aggravated in super-high flux transmutation plants. Some new ones arise too. We shall consider them one by one with the emphasis on superfuel case needed to burn Np-237.

3. THE DIFFICULTY OF GETTING HIGH EFFECTIVE FLUX

First of all very high flux is vital for the scheme which is illustrated by Fig.1 and 2. Fig.1 taken from [6] shows crucial neutronics parameter - the number of neutrons required per fission as a function of thermal neutron flux in Np-237 transmuting system. When it crosses a horizontal line indicating the number of neutrons released in fission event absorber turns into fuel. Fig.2 demonstrates that the rate of fission drops

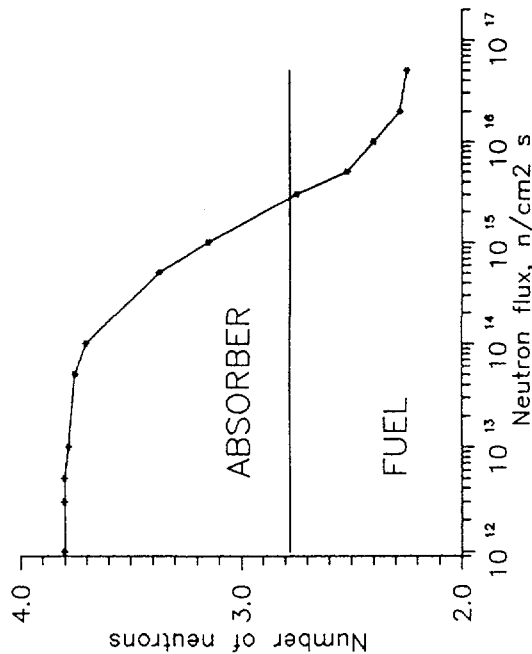


Fig 1. Number of neutrons required to fission Np-237 in a thermal spectrum as a function of flux level [7]. The horizontal line indicates the neutrons released in fission.

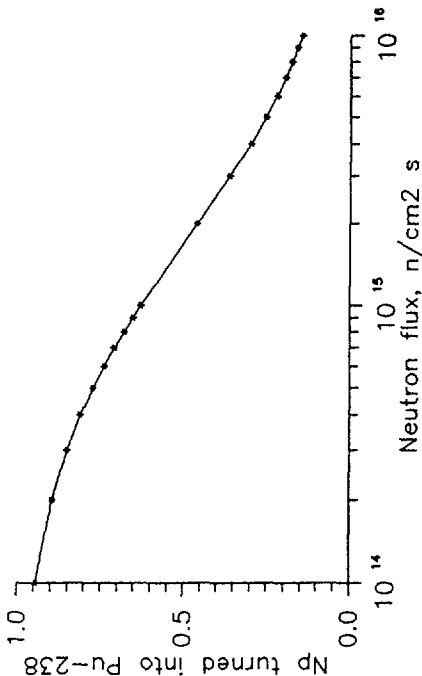


Fig 2. The share of Np-237 turned into Pu-238 instead of fission as a function of neutron flux.

fast with decreasing flux and its place is taken by production of Pu-238 which is hardly less of a radiational hazard than initial Np-237.

One serious difficulty in getting high enough flux is this: FMC spends only part T_1 of the full circulation period in the blanket. The rest of the time T_2 it is pumped through the pipes, heat exchangers and radio chemical units. If both T_1 and T_2 are much less than relevant decay half-time then effective neutron flux ϕ_{eff} enters a simple basic equation determining the equilibrium concentrations of the key short lived well fissioning nuclides

$$\rho_{N+1} = \rho_N \gamma N \phi_{eff} / (\lambda_{N+1} + \sigma_t(N+1)) \phi_{eff} \quad (1)$$

where N is nuclide's neutron number, ρ - density, λ - decay constant. ϕ_{eff} differs from physical neutron flux ϕ

$$\phi_{eff} = \phi \tau; \quad \tau = T_1 / (T_1 + T_2). \quad (2)$$

Operating NPPs have τ values varying from approximately 1/20 (PWR) to 1/300 (BN-800 type FBR). For specially designed blankets [7] $\tau \approx 0.2$ and the effective flux is reduced to almost 2×10^{14} instead of physical 10^{15} . The necessity to keep τ value as close to unity as possible conflicts seriously with thermophysical requirements. FMC heating in the liquid fuel blankets is very effective volume process without separating walls. Cooling is much less effective, slower, especially in multi-circuit systems. τ value may be improved substantially

only by refusing to produce electricity and using much colder water in external circuits. But even in this case τ values much higher than 0.3 are hardly realistic. Impossibility to produce power in ATW type scheme would not only complicate - and considerably - the economic side of the problem but also undermine the principal advantage of subcritical safety if a standard critical NPP supplying the linac with electricity becomes an indispensable part of the scheme. So ATW project is rightly planned as power self-sufficient and even electricity producer.

The spatial nonuniformity of the flux is equivalent to time nonuniformity. With a central axial neutron source the volume with maximum flux is limited and space averaging reduces resulting effective value.

The burning half-lives are crucial parameters of any transmutation plant. They depend on the load factor of the plant which enters denominators. In electronuclear transmutation the total load factor is a product of partial ones - for the linac, target complex, blanket plant and chemical by-pass. It may reduce its value considerably and prolong transmutation cycles acting just like decreasing of the effective flux.

4. THE DIFFICULTIES OF HANDLING HIGH EFFECTIVE FLUX

4.1. Tritium production

Accumulation of tritium would be very serious problem in high flux heavy water blankets. It accumulates due to neutron absorption in deuterium. Thermal cross section of the process is just 0.53 mb but in the flux of 10^{16} neutrons/cm² it results in tritium production rate about 1000 Ci per ton of heavy water a day and equilibrium concentration to be reached in some twenty years is about 10 million Ci/t. Hundreds and even dozens of Curies of chemically unseparable tritium in cooling water are considered as significant problem in PWRs, so it's clear that tritium quantities 5-6 orders higher would create grave difficulties indeed if the fluxes like 10^{16} cm⁻² s⁻¹ in heavy water become reality.

4.2. Heavy water radiolysis and hydrogen release

In the solid fuel reactors first protective barriers - fuel slug material and fuel pin shell - prevent not only accumulation of radioactive fission fragments in the coolant but radiolitic destruction of its molecules by fission fragments as well. The radiolysis of water leads to production of hydrogen and oxygen. The latter reacts with the water forming peroxide. Hydrogen is mainly released because recombination processes are relatively weak in pure water. Approximately one million of H₂ molecules is created by every act of fission in the water [11]. At GWT power level it means the release of a few cubic meters of hydrogen (npt) per second in heavy water blankets [10]. This hydrogen is very radioactive due to tritium component and means high explosion danger. The radiolysis may be inhibited in normal

operating conditions by recombination catalyzers like copper but in case of an accident leading to intense boiling recombination goes down while radiolysis intensity is some ten times higher for the vapor than for the water [11]. Addition of the copper inhibitors (the quantities needed are up to mol/l) absorb neutrons damaging neutron balance already vulnerable at high fluxes. Peroxide is strong oxidizing and corrosion agent. These problems speak for themselves. They are not directly connected with the flux itself and are determined just by total power of fragments released into the water.

The precursors of the delayed neutrons also circulate in the primary circuit (see section 4.4.3 below) so there will be not negligible release of neutrons producing fissions in all sections of the circuit with resulting production of hydrogen. It would be by a few orders of magnitude slower than in the blanket itself but may become a serious complicating factor in heat exchangers and pumps calling for special measures to deal with and seriously affecting safety.

4.3. Radioactivity and heat release in the primary circuit

If a radioactive nuclide is produced during the irradiation of a liquid circulating fuel in the core the share of its nuclei in the core at a stationary irradiation regime may be expressed in terms of decay constant λ and the times spent inside and outside the core T_1 and T_2 [12]

$$N_C/N_{\text{tot}} = 1 - \frac{(1 - e^{-\lambda T_1})(1 - e^{-\lambda T_2})}{T_1 \lambda (1 - e^{-\lambda(T_1 + T_2)})} \quad (3)$$

If all the exponential arguments are much less than unity this ratio turns into $N_C/N_{\text{tot}} = \tau = T_1 / (T_1 + T_2)$. That's the case for most of the fission and capture products so only a part of total radiation and decay heat equal τ is released in the core and the rest in other parts of the primary circuit. Without on line chemical reprocessing this heat and radiation release grows with the time during the campaign reaching typically about 8 per cent of total power. On line cleaning is rather slow and reduces only slightly the short-lived radioactivity which makes a lion's share of the release so the problem survives.

In the heavy water blanket burners there is a considerable addition to this effect due to very high radioactivity of the short-lived well burning actinides Np-238, Am-242 and Am-244. Their periods were indicated in Table I, average beta- and gamma-energies are given in Table II.

The periods of all the nuclides in Table II are more than 10 hours, 51 hours for most important Np-238 so they may affect considerably the cooling after shut down. In general the extremely high levels of the radioactivity of the primary circuit liquid fuel-coolant deserve special detailed discussion. Gamma-heating of the uncooled shielding may become a problem.

TABLE II. AVERAGE β - AND γ -ENERGIES OF THE MINOR ACTINIDES PER ACT OF DECAY

Nuclide	E, keV	E, keV
²³⁸ Np	190	647
²⁴² Am	159	18
²⁴⁴ Am	109	806

4.4. Complications in blanket kinetics

Kinetic problems may seem of minor importance for subcritical facilities but it's only true for the dangers associated with the instabilities of steady critical condition which cause most concern in traditional NPPs. Attaining and sustaining stable neutron multiplication and flux on planned level would be difficult task with heavy water blankets as the examples considered below illustrate.

4.4.1. Strong positive flux-reactivity feed-back

Equilibrium concentrations of the well fissioning nuclides grow with the flux and so does reactivity. This positive feed-back is a potential source of instabilities calling for investigation. Here we shall give only one example estimating and comparing reactivity time derivatives after a stepwise flux increase for two cores: uranium enriched to 3% of U-235 in the flux 10^{15} and neptunium with ¹⁵equilibrium concentrations of Np-238 for the flux values 10^{15} and 10^{16} .

The single most sensitive factor of the four determining k_{eff} (which we shall designate simply k) is η and for present limited purpose we can approximately take $k \approx \eta$. Then

$$\frac{\partial \rho}{\partial t} = \frac{1}{k^2} \frac{\partial k}{\partial t} \approx \frac{1}{\eta^2} \frac{\partial \eta}{\partial t} = \frac{1}{\eta^2} \frac{\partial \eta}{\partial n_f} \frac{\partial n_f}{\partial t} \quad (4)$$

where n_f is the density of fissioning nuclide. Let n_a be the density of the absorbing nuclide. Then

$$\eta = \nu_f \frac{\sigma_{ff} n_f}{\sigma_{tf} n_f + \sigma_a n_a} \quad (5)$$

n_f will start changing linearly with time after small "flux jump" $\Delta \Phi$: $n_f = n_f^0 + \beta t$. $\beta = N_a \sigma_a \Delta \Phi$ for neptunium and $\beta = -N_f \sigma_{tf} \Delta \Phi$ for uranium. Taking that into account we get from (4-5)

$$\frac{\partial \rho}{\partial t} = \frac{n_a}{n_f} \frac{1}{\sigma_a} \frac{\partial \sigma_a}{\partial \Phi} \Delta \Phi \quad \text{and} \quad \frac{\partial \rho}{\partial t} = \frac{n_a}{n_f} \frac{1}{\sigma_{tf}} \frac{\partial \sigma_{tf}}{\partial \Phi} \Delta \Phi \quad (6)$$

TABLE IV. HALF-LIVES AND THERMAL CROSS SECTIONS OF EU ISOTOPES

Nuclide	Eu-151	Eu-152	Eu-153	Eu-154	Eu-155
Half-life	stable	13 a	stable	8.5 a	4.96 a
σ_{γ} , barns	9200	12800	390	1340	3950

practically coincides with that of Np-238 - about 50 hours - so the rate of reactivity dropping after a shut down increases and after a few days a restart may become impossible without complete cleaning out of samarium.

If Eu isotope yields become significant for heavier MA europium poisoning may be important as Table IV shows. Eu-151 poisoning will increase almost five-fold, Eu-152 poisoning - almost four-fold etc.

4.4.3. Circulating precursors of the delayed neutrons

The delayed neutrons play no special role in subcritical accelerator driven systems. But there still remains the problem of accidental criticality no matter how small its probability is and in the case of such emergency low number of the delayed neutrons aggravates the dangers. There is little direct experimental information on the delayed neutron parameters of the key short-lived actinides burning in high fluxes, total yields may be evaluated from systematics [14] which gives $\nu_d=0.01$ for ^{238}Np and 0.005 for ^{242}Am . Those values are much lower than for uranium. Besides a considerable portion of delayed neutrons (up to about one half) are emitted outside the core decreasing the effective average further. Eq.(3) must be applied in full form to calculate the in-core share of delayed neutron precursors because most of them are very short-lived. Whether the delayed neutrons induce fission and multiply significantly outside the blanket depends on the geometry of the circuit but in any case neutron background would be very strong around the circuit tubing because $10^{-4}-10^{-3}$ of all the neutrons are produced there.

5. LOWER FLUXES FOR PLUTONIUM DOMINATED MIXTURES

Plutonium isotopes are the main part of the spent fuel actinide composition, uranium excluded, so the policy toward plutonium is decisive in designing transmutation schemes. It may be considered either as valuable major fuel element or just as another waste component to dispose of. Intermediate concepts are possible and the latest ATW version [7] seems to be one of them. It uses blanket configuration similar to CANDU core, physical flux 10^{15} and waste mixture which is 89 per cent plutonium for fuel (see Table 5). This version is already rather close to

for neptunium and uranium correspondingly. Np-238 and U-235 are fissioning nuclides, Np-237 and U-238 are absorbers. Multiplying and dividing right hand sides of the expressions (6) by ϕ we can present both derivatives in the form $r\Delta\phi/\phi$ and substituting the values of cross sections and densities to calculate crucial coefficient r determining the reaction of two systems on the same relative "jump" of the flux. The results are $-3.8 \times 10^{-9} \text{ s}^{-1}$, $3.5 \times 10^{-6} \text{ s}^{-1}$ and 6.7×10^{-6} for uranium at $\phi=10^{14}$, neptunium at $\phi=10^{15}$ and $\phi=10^{16}$ correspondingly. So the neptunium system is in a sense approximately 1000 times more dynamic than uranium one in the considered conditions. And with uranium the feed-back is negative (stabilizing) while it's positive (destabilizing) for neptunium.

4.4.2. Poisoning

Very high flux complicates poisoning problems, sometimes qualitatively. First, some "weak" poisons turn "strong". Second, equilibrium concentrations are reached faster. Third, intense neutron capture transforms some decay chains strengthening minor branches. Fourth, increased poisoning after shut down grows with the flux. Xenon, main poison in solid fuel, gets eliminated from the liquid very fast, in a minute or so [13], but even that is probably not fast enough because xenon burning half-time is only 26 seconds in the flux 10^{16} . Prometium-samarium chain becomes most important and will be considered now. Heavier isotopes of samarium must be taken into account. The parameters of samarium poisoning in high fluxes are given in Table III.

TABLE III. SAMARIUM POISONING IN HIGH FLUXES

$\phi, 10^{-2} \text{ s}^{-1}$	$T_{1/2}^{\text{eff}}$, hours			$\rho_{\text{Pm}}/\rho_{\text{Sm}}$	
	^{149}Pm	^{149}Sm	^{150}Sm	^{151}Sm	
10^{14}					
10^{15}	1.73	51	48	2.2a	128
10^{16}	1.73	38	4.8	80d	12.8
	1.73	11	0.48	8d	1.28
					190

The yields of specific fission products are not known for short-lived MA so it's impossible to normalize their poisoning data properly, but the yields of heaviest fragments grow with the mass number of the fissioning nuclei so the Pm-Sm chain yield of about 1% is a reasonable assumption.

The periods in the Table III are to be compared with chemical by-pass period, it's planned value being about a month. At the flux level 10^{16} all three Sm isotopes reach equilibrium concentration so the poisoning is tripled. "Samarium well" deepens manifold as the last column of Table III shows and unlike iodine well it does not disappear. Pm-149 half-life

TABLE V. INITIAL AND EQUILIBRIUM COMPOSITIONS AND NUCLEAR DATA OF THE FUEL MIXTURE OF THE MODERATE FLUX ATW VERSION [7] ($\Phi = 10^{15}$, $\Phi_{eff} = 2 \times 10^{14}$).

Isotope	Rel. concentration	σ_f, b	σ_f, b	$T_{1/2}$
Initial Equilibrium [7]				
237Np	0.0449	0.0317	176	2.14×10^6 a
238Np	0.	0.0003	203	2.117d
Total Np	0.0449	0.0321		
238Pu	0.0140	0.0230	540	87.71a
239Pu	0.5148	0.0610	269	24 100a
240Pu	0.2372	0.1003	289	6569a
241Pu	0.0785	0.0467	358	14.355a ⁵
242Pu	0.0481	0.2630	18.5	3.763×10^4 a
243Pu	0.	0.0002	87	4.956h
244Pu	0.	0.0026	1.7	-
Total Pu	0.8925	0.4968		8.1×10^4 a
241Am	0.0513	0.0048	587	432.6a
242Am	0.	0.0001	-	16.1h
242mAm	0.0001	0.0001	1344	152a
243Am	0.0092	0.0874	75.1	7730a
Total Am	0.0607	0.0924		
242Cm	0.	0.0213	16	162.8d
243Cm	0.	0.0006	130	28.5a
244Cm	0.	0.2794	18.1a	18.1a
245Cm	0.0018	0.0051	369	8500a
246Cm	0.0001	0.0004	1.22	4700a ₇
247Cm	0.	0.0094	57	1.6×10^5 a
248Cm	0.	0.0625	2.63	3.5×10^4 a
Total Cm	0.0019	0.3787		

normal thermal reactor as far as the neutron balance and the burning rates of various isotopes are concerned, its effective flux is only 2×10^{14} . The well fissioning MA - which are the heart of initial ATW scheme - play only secondary, almost negligible role. As a consequence very hazardous isotopes like Pu-238, Pu-242, Cm-244, Cm-248 are accumulating.

6. A POSSIBILITY TO EXPLORE: STATIC LIQUID BLANKETS

The facilities with circulating liquid fuel may be called "inside-out" blankets because usual situation is vice versa - a coolant circulates through a static fuel. That's quite possible for the liquid fuel too and even looks preferable from many important points of view. The possibility is mentioned for heavy water electronuclear actinide burners in [] so it's not quite clear why should fuel circulate at all.

The first major difficulty eliminated in static blanket is the distinction between physical and effective fluxes. The difference may be decisive. The second is not less important: the primary circuit becomes practically not radioactive and the number of circuits may be reduced to firm two and even a channel type single circuits with drum-separators are not unthinkable because the tubes walls may be made much thicker than fuel pin shells and practically hundred per cent reliable.

Small actinide inventory of the blankets with high thermal neutron flux is considered as one of their main advantages, increasing safety and enabling continuous chemical reprocessing. In earlier versions inventory gain was estimated as approximately hundred-fold, in later versions it's almost vanished. Fuel circulation increases the inventory 1/r times, i.e. manifold. It's one more argument in favor of static blankets.

7. CONCLUSIONS

1. The reactor physics and safety problems of heavy water MA blanket burners with the neutron flux close to $10^{16} \text{ cm}^{-2} \text{ s}^{-1}$ look very serious indeed, the most important are following: the difficulty of providing needed effective flux; tritium accumulation if the flux is reached; high radioactivity of the primary circuit liquid; water radiolysis by fission fragments with profuse hydrogen release; samarium poisoning.
2. Tritium accumulation and poisoning relax in the schemes with relatively moderate flux of a few units by 10^{14} but radiolysis and radioactivity in the primary circuit depend on the total power only and stay high. The fission rate of MA goes down to a hardly acceptable level with simultaneous increase of the accumulation of heavier, long-lived and poorly fissioning nuclides like Pu-238, Pu-242 and Cm-248.
3. Static liquid fuel blankets deserve detailed consideration although they may help to solve only some of the problems listed above.
4. In fact an accelerator in subcritical system plays role of an inertialess emergency shut down rod and from the point of view of actinide burning eliminates the problem of delayed neutron deficit only. Very high costs call for detailed discussion of the efficiency of its use in such a role because, as its follows from above consideration, other reactor problems get more complicated. The accelerator problems and, especially, the difficulties of continuous radiochemical processing of a liquid fuel mixture with short cooling times must be considered separately.

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SOME PHYSICS ASPECTS OF MINOR ACTINIDE RECYCLING IN FAST REACTORS

U.K. WEHMANN
Siemens AG,
Bergisch Gladbach,
Germany

Abstract

In the paper some results of studies on cores with the potential for a reduced SVE are described and the influence of MA recycling on these cores will be discussed.

1. INTRODUCTION

In a previous study / 1 / it was shown that the introduction of Minor Actinides (MA) into a large sodium cooled fast reactor core has the main consequences to

- increase the positive sodium void effect (SVE) with a simultaneous reduction of the Doppler constant
- to reduce the burnup reactivity loss.

Since an increased SVE is an important drawback for the core behaviour in accidental conditions on one side and for the public acceptance of fast reactors on the other side, there is a strong incentive for SVE reductions especially for MA burning cores.

Most of this work is part of a paper to be presented at the Tokyo Conference 1992 /2/.

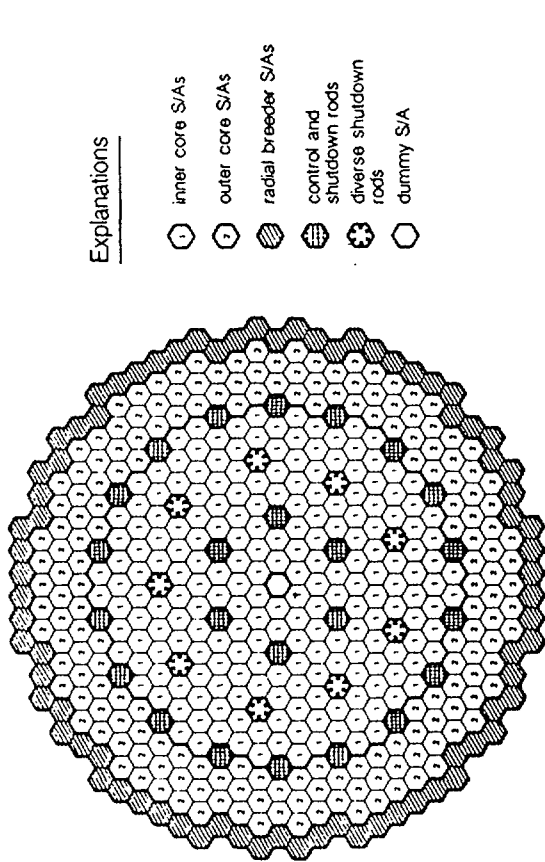
2. STUDIES DEVOTED TO SODIUM VOID EFFECT REDUCTION

In this study the core size reduction was applied as instrument to reduce the sodium void effect. Starting point was the European Fast Reactor EFR with a thermal power of 3600 MW, a cross-section of which is shown in Fig. 1.

Core size reduction was realized by, firstly, reducing the core height from 1 m to 0.7 m and 0.5 m with constant core diameter of 4 m and, secondly, by reducing the core diameter from 4 m to 1.8 m. This was achieved by reducing the number of fuel S/As and the number of pins per S/A. Fig. 2 shows a cross-section of the smallest core.

Tab. 1. Design and performance parameters of the core variants with different core size (EOEC)

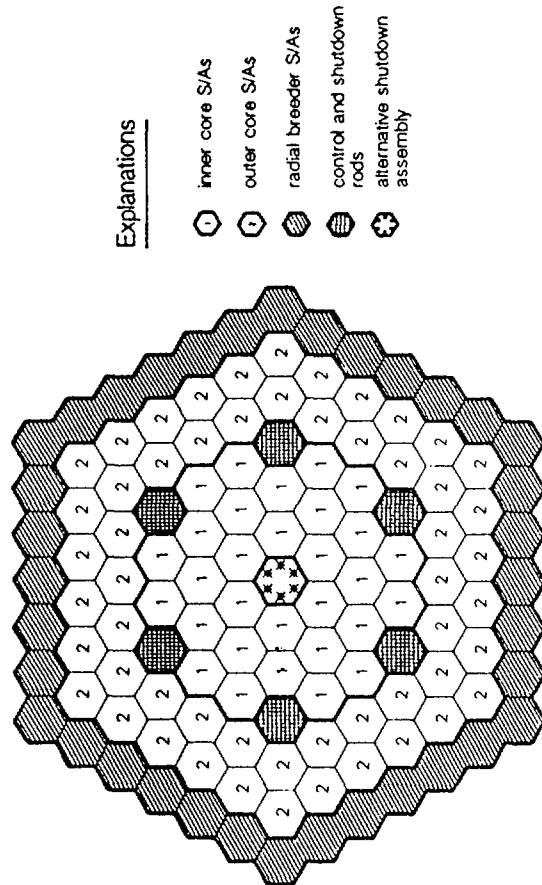
parameter	unit	core 1	core 2	core 3	core 4
thermal power	MW	3600	2600	2000	450
core height	m	1.0	0.7	0.5	0.7
core diameter	m	4.0	4.0	4.0	1.7
no. of fuel S/As	-	375	375	375	84
no. of control rods	-	24	24	24	6
no. of pins per S/A	-	331	331	331	271
fissile SVE	\$	5.9	4.4	3.0	2.5
total SVE	\$	5.5	3.6	2.2	1.9
fissile Doppler constant	\$	-1.7	-1.6	-1.4	-1.1
reactivity loss per full power year	\$	-8.9	-10.4	-12.8	-12.3
worth of control rods	\$	25	23	19	31



Explanations

- ① inner core S/As
- ② outer core S/As
- ③ radial breeder S/As
- ④ control and shutdown rods
- ⑤ diverse shutdown rods
- ⑥ dummy S/A

Fig. 1. EFR core cross-section



Explanations

- ① inner core S/As
- ② outer core S/As
- ③ radial breeder S/As
- ④ control and shutdown rods
- ⑤ alternative shutdown assembly

Fig. 2. Cross-section of the small core with 450 thermal MW

The design data of the four different cores are gathered in Table 1 together with the main performance data. For the SVE two values are given: one for the case that only the fissile zones are voided and another one (total SVE) which also includes the void effect of the upper axial blanket and the short fission gas plenum above.

Table 1 shows the strong reduction of the fissile SVE from 5.9 \$ to 2.5 \$ when going from core 1 to core 4 which is even more pronounced for the total SVE (5.5 \$ to 1.9 \$) since the enhancement of the axial leakage in case of voiding is stronger in the flatter cores.

This strong SVE reduction should offer the potential to design minor actinide containing cores without reaching the SVE level of the large cores.

3. STUDIES ON CORES WITH MINOR ACTINIDES

For this part of the study, which has been performed within a contract with the European Community, the cores no. 1, 2 and 4 have been chosen as potential MA-burners.

Tab. 2. Performance parameters of cores with different MA contents (EOEC)

parameter	MA-content	core 1	core 2	core 4
fissile SVE [\$]	0 %	5.9	4.4	2.5
	5 %	6.5	5.1	3.1
	15 %	7.5	6.3	4.8
Doppler constant [\$]	0 %	-1.7	-1.6	-1.1
	5 %	-1.3	-1.2	-0.8
	15 %	-0.7	-0.6	-0.5
Δg per full power year [\$]	0 %	-8.9	-10.4	-12.3
	5 %	-4.6	-6.7	-8.9
	15 %	+1.6	-1.5	-4.9
control rod worth [\$]	0 %	25	23	31
	5 %	24	21	30
	15 %	20	17	27

In these cores 5 % and 15 % of the heavy metal content were replaced by MA with a composition of

Np237 : Am241 : Am243 : Cm244 = 49 : 37 : 11 : 3,

which is typical for a Light Water Reactor UO₂-fuel with an average discharge burnup of about 40 MWd/kg HM. As it was done for the cores without MA, the core was burnt starting from the BOL state to beginning and end of equilibrium cycle with a total fuel residence time of 6 years. The main core characteristics calculated for the end of equilibrium cycle are collected in Table 2.

Concerning the SVE it is interesting to point out the different burnup dependence in cores with and without MA. The BOL and EOEC SVE values in the three considered cores are the following (in \$):

	core 1	core 2	core 4
0 % MA {			
BOL	3.6	2.2	0.7
EOEC	5.9	4.4	2.5
15 % MA {			
BOL	7.4	5.8	3.9
EOEC	7.5	6.3	4.8

One can see that the SVE increase with 15 % MA is much smaller than without MA. This is caused by the fact that MA and fission products have a similar influence on the neutron spectrum so that the effect of the fission product built-up during burnup is weakened due to the presence of the MA.

Generally, one can conclude that the increase of the SVE at EOEC due to MA introduction is increasing with core size reduction: it changes from + 1.6 \$ in core 1 to + 1.9 \$ in core 2 and + 2.3 \$ in core 4. But the value for core 4 with 15 % MA (4.8 \$) is still smaller than the value for core 1 without MA (5.9 \$).

The other parameters mentioned in Table 2 are also strongly influenced by the MA introduction:

- The Doppler constant is strongly reduced.
- The reactivity loss per full power year is strongly improved.
- The control rod worth is slightly reduced but this is of no relevance for the fulfillment of the shutdown requirements due to the reduced burnup reactivity loss.

4. CONCLUSION

The study has shown that core size reduction is a very effective way to reduce the SVE of the fissile zones. It requires, however, a reduction of the total power if the design limits and the basic design data, like fuel pin diameter, are to be kept constant.

Due to their reduced SVE the smaller cores have the potential for an introduction of up to 15 % Minor Actinides without exceeding the SVE level of the reference core 1. The acceptability of the reduced Doppler constant of the MA containing cores has, however, still to be demonstrated.

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ROLE OF FAST REACTORS IN REDUCTION OF LONG LIVED WASTE

A.G. TSIKUNOV, V.S. KAGRAMANYAN,
L.A. KOCHETKOV, V.I. MATVEYEV
Institute of Physics and Power Engineering,
Obninsk, Russian Federation

Abstract

Role of fast reactors in the problem of lowering of minor actinides (MA) quantity in the fuel cycle of nuclear power is considered in the report. The requirement to MA quantity in closed fuel cycle is formed. The results of analysis of BN-800 reactor possibilities for MA quantity reduction at PO MAYAK are discussed in the report. The recommendations are given on choice of actinide extraction coefficient values during the chemical reprocessing of spent fuel. The coefficients are the following: for Pu and Am - 0.9998, for Np and Cm - 0.98.

Development and construction of NPP with fast reactors and their further progress are tightly connected with the closed fuel cycle creation. It is necessary not only for the concept of nuclear fuel breeding realization but for solution of more urgent task for the present - environmental problems.

Considerable quantity of NPP in operation to-day, using once-through fuel cycle produce radioactive waste, disposal of which is one of the most important tasks of global character. Fast power reactors can play an important role in solution of the problem.

In the paper the above-mentioned role of fast reactor in the nuclear power system is being considered. The analysis of the question should be based on the concept that in long term perspective, under conditions of nuclear power system normal organization any increase of the environmental radioactivity level must be excluded.

To realize the concept, at least, two problems should be solved:

- organization of a reliable burial of fission products under surveillance (for hundreds of years);
- organization of produced actinides burning.

As a matter of fact, radiotoxicity of fission products remains higher than that of natural uranium for about the first 500 years.

Besides, in the process of nuclear reactors operation not only fission products are formed, but at the same time

activation products are arising. The most important ones from ecological point of view are the long-lived actinides (Pu, Am, Cm), which determine radiotoxicity of high level activity waste after about 500 years of cooling (storage) and the level of their danger does not diminish during a million of years.

One of the most dangerous (from ecological point of view) elements among these actinides in the spent fuel is plutonium. Its potential deleterious effect on environment about 10 times higher than that of all other actinides (Np, Am, Cm) in total. Closed fuel cycle formation i.e. repeated use of not completely burned up and build up fission materials, means actually beginning of waste minimization process or, if other things equal, reduction of uranium demand. Really all the countries with developed nuclear power follow this approach in the fuel cycle organization. Nevertheless, up to now under consideration were only fuel cycles with extraction of U and Pu for further recycle in reactors. In this case because of long-lived α -emitters ^{241}Am ($T_{1/2}=433$ yr.) and ^{237}Np ($T_{1/2}=2.14 \cdot 10^6$ yr.) build up one should put forth the task of out of reactor fuel cycle time reduction.

It is reasonable to put a question: what type of reactor is the most suitable for use of ecologically dangerous plutonium? The question is particularly sharp for plutonium with high content of isotopes, having mass number $M > 239$.

Conducted evaluations show that the specific (per GW(e)yr) radiological hazard of the long-lived actinides in the spent fuel for fast reactors is less than that for thermal reactors. The conclusion is true for both MOX and uranium fuel.

Calculations showed that it is possible to use as a nuclear fuel in fast reactor not only plutonium but neptunium, americium, curium.

When considering this problem one should have in mind that the benefit of transmutation (as compared to once through cycle) is exhibited after relatively long time interval. Proceeding from this fact such a requirement determining the efficiency of minor actinides transmutation can be formulated: - the total content of actinides at all stages of closed fuel cycle should amount 0.1-1.0 % as related to their quantity produced in once through cycle.

The chosen time interval may correspond, for instance, to the reactor life-time, or life-time of the nuclear power system. This condition has been formulated mainly on the base of requirements to the depth of actinides extraction from spent fuel in reprocessing (actinides content must be reduced by 10²-10³ times).

Radiotoxicity of all minor actinides in this fuel cycle after 500 years of storage would not be higher than radiotoxicity of initial natural uranium.

Conducted evaluations showed, that in the case of minor actinides recycle their content in the fuel cycle arrives at the steady state level. Minimum of minor actinides will be in the system consisting of fast reactors only. In this case total quantity of minor actinides in uranium-plutonium fuel entering reactors will be at the level of 0.7%. Minor actinides quantity in the total closed fuel cycle on the base of fast reactor will not be higher than 1% of minor actinides, which could be produced in the nuclear power system during 150-200 years of thermal reactors operation having the same power and once through fuel cycle.

In reality fast reactors will represent only certain part of the nuclear power system. Under these conditions minor actinides content in the fast reactors fuel cycle will surpass the 0.7% value. Obviously, such a reactor should be adapted to the use of actinides bearing fuel. Inclusion of minor actinides in the traditional fuel gives to it some specific features in subassemblies management and in the core neutronics.

If the traditional BN-800 fast reactor is used for actinides incineration, their quantity in the fuel will be restricted by the condition of initial core characteristics retention, and first of all those accounting for reactor safety. The minor actinides content in the fuel should not be higher than 3-4% of total quantity of heavy atoms. Already this quantity appreciably affects the reactor physical characteristics, especially sodium void reactivity, and requires partial refurbishing of fuel handling system, designed for the "standard" MOX fuel.

To increase the efficiency of actinides burning would be reasonable to reject ^{238}Pu as a fertile material producing high activity actinides in the course of irradiation. Further development of this idea results in concept of a core with an inertial diluent such as, for instance, zirconium, or zirconium based material.

Can the present day traditional fast reactors solve the practical task of bringing down minor actinides quantity in the fuel cycle?

To answer this and other questions, related to substantiation of a necessity (or expediency) to transmuted the minor actinides, the authors attempted to obtain on the base of their analysis of possible use for these purposes BN-800 reactor on the site of PO "MAYAK".

To the date of BN-800 reactor commissioning appreciable quantity of actinides is supposed to be accumulated as a result of spent fuel reprocessing from thermal uranium reactors VVER-440 and fast reactors BN-350 and BN-600.

The maximum life-time of NPP with BN-800 reactor assumed to be 60 yr. Storage time of fast reactors spent fuel before reprocessing assumed to be 3 yr. Efficiency of actinides (Pu, Np, Am, Cm) extraction in the reprocessing of spent fuel assumed to be 0.98 (conservative approach).

Three possible situations at the site of PO "MAYAK" are being considered.

1. BN-800 is not commissioned. Transuranium elements from VVER-440, BN-350 and BN-600 reactors are stored during 60 year after year 2000 and then directed to the long-time burial.

2. BN-800 reactor is commissioned in the year 2000 in the traditional concept of uranium-plutonium fuel cycle. The breeding ratio is equal to 1. So called "dirty" plutonium produced by thermal reactors VVER-440, as well as extracted from the spent fuel of the BN-800 reactor core is assumed to be used as its fuel. In the case of "dirty" plutonium deficiency it will be covered by "pure" plutonium produced in fast uranium reactors and in the radial blanket of BN-800 reactor. Minor actinides in this version are not extracted but buried together with fission products.

3. The BN-800 reactor to be commissioned in the year 2000 using uranium-plutonium fuel mixed with minor actinides. Maximum content of minor actinides amounts to 4% of heavy atoms in the very beginning, when there is a lot of minor actinides in the storage of spent fuel. Then, when minor actinides in the thermal reactors storage will be exhausted, their content in the fresh fuel of BN-800 reactor is diminished.

All data concerning estimates of different nuclides quantities on the site PO "MAYAK" by the year 2060, for three considered situations, are presented in the table 1.

TABLE 1. QUANTITY OF RADIONUCLIDES AT PO MAYAK FOR THREE DIFFERENT OPTIONS OF FUEL CYCLE, KG

Nuclide	Nuclide quantity as of 2060 year			
	quantity as of 2000y	Option 1	Option 2	Option 3
^{237}Np	550	720(720)	785(785)	60(45)
^{241}Am	420	2450(2450)	2040(2040)	370(170)
^{243}Am	150	150(150)	630(630)	195(45)
^{244}Cm	40	5(5)	25(25)	100(10)
^{238}Pu	165	110(110)	40(20)	275(75)
^{239}Pu	17600	17600(17600)	17230(1750)	17470(1730)
^{240}Pu	4370	4400(4400)	3810(840)	3770(830)
^{241}Pu	2350	165(165)	420(80)	410(90)

Comment: Nuclide quantities which go for final disposal are indicated in brackets.

TABLE II. POTENTIAL HAZARD INDEXES OF ACTINIDES IN WASTES AFTER 103 YEARS, LITERS OF WATER.

Group of actinides	Option of fuel cycle		
	1	2	3
Np	0.03*10 ¹⁰	0.04*10 ¹⁰	0.002*10 ¹⁰
Pu	100*10 ¹⁰	14*10 ¹⁰	14*10 ¹⁰
Am	170*10 ¹⁰	140*10 ¹⁰	12*10 ¹⁰
Cm	0.07*10 ¹⁰	0.3*10 ¹⁰	0.14*10 ¹⁰
Total	270*10 ¹⁰	154*10 ¹⁰	26*10 ¹⁰

Analysis of these figures makes it possible to conclude:

1. In spite of small difference in the total quantity of nuclides in versions 1 and 2 the latter gives an essential reduction of nuclides which need superlong-time burial. Quantity of plutonium to be buried in the version 2 is an order of magnitude lower than that in the version 1.

2. More radical is solution of ecological problems in the version 3. One can see that here not only the quantity of plutonium to be buried is an order of magnitude less than in the version 1, but quantities of neptunium and americium to be buried are an order of magnitude less than in versions 1 and 2 as well. Recycle of MA results in reduction of their quantity in all the system (storage+BN-800). Exclusion is 24Cm.

Whether such a minimization of actinides quantity is sufficient or not. Answer to this question can be obtained by the way of comparison of the waste to be buried radiological toxicity with that of natural uranium. Here the quantity of natural uranium is meant which was used for energy production by VVER-440 reactors and by BN-800 reactor during 60 of its operation.

According to estimates the quantities of fission products and artificial actinides accumulated due to VVER-400 reactors operation till the year 2000 and BN-800 reactor during 60 years amount to 100t and 30 t respectively. This process of energy production will result in disappearance in the nature of about 130 t uranium. Radiological hazard of any nuclide is accepted to evaluate by the potential hazard index (PHI). It is determined as a quantity of water (air), which is needed to dilute the specific content of a radionuclide to its tolerable concentration level.

The potential hazard index of 130t uranium is evaluated as 0.4 10¹⁰ liters of water (taking into account daughter products of uranium isotopes). PHI of actinides buried for different scenarios of fuel cycle development are expedient to compare with each other approximately 103 years later, when it will be difficult to rely upon artificial barriers and when relative contribution of actinides in potential hazard index will reach maximum.

In table 2 are listed the calculated PHI of actinides in wastes after 103 years for three above-mentioned options.

It is clear from table 2 that putting into operation the BN-800 reactor into any of the above-mentioned options of fuel cycle organization leads to lowering of level of wastes PHI in comparison with the case of final disposal of VVER-440 reactor wastes.

Under recycle in the BN-800 reactor plutonium only it is possible to reduce PHI two times. Involvement into the recycle

all minor actinides increases the scale of lowering of the VVER-440 disposed wastes PHI up to 10 times, but for the variant the value of the actinides PHI in the wastes after 103 years will be, however, approximately two orders of magnitude higher than ideal objective - 0.4*10¹⁰ liters of water.

The analysis shows that the ideal objective could be achieved only under the condition of lowering of Am and Pu (the main investors into PHI) discards value during reprocessing almost two orders of magnitude, i.e. from 2% to 0.02%. As for extraction coefficients of Np and Cm we don't need special tightenings up in comparison with the coefficients adopted in our calculations, namely, 0.98.

Summing up aforesaid, one can affirm that in near future the fast reactors could solve the problem of limitation (up to demand of the Earth's radiation "status quo" conservation) of long-lived and especially dangerous part of the nuclear power wastes like actinides.

ACTINIDE TRANSMUTATION IN THE ADVANCED LIQUID METAL REACTOR (ALMR)

C.L. COCKEY

GE Nuclear Energy,
San Jose, California,
United States of America

Abstract

The Advanced Liquid Metal Reactor (ALMR) is a US Department of Energy (DOE) sponsored fast reactor design based on the Power Reactor, Innovative Small Module (PRISM) concept originated by General Electric. The current reference design is a 471 MWt modular reactor loaded with ternary metal fuel. This paper discusses actinide transmutation core designs that fit the design envelope of the ALMR and utilize spent LWR fuel as startup material and makeup. Actinide transmutation may be accomplished in the ALMR by using either a breeding or burning configuration. Lifetime actinide mass consumption is calculated as well as changes in consumption behavior throughout the lifetime of the reactor. Impacts on system operational and safety performance are evaluated in a preliminary fashion.

1. INTRODUCTION

The objective of the Advanced Liquid Metal Reactor (ALMR) Program, under the sponsorship of the U.S. Department of Energy, is to develop a competitive reactor system aimed at improving safety, enhancing plant licensability, and simplifying plant operations. This activity, led by GE, is a national program involving wide participation by U.S. industries, the U.S. national laboratories, universities, and international contributors. One of the goals of the program is to develop a standardized design that can be licensed and certified. The current reference design (see Figures 1 & 2) is a 471 MWt modular reactor, with nine modules constituting a 1440 MWe plant (Ref. [1]). The reference core design is a breeder core utilizing ternary metal fuel under development by Argonne National Laboratory (Ref. [2]). This paper discusses actinide recycle (or burning) core designs that fit the design envelope of the ALMR and utilize irradiated LWR fuel as startup material and for makeup.

Actinide recycle (frequently referred to as "actinide burning" or "transuranic burning") has long been an intriguing concept associated with closing the nuclear fuel cycle and improving waste management. The concept involves transmutation or fissioning of the higher actinides (transuranic [TRU] isotopes) to shorter lived fission products. The primary incentives for transmutation of these TRU isotopes is to eliminate as many of them as possible from the ultimate waste stream via processing spent fuel and to recycle the TRU as an ALMR fuel source. LWR irradiated fuel contains up to about 16% minor actinides (MA-neptunium, americium and curium) relative to total TRU, depending on burnup as shown in Table 1. The current U.S. LWR spent fuel inventory averages about 7.7 w/o MA.

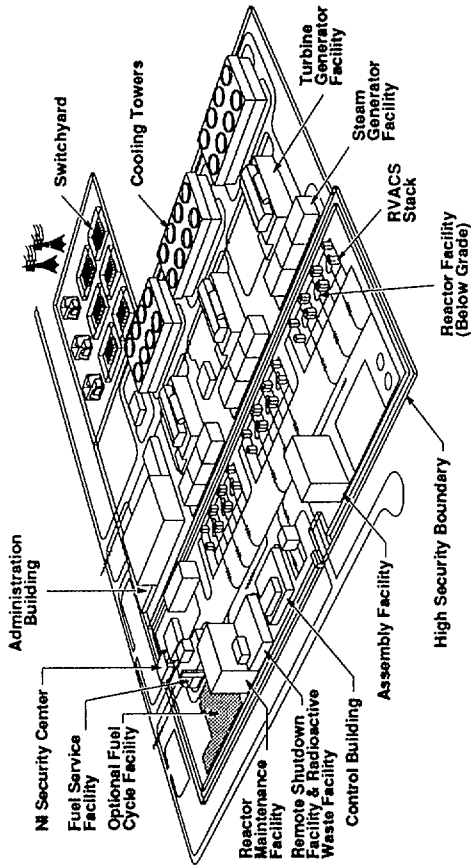


FIG. 1. ALMR Power Plant (3 Power Blocks) - 1440 MWe

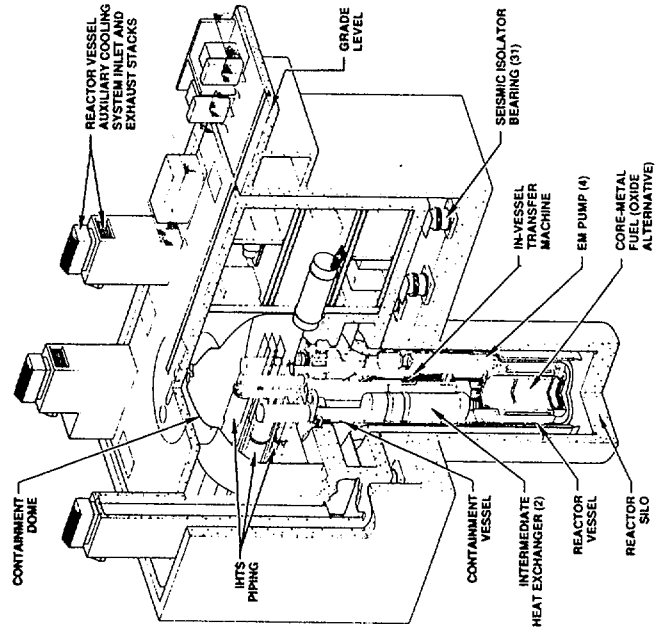


FIG. 2. ALMR Reactor Facility

TABLE I. COMPARISON OF ISOTOPIC COMPOSITIONS IN SPENT FUEL

	Ref. Equil.	Existing	Current	Future
	ALMR Fuel	US LWR Inventory	LWR 33 GWD/t	LWR 50 GWD/t
Plutonium Isotopes (%)				
Pu-238	0.9	1.2	1.5	3.4
Pu-239	75.4	60.1	58.2	55.3
Pu-240	21.3	23.9	26.6	20.1
Pu-241	1.5	10.4	8.5	11.7
Pu-242	0.9	4.5	5.2	9.4
Minor Actinides (%)				
Np-237	26.5	47.8	42.0	42.8
Am-241	60.3	39.6	48.3	40.8
Am-242M	3.3	0.1	0.1	0.1
Am-243	7.7	9.9	8.0	12.7
Cm	2.2	2.5	1.6	3.6
Total TRU (%)	97.4	92.3	89.2	83.4
Plutonium	2.6	7.7	10.8	16.6
TRU in Heavy Metal (%)	25.9	0.88	1.02	1.26

The purpose of the work described in this paper was to focus on 'safe actinide burners'. The ALMR is a reactor system that uses passive safety features, and the actinide burning cores should not have a significant impact on these features. The work was done in the context of this system and an evaluation was made of the impacts on the cores' safety features (Ref. [3]).

There are two basic processing concepts, aqueous and pyroprocess, to achieve actinide recovery from spent LWR fuel for recycle to the ALMR. Aqueous processes and pyroprocesses may be used for recovery of actinides from both LWR and ALMR spent fuel and recycle to the ALMR. The pyroprocesses for processing LWR and ALMR spent fuel are the reference processes in the ALMR program. Unlike the aqueous process, the pyroprocess does not separate Pu from other TRU. A specific Pu enrichment from LWR spent fuel therefore carries a specific MA enrichment. In this scenario, to continuously consume MA for a reactor's lifetime, it is necessary to continuously consume Pu.

2. PLANT DESCRIPTION

The ALMR plant utilizes nine reactor modules arranged in three identical 480 MWe power blocks. Each power block consists of three identical reactor modules, each with its own steam generator, that jointly supply steam to a single

turbine generator. A sodium-filled intermediate heat transport system (IHTS) provides normal heat removal for the reactor through an intermediate heat exchanger (IHx) and transports the energy to the steam generator. The reactor facility is seismically isolated to provide high margins in seismic capability.

The reactor, shown in Figure 2, is a pool-type containing the core, two intermediate heat exchangers, four primary electromagnetic (EM) pumps, and interim spent fuel storage. Containment is provided by a low leakage, pressure-retaining boundary, which completely encloses the reactor coolant boundary. Containment consists of a lower containment vessel surrounding the reactor vessel and an upper dome over the reactor closure.

The current reference reactor fuel is metallic uranium-plutonium-zirconium alloy, and ferritic alloy HT9 is used for the cladding and assembly ducts to minimize swelling associated with high burnups. Metal fuel provides competitive fuel costs and excellent negative reactivity feedback during transient overpower and loss of cooling events. An alternative oxide core is under development with international partners. The reference breeder core is a heterogeneous arrangement of blanket and driver fuel, with six control rods. Refueling occurs after 24 months of operation, with one-third of the driver fuel replaced each cycle. The fuel is not shuffled, but blanket assemblies are shuffled each outage. The burnup reactivity swing is small during the cycle for the equilibrium core, making axial swelling of fresh metal fuel assemblies the largest contributor to the cycle reactivity swing.

Reactivity control for normal operations of startup, load following, and shutdown is accomplished by bank movement of the six control rods. Each control rod absorber assembly has sufficient worth such that any one of the six can shut down the reactor to cold subcritical conditions. Positive reactivity addition by inadvertent withdrawal of the control rods is limited by electronically positioned mechanical rod stops.

For safety margin in the event of loss of primary coolant flow without scram, three gas expansion modules (GEMs) have been included at the periphery of the core. The GEMs are hollow assembly ducts, which are open to flow at the bottom like the driver fuel but are closed to flow at the top. The GEMs are filled with vessel cover gas before insertion into the core, and this gas is compressed as the GEMs fill with sodium. The GEMs are designed such that the sodium level inside the GEMs at normal operating pressures is above the top of the active core. When pumping is lost in the primary system and the pressures drop, the gas inside the GEMs expands and expels most of the sodium. The resultant void near the core periphery increases neutron leakage (provides negative feedback) and reduces the net reactivity of the core.

An ultimate shutdown system is included as a means of bringing the reactor to cold subcritical conditions in the event of a complete failure of the normal scram system. This device is manually actuated after the core has been brought to a safe stable state by the core thermal feedbacks.

3. CORE DESCRIPTION

Two major types of ALMR cores were studied; radial heterogeneous breeders and homogeneous burners (see Figure 3). For the reference breeder, LWR spent fuel fissile material was selected as the reference startup fuel for the ALMR and will be recycled in the ALMR until the TRU content approaches the equilibrium TRU value for an ALMR. This material contains Pu plus associated minor actinides from the LWRs. Only startup fissile material is required for the reference breeder ALMR. This type of core will be fissile self-sufficient and will consume LWR minor actinides loaded into the startup core plus two reloads.

If the ALMR is used to reduce the total amount of LWR actinides in waste destined for disposal, fissile material and other TRU isotopes can be transmuted or burned in ALMRs. Various amounts of TRU can be burned depending on the design of the ALMR core. In order to consume the largest amounts of TRU with metal fuel, a burner core was designed. The approach used in this case to modify an ALMR from a breeder to a burner was to change the core from a heterogeneous arrangement with blanket assemblies to a homogeneous arrangement with no blankets. The blanket assemblies were replaced with fuel assemblies and the radial blankets were replaced with an extra row of shield assemblies in order to compensate for the effect of the deleted radial blanket assemblies on shielding. The core height was decreased to provide decreasing conversion ratios and increasing TRU consumption rates. Impacts on system operational and safety performance were evaluated in a preliminary fashion. The power level remained at 471 MWt, and the designs were evaluated according to current ALMR design and safety parameters.

The driver fuel assemblies are divided into two enrichment zones, lower enrichment in the core center and higher enrichment in the outer core rings. This was done to reduce the radial power peaking. Core heights ranging from 135 cm (53 in - reference) to 89 cm (35 in) were studied for actinide burning benefits and safety characteristics. Cycle lengths were adjusted to account for peak fast fluence limits on the fuel. The reference core has a 24 month cycle, while the 135 cm burner core has an 18 month cycle and the 89 cm core has a 13 month cycle.

A 89 cm core height burner design does not meet ALMR design/safety criteria due to high burnup reactivity swing (\$12) and peak linear power constraints, as defined for metal fuel by ANL. This core is considered to be beyond the envelope of ALMR burner core design. A 102 cm (40-in) core is marginal in performance due to its burnup reactivity swing of 11\$, although this value is probably within the bounds of control design. An added benefit of reducing core height is the reduction of sodium void worth, but this is a minor safety benefit which is offset by other more serious safety concerns. Greater core heights give larger safety margins and larger actinide storage masses.

4. TRU MASS CALCULATIONS

The TRU and minor actinide consumption rates were calculated for specific cores. As discussed above, the reference ALMR has a net annual TRU gain (but MA loss) whereas the burner cores show a net TRU consumption (refer to values

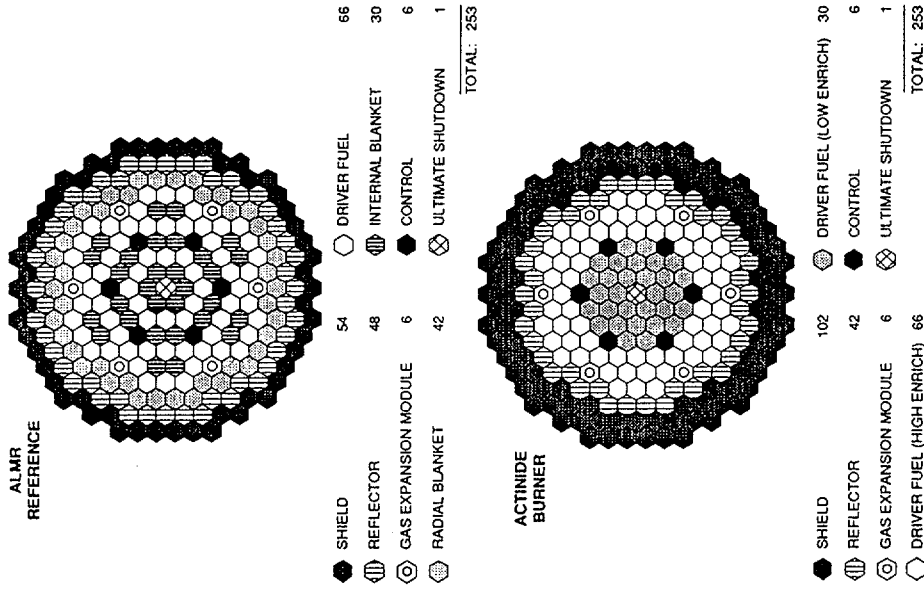


FIG. 3. Core Layout Comparison

shown in Table II). Table II shows the results of equilibrium diffusion and burnup calculations with no fuel recycle. This produces "snapshot" core values calculated approximately at Cycle 3. TRU and MA lifetime reactor consumption totals should be based on core lifetime evaluations rather than "snapshot" intervals, as consumption changes with changing composition. As the core height is reduced and the uranium-238 content is reduced, the TRU consumption rate is increased while the total core heavy metal inventory is decreased. TRU consumption is variable with the time the fuel spends in the reactor, but tends to approach an equilibrium value near the end of reactor life.

TABLE II. ALMR CORES EVALUATED FOR ACTINIDE RECYCLE

	Actinide Burning Cores	
	Reference Startup ALMR Core 135 cm	Active Core Height (cm) 102
Breeding Ratio	1.09	0.77
Cycle Length (months)	24	18
Burnup Reactivity Swing (\$)	-2.3	-9.1
Peak Linear Power (kW/m)	31.2	28.2
Sodium Void Worth (\$)	5.85	4.63
TRU Enrichment (wt% in U-TRU-Zr)	30	15/19
TRU Inventory (kg/core 471 MWh)	2196	1867
TRU Consumption Rate kg/yr	-6.9	38.7
% inventory/yr	-0.3	2.1
		51.1
		3.3
		4.0
		20/24
		1424
		56.9
		4.0

*Does not meet ALMR design criteria

ORIGEN2 calculations were completed for two cores in order to show the effects of continually loading LWR-generated TRU into the ALMR actinide burning cores. The transuranics were processed and reloaded into the core for 42 cycles. The calculations included refueling and makeup with LWR TRU, as well as k-infinity and mass normalization. One-group ALMR cross-sections were overlaid on to the standard ORIGEN2 fast reactor cross-sections. An 18 month cycle, three-batch homogeneous core with a 135 cm core height (conversion ratio 0.77) was compared to a 15 month cycle homogeneous core with three batches and a core height of 102 cm (conversion ratio 0.69). Both changing TRU consumption rates and the differing behavior of each actinide isotope were evaluated during a core lifetime of 42 cycles.

Figure 4 shows the changing concentration of each MA in the 102 cm core. These values are for one batch and are not core-averaged results. The concentration of each minor actinide appears to approach an equilibrium value. Neptunium (Np) is converted at a steady rate. The rate of production of curium (Cm) levels off to a steady value, and then gradually begins to decrease. Americium (Am) is slowly consumed. The batch nature of the graph clearly shows that Am is consumed during each cycle, but also shows that more Am is loaded at the beginning of each cycle than is transmuted. The percentage of MA in total TRU stays approximately even at 10.7%-10.9%, due to the heavy TRU makeup required for this core. The reference heterogeneous ALMR, not requiring LWR TRU makeup, would have a substantially higher rate of change in the MA concentration (from 10.7% to 2.6%).

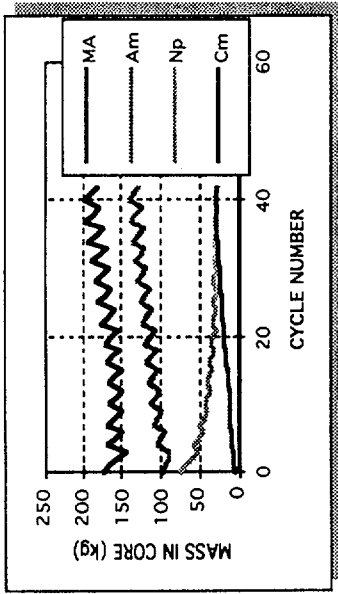


FIG. 4. Minor Actinide Mass

Every isotope exhibits a unique characteristic behavior in the actinide burner designs. Table III shows total loaded and consumption values for both the 102 cm and the 135 cm cores from ORIGEN2 calculations. No burns out steadily throughout the plant lifetime. A total of 87% of all Np loaded into the 102 cm core over 50 years is consumed, with a value of 74% for the 135 cm actinide burner. The three Am isotopes exhibit very different behavior. Am-241 is consumed in both cores (79% in the 102 cm core). Overall, the total amount of Am-242M is slightly increased. Am-243 is consumed, with values ranging from 16-23%. The various isotopes of Cm have differing behavior, but in general during the early cycles Cm is produced and it is only later in the composition lifetime (after 40 years) that any Cm isotope is consumed. The amount of Cm produced is small in terms of mass and large in terms of decay heat. Pu is consumed throughout the reactor lifetime (64-73%).

The 102 cm core loads a larger total amount of actinides due to its larger required makeup, and also consumes a larger mass of actinides over the same amount of time. 66 and 62 percent of the total TRU introduced to the ALMR is consumed over 50 years in the 102 cm and 135 cm high cores, respectively (3.3 and 3.1 MT/reactor). 68 and 63 percent of the MA introduced are consumed in the two cores, respectively (385 and 350 kg/reactor). These values show pure consumption without the additional benefit of storing TRU or MA in the ALMR. A total of 560 kg of MA are loaded into one of these cores, and 5 MT of TRU are loaded into these reactors.

Figure 5 gives curves that demonstrate the storage and consumption capability of these cores. The 102 cm core does both load and consume more TRU than the 135 cm core over 50 years. The crossover point where the additional makeup required by the 102 cm core is greater than the greater TRU loaded into the 135 cm core is at 22 years. The lower conversion ratio of the 102 cm core leads to greater consumption amounts for the lifetime of the 102 cm core.

Figure 6 shows TRU consumption rate results from two cores, the 135 cm actinide burning core and the 102 cm actinide burning core. The cores show

TABLE III. TOTAL TRANSURANICS CONSUMED IN 50 YEARS

Isotope	102 cm			135 cm		
	Total Loaded (Kg)	Total Consumed (Kg)	Percent Consumed	Total Loaded (Kg)	Total Consumed (Kg)	Percent Consumed
Np-237	193	168	86.7	168	141	83.9
U-238	9376	4997	53.3	11569	5372	46.4
Pu-238	114	55	48.6	93	32	34.4
Pu-239	2471	1802	73	2211	1342	60.7
Pu-240	1293	741	57.3	1103	507	45.9
Pu-241	250	177	71	216	145	66.9
Pu-242	262	132	50.3	218	96	43.9
Am-241	319	254	79.4	271	201	74.1
Am-242M	4	-1	--	4	-2	--
Am-243	66	10	16.3	53	3	4.8
Cm-242	0	-28	108.2	0	-19	--
Cm-243	0	0	--	0	0	--
Cm-244	5	-15	--	5	-12	--
Cm-245	3	-2	--	2	-2	--
Cm-246	0	-1	--	0	0	--
Total MA	566	385	68.0	501	308	61.4
Total TRU	4956	3293	66.4	4342	2429	55.9
TOTAL HM	14332	8290	57.8	15912	7801	49.0

parallel curves, but different consumption rates. The sharp changes in the first ten years are due to the burnup of Np-237. As the percentage of Np-237 (and other actinides) in the core is high relative to U-238, the burnout of Np-237 will have a larger effect on the rate of change of consumption in the early cycles.

Figure 6 shows an interesting view of actinide consumption. The consumption rate of minor actinides changes throughout the core lifetime. The first two cycles use LWR TRU for reload fuel. The following cycles use processed ALMR fuel and LWR TRU makeup. The marked change in the graphs is the point recycle begins. The rate of actinide consumption is changing due to the changing isotopic composition of the core. Also, the majority of the actinides loaded into the core are from the initial core. As the actinides are processed and reloaded, there is a higher probability of burnup due to the greater amount of core exposure. TRU consumption is approximately level, with some small changes due to changing composition.

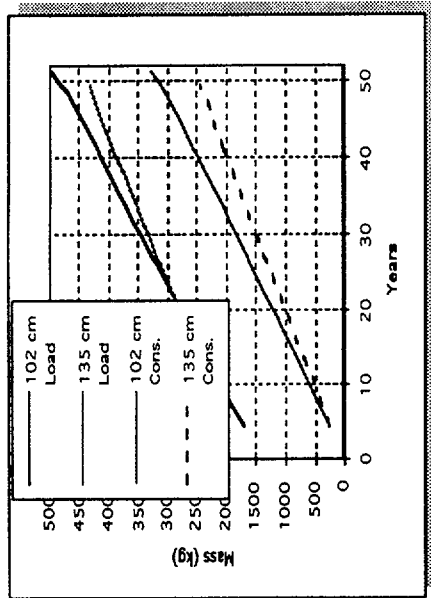


FIG. 5. TRU Storage

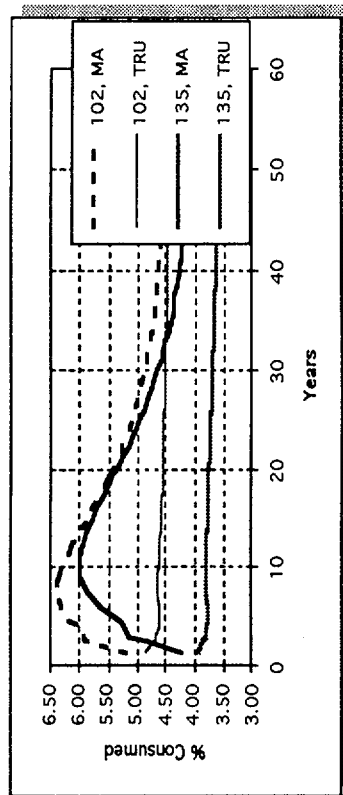


FIG. 6. TRU and MA Consumption Rates

5. IMPACTS OF ACTINIDE BURNING ON THE ALMR SYSTEM

LWR actinide recycle in the current ALMR reference configuration will have a small impact on reactor operation. The greatest impact would be the additional shielding requirements necessary for the greater amount of MA. Actinide recycle using a homogeneous actinide burner core does have system impacts. New safety designs would be needed to accommodate the much greater reactivity swings (e.g., 9% for the 135 cm core and 12% for the 89 cm core).

The greatest impact on the reactor relates to shielding of near core components. This is due to the difference in core assembly type. The reference

heterogeneous core has a row of radial blanket assemblies between the fuel and the reflector assemblies, while the homogeneous cores use only fuel assemblies. Neutrons are produced in the outer ring of fuel assemblies at a higher rate than in the radial blankets in the reference heterogeneous core. It is necessary to increase shielding for these homogeneous cores by converting a possible fuel assembly row into shielding. Near core shielding would need to be increased by a factor of 1.5-2.5 in order to counter the neutron damage incurred without the extra row of shield assemblies.

Actinide recycle impacts fuel handling equipment design because of the need to handle fuel assemblies with higher decay heat and radioactivity. This is due to higher concentrations of minor actinides for reference ALMR startup and burner ALMR reload fuel than for ALMR reference equilibrium fuel. It is anticipated that fuel handling equipment including transport casks can accommodate these higher decay heat and radioactivity levels without major design modifications. Total core decay heat is less than a startup core, so there are no impact on RVACS.

The impacts of actinide recycle on fuel processing and fabrication are expected to be relatively inconsequential because these functions are designed to be operated and maintained remotely. Remote/automated operation and maintenance of fuel processing and fabrication are required due to high radiation levels of plutonium (Pu238 and Pu240) as well as significant levels of fission products (primarily rare earth isotopes). Increased levels of minor actinides should not require additional shielding, with the possible exception of transition areas, such as loading and unloading casks and fuel assembly transfers. It will be necessary to evaluate process impacts, such as the effect on melting point temperatures, process heat loads, volatility, alloying, and other attributes which could affect the processes and/or the product. These impacts are not presently known but, based on the characteristics of the minor actinides, are expected to be minimal and controllable.

For the homogeneous burner cores, there will be no blanket assembly fabrication, as all assemblies are fuel assemblies. Even this change has no major impact on the fuel cycle facilities because for the pyroprocess, blanket assemblies were assumed to be fabricated in the (plutonium) fuel cycle facility and not in a separate facility. Two fuel enrichments are required in the core which will result in a minor complication in fuel processing and fabrication compared to only one enrichment.

6. CONCLUSIONS

Overall, actinide recycle (consumption) appears promising as a waste management approach as well as providing fuel for ALMR startup and deployment for power production. Processing spent fuel provides an opportunity for removal of the actinides and selected fission products from the waste streams as well as potentially improved waste forms. Actinide burning cores designed for safety as well as consumption are certainly feasible within the ALMR design envelope. The ALMR is very flexible and well positioned to provide actinide recycle in both modes, breeder and burner. Recycle of fuel is a natural characteristic of the ALMR.

ACKNOWLEDGEMENTS

Work performed under DOE contract No. DE-ACO3-89SF17445.

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MINOR ACTINIDE CONTAINING FUELS FOR TRANSMUTATION PURPOSES

L. KOCH, M. COQUERELLE, K. RICHTER
Institute for Transuranium Elements,
Commission of the European Communities,
Joint Research Centre,
Karlsruhe

Abstract

In the last 20 years the Institute for Transuranium Elements is developing MA containing fuels for fast reactors. There are several incentives to recycle these materials in fast reactors: increased energy generation, prolonged burn-up reactivity, last but not least reduced radiotoxicity of discharged waste. The basic knowledge to design fuels on an oxide and alloy basis has been obtained from experimental studies and simulation experiments. Fuel pins of different MA compositions have been and will be irradiated in PHENIX. First results of the post-irradiation examinations are given.

1. INTRODUCTION

During the last twenty years the European Institute for Transuranium Elements studied the possibilities to recycle minor actinides to fission reactors [1]. There is an economic incentive to recover these elements for energy generation in the driver fuel of a fast reactor. The gain in extra electricity compensates the additional costs of reprocessing and fuel make-up [1]. They have been assessed to be 4.8% [2] to 5.2% [3] of the electricity generating cost in case of the less favourable PWR. The gain in extra energy by MA fission will amount to about 4% of the energy generation [1] and consequently increase the breeding gain as well as prolong the burn-up reactivity of the fuel [4, 5]. Moreover the recycling of MA would save 15% of uranium - an argument less relevant to FBR's because of more efficient use of uranium in this reactor type than in LWR. Recently the aspect of the long-term reduction of radiotoxicity as an ecological advantage of MA recycling attracts more attention than the economic benefit since the electricity generating cost of a FBR can not yet compete with those of a LWR.

2. FUEL DESIGN

The renewed interest in partitioning and transmutation brought concepts other than the fast reactor into discussion, which are summarised in [6]. The fuel types proposed enclose aqueous solutions, slurries, molten salt alloys, nitrides and, last but not least, oxides. Principally our institute is open to engage in any fuel development provided the appropriate funding is given. Presently we continue with our studies on mixed oxides [7] and Zr-based alloys [8] and will look into the design of Tc targets and minor actinides in inert matrix [9].

The research of mixed oxide fuels will not be limited to an application in a FBR. Since presently the LWR is the nuclear reactor of choice transmutation studies with this reactor type offer the opportunity to study the impact of P & T on an existing fuel cycle and at the same time keep the relevant technology alive for a possible application of P & T in a more effective FR economy.

To design a fuel for specified irradiation conditions certain physical and physico-chemical data have to be known [10]. They include data on:

- effective neutron-cross sections to estimate the burn-up evolution in the fuel with time,
 - mechanical properties such as Young's module, thermal creep plastic deformation,
 - chemical-physical data such as phase diagrams, melting point, oxygen potential, thermal expansion, heat capacity, vapour pressure.
- From such data possible fuel compositions can be derived which need further testing of their technical feasibility.

3. FUEL TESTING

The scope of information required to license an irradiation experiment varies with the chosen reactor and its license authority. It depends on fuel mass to be tested and the computer model used to simulate the irradiation behaviour.

Certain features of a fuel under irradiation can be simulated in a laboratory:

- fuel restructuring in a thermal gradient by electrical heating,
- fuel-clad interactions,
- compatibility of fuel with coolant.

Fuel-swelling, radiation induced creep as well as other radiation dependent fuel parameters can be determined only in an irradiation experiment. Therefore - if their influence to the irradiation behaviour can not be sufficiently predicted - a pre-test with a smaller quantity - (maybe in a different reactor) - will be asked by safety authorities. Such an example is the below mentioned irradiation experiment in KNK II, where a pre-irradiation examination was made in FR-2.

During post-irradiation examination the irradiation behaviour is revealed. Routinely the following tests are foreseen in our institute:

- visual inspection of outer and inner cladding, X-ray examination,
- profilometry, Eddy current, gamma and neutron scanning,
- optical and scanning electron microscopy, EMPA,
- fuel density, porosity, metallography, ceramography, oxygen potential,
- elemental and isotopic assay by IDA-MS, laser ablation ICP-MS.

4. EXAMPLES OF MA-FUELS

Table I summarises the fuel types that have been tested under irradiation, of which the post-irradiation examinations of some of them have not yet been concluded. They all consist of mixed oxides. The basic data needed for fuel design have been presented earlier [10]. First results of the post-irradiation examinations were published [11]. From these and recent results it can be concluded that the MA-containing fuel of the homogeneous type doesn't behave significantly different from the standard fuel under irradiation. Hence a long-term irradiation experiment of mixed oxide in a cooperation with CEA, France is foreseen in PHENIX reactor.

In the test-irradiation of an $(U_{0.5}Am_{0.5})O_2$ in the thermal flux of FR-2 it became apparent that the strong resonance shielding could not be sufficiently accurately predicted. During irradiation only 15% of the expected linear power increase was observed due to the strong radial gradient of Am transmutation in the pellets (Fig. 1) [12]. Consequently the strong temperature gradient leads to circumferential crack formation, what made the outer region of the pellet to decay

TABLE I. MA CONTAINING FUELS TESTED UNDER IRRADIATION

Fuel composition	Reactor	Burn-up
(Am _{0.5} U _{0.5})O ₂	FR-2	<1%
(U _{0.73} Pu _{0.25} Np _{0.02})O ₂	KNK II	1%
(U _{0.73} Pu _{0.25} Am _{0.02})O ₂	KNK II	1%
(Am _{0.5} U _{0.5})O ₂	KNK II	1%
(U _{0.74} Pu _{0.24} Np _{0.02} O _{1.97})	PHENIX	6.4%
(U _{0.74} Pu _{0.24} Am _{0.02} O _{1.96})	PHENIX	6.4%
(U _{0.55} Np _{0.45})O ₂	PHENIX	4.5%
(U _{0.6} Np _{0.2} Am _{0.2})O _{1.93}	PHENIX	4.5%

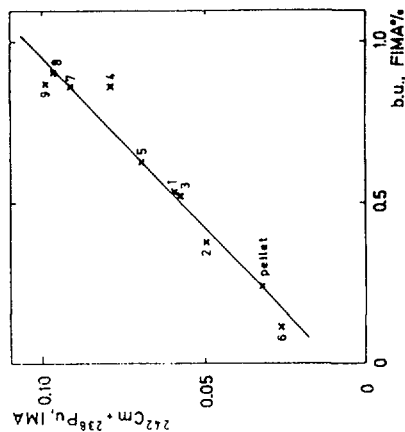


FIG. 1. Range of burn-up and ²⁴²Cm formation (including its daughter product) along the radius of a (U_{0.5}Am_{0.5})O₂ pellet irradiated in FR-2.

into pieces. From this results it appears doubtful if fuels of such a high ²⁴¹Am content can be irradiated in a LWR for the purpose of transmutation.

The burn-up achieved in the KNK II irradiation experiment was low - as given in Table I - hence conclusion on the long-term material irradiation behaviour can not be drawn. So far no irregularities were observed. The main emphasis was given in comparing the experimentally obtained isotopic composition with the predicted one, which has been presented in [13]. The agreement for most of the nuclides is sufficient (Table II).

TABLE II. RATIO OF CALCULATED TO EXPERIMENTAL ISOTOPE CONCENTRATIONS FOR (U_{0.75}Pu_{0.25}Am_{0.02})O₂

Isotope	JENDL. 3 SAND(DK8)	ENDF/B V(IV) SAND(DK8)	KFKIRUZJEP-2 SAND(DK8)	KFKIRUZJEP-2 DJEP(DZFI)	KFKIRUZJEP-2 SAND(DK2)
²³⁸ Pu	1.2593	1.2137	0.9762	0.8345	1.2397
²³⁹ Pu	0.9865	1.0023	1.0029	1.0280	0.9718
²⁴⁰ Pu	1.0371	1.0343	1.0024	0.9933	1.0169
²⁴¹ Pu	1.1843	1.0467	1.1168	1.0058	1.2602
²⁴² Pu	1.1107	1.0742	1.0604	0.9878	1.1821
²⁴¹ Am	1.0133	1.0176	1.0529	1.0639	1.0294
^{242m} Am	1.4532	1.3674	1.0404	0.9078	1.2718

(Neutronspectrum obtained by Fluxmonitors + SAND programme or by 3-dimensional diffusion calculations. 3 different cross-section libraries were applied.)

TABLE III. FABRICATION DATA OF MA FUEL TO BE IRRADIATED IN PHENIX

Fuel compositions	(U _{0.74} Pu _{0.24} Np _{0.02})O ₂	(U _{0.74} Pu _{0.24} Am _{0.02})O ₂	(U _{0.55} Np _{0.45})O ₂	(U _{0.6} Np _{0.2} Am _{0.2})O ₂
thermal treatment of GSP particles	400°C/2h argon 400°C/2h air 850°C/2h air 850°C/2h Ar+5% H ₂	400°C/2h argon 400°C/2h air 900°C/4h air 1050°C/2h Ar+5% H ₂	400°C/1h argon 400°C/4h air 865°C/3h air 865°C/4h Ar+5% H ₂	400°C/1h argon 400°C/4h air 900°C/2.5h air 900°C/3h Ar+5% H ₂
pressing	5.1-6.4 U/cm ²	4 + 5.6 U/cm ²	6.1 U/cm ²	6.1 U/cm ²
free density	58.6% TD	58.0% TD	55.6% TD	56.4% TD
sintering	1620°C/6h Ar+5% H ₂	1620°C/6h Ar+5% H ₂	1620°C/6h Ar+5% H ₂	1620°C/6h Ar+5% H ₂
pellet Ø	5.363 ± 0.052 mm	5.417 ± 0.029 mm	5.518 ± 0.027 mm	5.434 ± 0.013 mm
pellet density	97.5 ± 0.8% TD	96.8 ± 0.7% TD	95.1 ± 0.6% TD	95.9 ± 1.0% TD
O/M	1.973	1.957	1.996	1.928
X ray diffraction parameter	5.4578 ± 5	5.4570 ± 5	5.4545 ± 5	5.4735 ± 5 5.4696 ± 5

The fabrication data of fuels irradiated in PHENIX are summarised in Table III [14]. The limiting parameter of the oxide fuels is their oxygen potential, which has to be adjusted (by sintering in argon atmosphere containing 5% H₂ to such a level that a reaction with the Na coolant is minimised.) Since a low O/M ratio lowers the thermal conductivity too, the irradiation of a (U_{0.5}Am_{0.5})O_{2-x} becomes impossible. The other fuel parameters are close to those of the standard fuel. Worth to mention is that the gel-supported-precipitation (GSP) was used to avoid any aerosol formation as observed by using mixed oxide powders. The fuel specimen reached a linear power in the range of 300 to 400 W/cm. As pointed out earlier the

TABLE IV. SUMMARY OF THE MICROSTRUCTURE ANALYSIS OF SPENT MA FUELS IRRADIATED IN PHENIX

Pin	(U _{0.74} Pu _{0.24} Np _{0.02})O ₂ 380/325 W/cm	(U _{0.6} Am _{0.2} Np _{0.2})O ₂ 174/273 W/cm	(U _{0.55} Np _{0.45})O ₂ 206/283 W/cm	(U _{0.74} Pu _{0.24} Am _{0.2})O ₂ 380/325 W/cm
Cladding				
r _c [mm]	3.294	3.293	3.303	3.293
Δ r/r [%]	0.58	0.55	0.84	0.55
wall thickness [mm]	0.448	0.449	0.454	0.446
Fuel-cladding interaction				
Δ r [μm]	19	3	3	19
Fuel				
r fuel [mm]	2.829	2.840	2.841	2.826
Δ r/r [%]	4.1	5.9	4.9	4.3
Fuel restructuring				
Zone 1: outer ring (r/r ₀)	1-0.66	1-0.66	1-0.58	1-0.67
Zone 2: equiaxial grain growth	0.66-0.50	0.55-0.26	0.58-0.22	0.67-0.48
Zone 3: columnar or elongated grain growth	0.50-0.11	0.26-0.00	0.22-0.00	0.48-0.11
Zone 4: central void	0.11-0.00	0.98, 0.63		0.11-0.00
circular crack (position r/r ₀)	0.83			0.89
Secondary phases				
metallic precipitates: (Ø = 1 μm)	Zone 1 - 3	Zone 1 - 3	Zone 1 - 3	Zone 1 - 3
(Ø = 5 μm)	Zone 3			Zone 3
ceramic phase: (Ø = 1 μm)	Zone 3		Zone 2	
(Ø = 1 μm)				

behaviour did not deviate strongly from the standard-mixed oxide except for the He formation stemming from ²⁴²Cm decay and leading to a higher gas pressure inside the pin. The recent results of the post-irradiation examinations are summarised in Table IV [15]. The fuels with the high MA concentrations did not reach the same linear power and consequently were exposed to lower burn-up. This explains the difference to the two other fuels, which are more similar to the standard fuel. Further results on fuel-clad-interaction and isotopic composition (in comparison to predictions) will become available in the future.

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PHYSICS CONSIDERATIONS IN THE DESIGN OF LIQUID METAL REACTORS FOR TRANSURANIC ELEMENT CONSUMPTION

H. KHALIL, R. HILL, E. FUJITA, D. WADE
Argonne National Laboratory,
Argonne, Illinois,
United States of America

Abstract

The management of transuranic nuclides in liquid metal reactors (LMR's) is considered based on the use of the Integral Fast Reactor (IFR) concept. Unique features of the IFR fuel cycle with respect to transuranic management are identified. These features are exploited together with the hard spectrum of LMR's to demonstrate the neutronic feasibility of a wide range of transuranic management options ranging from efficient breeding to pure consumption. Core physics aspects of the development of a low sodium void worth transuranic burner concept are described. Neutronics performance parameters and reactivity feedback characteristics estimated for this core concept are presented.

1. INTRODUCTION

The desire to reduce the long term radiological hazard associated with the disposal of spent nuclear fuel has motivated numerous studies in several countries (e.g. Refs. 1-6) of ways to destroy the transuranium products of fuel irradiation. Recent efforts in the U.S. at Argonne National Laboratory [6-8] have focused on evaluating the potential for accomplishing this with liquid metal reactors (LMR's), particularly metal fuel systems based on the Integral Fast Reactor (IFR) concept. These evaluations have addressed both the recycle of self-generated actinides and the consumption of externally produced inventories of transuranics, e.g. the transuranic species in spent LWR fuel. Core physics aspects of these evaluations will be discussed in this paper.

2. THE IFR CONCEPT

Key features of the IFR concept [9] include the metallic fuel form (U-Pu-Zr) and the compact and inexpensive techniques being developed for fuel reprocessing (pyrometallurgical process) and refabrication (injection casting). Neutronic feedback mechanisms and other inherent physical phenomena provide for neutronic shutdown and decay heat removal in accident sequences, resulting in a high degree of passive safety. Fuel burnup levels in excess of 15 at% are being demonstrated for the U-Pu-Zr alloy in EBR-II experiments. Current designs [10] are self-sufficient with respect to fissile mass, providing for favorable fuel cycle economics, and can be modified to yield a large net excess fissile production if desired. Conversely, favorable neutron economy can be sacrificed by reduction of fertile content and core geometric "spoiling" to permit the net consumption of transuranic species instead of their net creation through breeding.

The IFR fuel cycle has a number of attractive features in connection with the management of man-made actinides. They stem from the following:

1. The main reprocessing step (electrorefining) directly provides for the separation of the bulk of the transuranics from uranium and fission products. The small portion (~1%) of transuranics

that is not automatically recycled can, with techniques being developed, be stripped from the initial waste and returned to the reactor at the expense of simultaneously recycling an increased proportion of the rare-earth fission products.

2. The higher radioactivity levels associated with the increased fission product carryover to the transuranic product stream does not significantly complicate fuel refabrication, which is done remotely because of the presence of higher actinides in the repeatedly recycled fuel and the incomplete fission product separation inherent in the basic reprocessing method.
3. The hard neutron spectrum favors destruction of transuranics by fission over creation of higher transuranics via neutron capture. Thus, the relative concentrations of minor actinides do not build up to levels at which their radioactivity becomes intolerable. The spectrum hardness also mitigates the adverse poisoning effect of the recycled portion of the rare-earth fission products.

3. RECYCLE OF LWR SPENT FUEL

In addition to the main IFR reprocessing and waste treatment steps (i.e. those concerned with recycle of LMR discharge fuel and blanket assemblies), pyrochemical processes are also being developed and demonstrated for the purpose of extracting the transuranic species from LWR spent oxide fuel and for concentrating these transuranics in a metallic form suitable for introduction into the IFR fuel cycle. The goal of the process development is to recover at least 99.9% of the LWR discharge transuranics for use in LMR's. The processes being studied involve the decladding of the spent LWR fuel pins by suitable mechanical and chemical means, and the subsequent pyrochemical decomposition of the spent fuel into (a) a product stream containing the transuranics and the major portion of rare-earth fission products, (b) a uranium-rich component suitable for storage and potential future use as the source of LWR or LMR fuel, and (c) waste streams that can be processed to recover residual actinides and then converted and packaged into forms acceptable for geologic disposal.

It should be noted that the elimination of fission products from the transuranic output of the above processes is not required, because this output is designed for introduction into the IFR electrorefining step, which accomplishes the requisite degree of fission product removal. The high radioactivity and low purity of the transuranic product limit both the risk of its diversion and its attractiveness for weapons applications. On the other hand, the effected separation of the bulk of the uranium is important because it helps preserve the compactness of the electrorefining process by allowing this process to deal with only about 1 to 2% of the total heavy metal in the spent fuel.

Currently, three different pyrochemical separation concepts, referred to as the "salt transport", "magnesium extraction", and "zinc-magnesium" processes, are being investigated with the objective of identifying and further developing the most promising concept.

4. TRANSURANIC MANAGEMENT IN SELF-SUFFICIENT LMR'S

In a self-sufficient LMR, transuranic losses by fission are compensated by transuranic (Pu-239) breeding, allowing for sustained power production with only fertile material (e.g. depleted U) supplied as makeup. Table 1 compares the transuranium isotopic mix of discharged fuel from a LWR (once-through) and a 1200 MWt LMR (equilibrium/recycle, based on the IFR concept). The relative concentrations of the vast majority of minor actinides are substantially smaller in the LMR discharge, while the Pu-239 proportion therein is greater. Thus, a self-sufficient LMR using the recycled transuranics from LWR discharge as its startup fissile source would, in addition to producing power over its operating life, result in an equilibrium fuel composition with reduced long-term radiological toxicity owing to the reduced proportion of higher actinides. Of particular importance is the reduced

Table I. COMPARISON OF LWR AND LMR TRANSURANIC ISOTOPICS

Fraction of Transuranic Mass	LWR		1200 MWt LMR		1200 MWt LMR		Ratio of LWR (3.2 y Decay) to LMR (2.0 y Decay)	
	Discharge (3.2 y Decay)	LWR (3.2 y Decay)	Discharge	LWR	Discharge	LWR	(2.0 y Decay)	(2.0 y Decay)
Np-237	5.35-2	5.40-2	6.16-3	6.19-3	6.19-3	6.19-3	8.7	
Pu-236	2.41-7	1.12-7	6.12-8	4.08-8	4.08-8	4.08-8	2.7	
Pu-238	9.08-3	1.01-2	6.27-3	6.19-3	6.19-3	6.19-3	1.6	
Pu-239	0.503	0.508	0.747	0.747	0.747	0.747	0.68	
Pu-240	0.201	0.199	0.200	0.200	0.200	0.200	1.0	
Pu-241	0.157	0.134	2.13-2	1.96-2	1.96-2	1.96-2	6.8	
Pu-242	3.93-2	3.88-2	8.49-3	8.49-3	8.49-3	8.49-3	4.6	
Am-241	3.44-3	2.51-2	6.94-3	8.63-3	8.63-3	8.63-3	2.9	
Am-242m	1.14-4	1.11-4	5.11-4	5.07-4	5.07-4	5.07-4	0.22	
Am-243	2.51-2	2.48-2	1.74-3	1.74-3	1.74-3	1.74-3	14.0	
Cm-242	1.31-3	9.73-6	3.56-4	2.36-5	2.36-5	2.36-5	0.41	
Cm-243	8.55-5	7.86-5	1.93-5	1.85-5	1.85-5	1.85-5	4.2	
Cm-244	6.32-3	5.52-3	7.96-4	7.44-4	7.44-4	7.44-4	7.4	
Cm-245	5.15-4	5.08-4	1.63-4	1.63-4	1.63-4	1.63-4	3.1	
Cm-246	6.42-5	6.31-5	4.72-5	4.72-5	4.72-5	4.72-5	1.3	
MA/Fissile Pu	0.137	0.172	0.022	0.024	0.024	0.024	7.2	
MA/Total Pu	0.099	0.124	0.017	0.018	0.018	0.018	6.9	
Np-237/MA	0.591	0.490	0.368	0.343	0.343	0.343	1.4	
Am-241/MA	0.038	0.228	0.415	0.478	0.478	0.478	0.48	
Am-243/MA	0.278	0.225	0.104	0.096	0.096	0.096	2.3	
Np chain	0.214	0.213	0.034	0.034	0.034	0.034	6.3	

MA = sum of minor actinides

Fissile Pu = Pu-239 + Pu-241

Np chain = Np-237 + Am-241 + Pu-241

fraction of Pu-241, whose decay product (Np-237) dominates the long-term hazard. The energy production and long-term waste radiotoxicity reduction benefits are realized at the costs associated with the reprocessing of LWR fuel and the increased amount of fission product waste.

5. TRANSURANIC CONSUMPTION IN PURE BURNER CONCEPTS

Characteristics of LMR concepts designed for net consumption of pre-existing transuranic inventories have also been investigated. Taken to an extreme, no fertile U-238 is utilized in these concepts, and only transuranics are used for the startup inventory and as the source of makeup material added to the actinides recovered from self-discharged fuel. The neutronic feasibility of such "pure burner" concepts, which maximize the transuranic consumption per unit of energy production, has been verified in 1200 MWt LMR's -- both for the case in which all transuranic species in the LWR discharge are used as fuel and for the case in which only the minor actinide species (no Pu) are used.

Table II. COMPARISON OF EQUILIBRIUM CYCLE CHARACTERISTICS FOR 1200 MWt ACTINIDE RECYCLE CONCEPTS

	Fissile Self-Sufficient LMR		Pu and MA Pure Burner		MA Pure Burner
No. of Batches	4		1		1
Cycle Length, days	365		254		453
Capacity Factor, %	89		80		80
Driver Average Burnup, MWd/kg	104		111		111
Driver Peak Burnup, MWd/kg	140		150		158
Driver Peak Fast Fluence, $10^{21}n/cm^2$	3.51		1.30		1.76
Driver Peak Linear Power, kW/ft	13.0		13.6		14.5
EOC-BOC Δk Swing, %	0.27		-12.1		-4.33
Driver HM Loading, kg/y	2770		4040		3040
%U	77.2		0		0
%Pu	22.4		64.6		37.5
%MA	0.4		35.4		62.5
Blanket HM Loading, kg/y	3910		-		-
%U	100		-		-
%Pu	0		-		-
%MA	0		-		-
Makeup Feed, kg/y	466		352		346
%U	100		0		0
%Pu	0		87.6		0
%MA	0		12.4		100

HM = Heavy Metal

MA = Minor Actinides

Illustrative design and performance characteristics for both cases are summarized in Table II. It should be emphasized that the fuel compositions being employed in such cases are quite exotic in containing essentially no uranium, and that the assembly/core configurations using such compositions (and constrained by a beginning of cycle excess reactivity requirement) are nonconventional. Thus while these concepts are feasible neutronicallly, the establishment of their overall viability requires extensive technological efforts, a discussion of which is beyond the scope of this paper.

The consumption of transuranics in LWR's is considerably less attractive. Not only does the thermal spectrum favor the buildup (in proportion) of the higher actinides, but it also renders most of

the minor actinide species as poisons whose accommodation requires greater fissile enrichment; a thermal system fueled entirely with the LWR-discharge isotopic mix of minor actinides is not feasible neutronically based on the fundamental requirement of criticality. Moreover, the technological difficulties of adapting current aqueous reprocessing methods to compositions rich in higher actinides are likely to be severe.

6. CONCEPTUAL DESIGN FOR TRANSURANIC CONSUMPTION

The focus of recent conceptual design efforts at ANL [7] has been the development of core concepts for net transuranic consumption whose design parameters, unlike those of the pure burner concepts, remain within the bounds of the in-place development program. For example, limits on transuranic enrichment (28 wt% in heavy metal), linear power (50 kW/m), discharge burnup (15 at%), and peak fast fluence (3.5×10^{22} n.cm⁻²) were observed. A key additional objective of the core development was to achieve a low (near-zero) sodium void worth. Finally, the core concepts were developed so that net fissile breeding, if desired, could be readily achieved as an alternative to net transuranic consumption. Such flexibility for increased breeding was achieved by use of external blankets to minimize the effect on reactor design and to mitigate the inevitable increase in sodium void worth that would occur with enhanced internal conversion.

A 1575 MWt (600 MWe) design concept satisfying these objectives and constraints was developed [7]. Key features of this concept, in its transuranic consumption mode, are (a) elimination of internal and external blankets to minimize transuranic production by U-238 capture, (b) adoption of a pancaked core shape to reduce breeding and void worth, and (c) incorporation of a non-fueled central region in an annular core configuration. Detailed evaluations of this core configuration were carried out based on the use of LWR-discharge transuranium isotopics as the reactive fuel feed component; limited analyses of this core with its equilibrium (infinite-recycle) fuel composition have also been performed but are not described here in detail. The use of a flat, annular core shape enabled the achievement of a low void worth (\$0.16 for EOC voiding of the entire core and upper plenum region) while simultaneously satisfying the assumed 28% transuranic enrichment limit. In particular, the annular geometry mitigates the central power peaking tendency without recourse to enrichment zoning, thereby enabling the enrichment and linear power limits to be satisfied concurrently with meeting the criteria of transuranic consumption and near-zero void worth.

The breeding ratio of 0.53 permits the net consumption of 234 kg of LWR transuranics on an annual basis. The fissile Pu component of this net loss (218 kg/y) would, with repeated recycle, decrease somewhat as the nuclides that are less likely to fission increase in proportion. Thus some changes in core performance would be observed with repeated recycle as the transuranic isotopic mix gradually shifts to an equilibrium recycle distribution characteristic of the core geometry and fuel cycle parameters. Preliminary calculations suggest that the coolant void worth of the equilibrium core is nearly \$1.0 greater.

Additional performance results are summarized in Tables III and IV. Of particular note are the burnup reactivity loss of 4.2% Δk (5.7% Δk without midcycle replacement of central absorber assemblies), which is significantly greater than the nominally zero reactivity swing achievable in fissile self-sufficient designs. The small Doppler coefficient can be attributed to the hard spectrum and the low U-238 concentration; contributions to the Doppler effect by the transuranics are minimal

Table III. EQUILIBRIUM-CYCLE PERFORMANCE PARAMETERS

Breeding Ratio	0.533
EOC-BOC Reactivity Swing, % Δk	-4.17
Average Discharge Burnup	
MWd/kg	82.6
Atom %	8.8
Peak Discharge Burnup	
MWd/kg	118.6
Atom %	12.6
Peak Linear Power, W/cm	
BOC	495
EOC	462
Power Peaking Factor	
BOC	1.64
EOC	1.53
Peak Flux, 10 ¹⁶ cm ⁻² s ⁻¹	
BOC	4.84
EOC	5.02
Peak Fast Flux, 10 ¹⁶ cm ⁻² s ⁻¹	
BOC	3.68
EOC	3.77
Peak Fast Fluence, 10 ²² cm ⁻²	
Heavy Metal	5,790
Transuranics	1,492
Fissile Pu	958
Net Loss, kg/y	
Heavy Metal	508
Transuranics	234
Fissile Pu	218

because of their presence as dilute species in the reprocessed LWR discharge composition. Finally, the flat core shape results in the radial expansion reactivity coefficient being significantly more negative, and the axial expansion coefficient being less negative, than in conventional designs. For the same reason, and because of the higher control rod worth dictated by the greater burnup reactivity loss, the control rod driveline expansion coefficient is much more negative than for conventional cores. Dynamics analyses [11] utilizing the computed feedback parameters have demonstrated significant passive safety margins for unprotected loss-of-flow and reactivity insertion accidents.

Table IV. LOW VOID WORTH ACTINIDE BURNER REACTIVITY COEFFICIENTS

	BOC*	EOC
Sodium Void Worth, \$		
Core	1.78	2.85
Plenum	-2.20	-2.70
Total	-0.43	0.16
Doppler Coefficient, $-10^{-3}\Delta k/dT$		
Flooded Core		
Fuel Structure ^b	0.73	0.88
Voided Core	0.30	0.35
Fuel Structure ^b	0.36	0.46
	0.21	0.25
Axial Expansion Coefficient, \$/cm		
Fuel	-1.50	-1.19
Fuel and Structure ^b	-1.37	-1.01
Radial Expansion Coefficient, \$/cm	-1.43	-1.48
Control Rod Drive Line		
Expansion Coefficient, \$/cm	-0.778	-0.414
Effective Delayed Neutron Fraction	3.50E-3	3.47E-3
Prompt Neutron Lifetime, s	2.17E-7	2.47E-7

*BOC values are calculated for critical configuration, with primary rods inserted 24 cm.

^bValues reflect perturbation of total structure; clad effects can be estimated as 63% (volume fraction of clad in the total structure) of the total structure effect.

7. SUMMARY

The IFR fuel cycle has a number of attractive features for the management of transuranics. These features, when successfully demonstrated, can be exploited along with the hard neutron spectrum characteristic of LMR's to achieve substantial flexibility in transuranic management options, ranging from efficient breeding to destruction in burner concepts.

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**ENGINEERING ASPECTS OF
TRANSMUTATION USING FAST REACTORS**

(Session 3)

Chairmen

L.A. KOCHETKOV
Russian Federation

H. SZTARK
France

RADIOWASTE TRANSMUTATION IN NUCLEAR REACTORS

A.N. SHMELEV, G.G. KULIKOV,
V.A. APSE, V.B. GLEBOV
Moscow Institute for Physics and Engineering
D.F. TSURJKOV, A.G. MOROZOV
I.V. Kurchatov Institute of Atomic Energy
Moscow, Russian Federation

Abstract

The burnup of minor actinides may be realized both in fast and thermal reactors. The principal difference of minor actinides burn-up in thermal and fast reactors concerns with the neutron balance: in thermal reactor the neutron balance is negative with neutron consumption and otherwise in fast reactor the neutron balance is positive with production of excessive neutrons. Applying of fuel containing minor actinides in FBRs leads to degradation of safety characteristics, namely to decreasing of effective fraction of delayed neutrons β_{eff} and to high positive sodium void effect of reactivity to upgrade the value of β_{eff} it is supposed the addition of the fissile nuclides characterized by high fraction of delayed neutrons in fuel composition. The decreasing of sodium void reactivity may be achieved by use of potassium as a coolant (or potassium alloys Na-K, K-Pb).

The two important aspects may be emphasized in the transmutation problem of long-lived radioactive waste (RAW) produced in reprocessing of nuclear reactor spent fuel. The first one is related with the safe burn-up of minor actinides (Np, Am, Cm) determining RAW potential hazard during a thousand years or more (see Fig.1a). The second one concerns with the transmutation of long-lived fission products (FPs), mainly Sr-90 and Cs-137, with duration of significant radiological hazard up to several centuries (see Fig.1a). Here it may be noted the following point. As it is shown at Fig.1a the potential hazard of RAW getting into organs of digestion (IBHwater-Index of Biological Hazard defined as a water volume needed for RAW dilution to safe concentration $1/l$) is determined by the mentioned FPs up to several centuries. During the next thousands of years IBH is determined by the actinides. Potential hazard of RAW inhalation is determined mainly by actinides during all time periods (see Fig.1b).

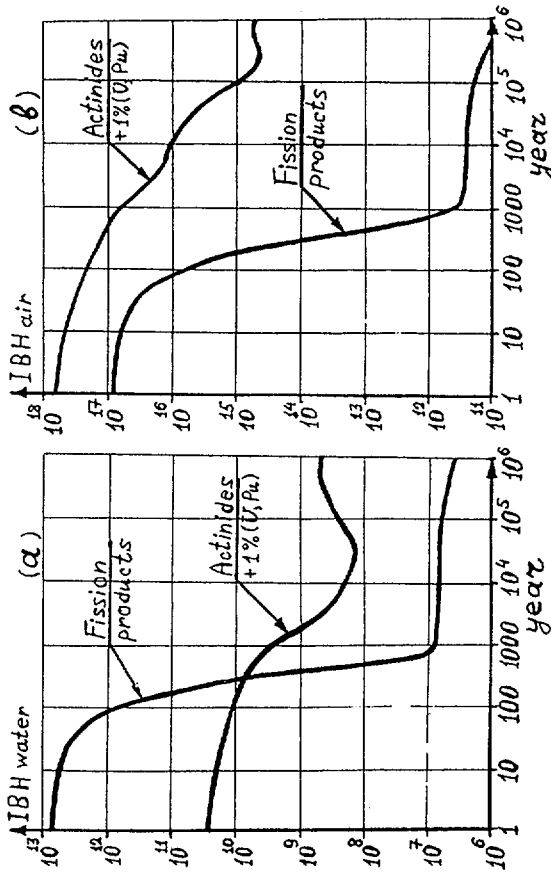


FIG. 1. Index of biological hazard (m^3 water or air/GW(e).year) as a function of waste decay time (PWR type reactor).

Neutron consumption in minor actinides burn-up in fast and thermal reactors.

The burn-up of minor actinides may be realized both in fast and thermal reactors. Fission cross-sections of minor actinides have the thresholds in the 0.1-1.0 MeV energy region, so it can be fissioned in fast reactors. In thermal reactors the minor actinides can be transmuted by neutron capture to the well-fissionable nuclides (Pu-239, Cm-245) with following burn-up. As a result the long-term potential hazard of minor actinides (thousands of years) converts into the short-term hazard of the FPs (hundreds of years).

The principal difference of minor actinides burn-up in thermal and fast reactors concerns with the neutron balance: in thermal reactor the neutron balance is negative with neutron consumption and otherwise in fast reactor the neutron balance is positive with production of excessive neutrons. This difference is demon-

TABLE 1. ACTINIDE BURNING AND NEUTRON BALANCE

	100 ²³⁹ Np-nuclides		100 Actinide nuclides	
	PWR	FBR	PWR	FBR
Number of Act. Fissions	33.1	39.8	35.8	38.0
Number of neutrons	-83.1	+31.0	-91.0	+29.8
Number of ²³⁵ F-fissions	58.5	0	64.1	0
(²³⁵ U ₀)	(24.6)	(0)	(26.9)	(0)
Total Fissions	91.6	39.8	99.9	38.
	(31.1+58.5)		(35.8+64.1)	
Number of Transmutations (S+G+I+T)	0	149.0	0	143.3
	Fissions			
Np-237	0.5	19.9		
Np-238	1.0	0.3		
Pu-238	2.3	18.4		
Pu-239	25.6	1.2		
Pu-241	3.7	-		
Total:	33.1	39.8		

treated by the data of Table 1 where the calculational results of /2/ are presented. During the 1000 days one hundred nuclei of Np-237 and one hundred nuclei of minor actinides from spent fuel of FWR /3/ were irradiated in neutron spectra of FWR-type thermal reactor and fast breeder reactor (FBR). It is shown that during irradiation in FBR spectrum the more actinide fissions occurred than in thermal reactor spectrum. In the spectrum of FBR the main fraction of fissions is related with Np-237 and Pu-238; for thermal reactor spectrum Pu-239 is the main fissile nuclide. Transmutation of 100 Np-237 nuclei in thermal reactor spectrum led to consumption of 83.1 neutrons (including the fission neutrons) but irradiation in FBR spectrum was accompanied by the excessive neutrons production (31 neutrons). For the generation of 83.1 neutrons in thermal reactor it is needed to occur the 58.5 fissions of U-235 accompanied by the conversion of 24.6 U-235 nuclei to U-236, so the total consumption of U-235 is 83.1 nuclei. Neutron

balance of actinides irradiation in FBR is characterized by positive value, the excessive neutrons are produced. Thus, irradiation of 100 Np-237 nuclei in thermal reactor during the 1000 days resulted in fission of 33.1 nuclei of Np-237 and its daughter products, but in sum with U-235 fissions for neutron generation the total fissions quantity is 91.6. Irradiation of 100 Np-237 nuclei in FBR during the 1000 days resulted in 39.8 fissions. So the actinides burn-up in FWRs leads to the generation of increased quantity of FPs (by 2.3 times) and to additional consumption of U-235. Otherwise the analogous process in FBRs is accompanied by the production of excessive 31 neutrons which may be used for the transmutation of long-lived FPs. The calculational results of one hundred minor actinides nuclei irradiation are the same as that for Np-237. It must be noted that following analysis showed the sufficiency of such neutron excess for transmutation of generated long-lived FPs which are expected to be the main hazardous component of RAW. This comparison illustrates the principal difference between fast and thermal reactor. Fast reactor is mainly the breeder of neutrons in conditions of fuel self-sustaining. If the excessive neutrons are absorbed in the fertile material then such reactor becomes the breeder of fuel. If the fuel breeding doesn't need as the main goal of fast reactor operation then these neutrons can be used for the transmutation of long-lived FPs.

Neutron consumption in transmutation of long-lived FPs.

In order to evaluate the neutron consumption in transmutation of the most hazardous long-lived FPs it is necessary to analyse the yields of these nuclides. It is known /4/ that in thermal fission of U-235 the total yield of the main long-lived FPs is approximately 0.205 nuclei per fission (see Table 2). If transmutation of these FPs would begin after the period of 30-year decay (as in / 5 /) then the total quantity of FPs would decrease to 0.142 per fission due to radioactive decay of Sr-90 and Cs-137. Neglecting the value of FPs neutron capture cross-sections and neutron losses it may be estimated that 0.142 neutrons per fission must be provided for these radionuclides transmutation. This

TABLE 2. YIELD OF FISSION PRODUCTS PER FISSION ^{235}U (n_{fp} , f)

Nuclide	$T_{1/2}$, y.	Yield	Yield Φ 30 yr. decay (unit fission)	IBH, $\frac{m}{\text{yr}} \cdot \frac{10^6}{\text{unit fission}}$
^{90}Sr	27.7	0.059	0.029	$1.86 \cdot 10^{11}$
^{137}Cs	30.	0.062	0.031	$4.07 \cdot 10^9$
^{129}I	$1.6 \cdot 10^7$	0.009	0.009	$5.25 \cdot 10^5$
^{99}Tc	$2.1 \cdot 10^5$	0.062	0.062	$6.45 \cdot 10^4$
^{151}Sm	87.	0.0082	0.0064	$7.49 \cdot 10^5$
^{79}Se	$6.5 \cdot 10^6$	0.0008	0.0008	$1.35 \cdot 10^5$
^{107}Pd	$6.5 \cdot 10^6$	0.004	0.004	$3.82 \cdot 10^4$
Total		0.205	0.142	$1.90 \cdot 10^{11}$

*Yield Φ neutron capture in subsequent isotopes.

is the minimal estimation because of the supposition of continuous purification of irradiated nuclides from daughter product and neglectation of neutron losses due to leakage and useless absorption. The demand on neutron quantity needed to transmuted the main long-lived FPs may be compared with rate of neutrons generation in fast reactor used for the production of excessive fissile isotopes in breeding regime. Suppose that FBR with metal fuel has breeding ratio $BR=1.4$ and breeding ratio of core $BRC=1.0$ for the compensation of fuel burn-up reactivity effect. Then the value $BR-1=0.4$ is the characteristics of the excessive fuel production in the reactor blanket. It means that one fission in core leads to generation of 0.4 neutrons in blanket with taking into account of the neutron leakage and useless losses. Comparing this excessive neutron quantity with minimal neutron consumption for transmutation of the main long-lived FPs (0.142 neutrons per fission after 30-year decay period) it may be concluded that for transmutation of the own long-lived FPs in blanket of such FBR the useless losses of 65% neutrons may be allowed. This reserve can be used, for instance, in the optimization of blanket composition, of irradiation regime with periodical purification

and loading the new FPs including FPs from other reactors. It is important to note that quantity of excessive neutrons generated in FBR with mentioned breeding parameters may be sufficient for transmutation of produced long-lived FPs. Now it must be noted that Sr-90 and Cs-137 have low values of neutron capture cross-sections, so the level of neutron flux in FBR blanket is insufficient for the effective transmutation of these nuclides. Long-lived FPs with rather high neutron capture cross-sections can be transmuted in blanket of FBR. The transmutation of Sr-90 and Cs-137 must be realized in high-flux reactor (see Fig.2), its concept will be discussed below. It is important to note that separate transmutation in two nuclear reactors may be characterized by the similar total neutron balance as in considered simplified scheme of long-lived FPs transmutation in Fast Reactor Blanket.

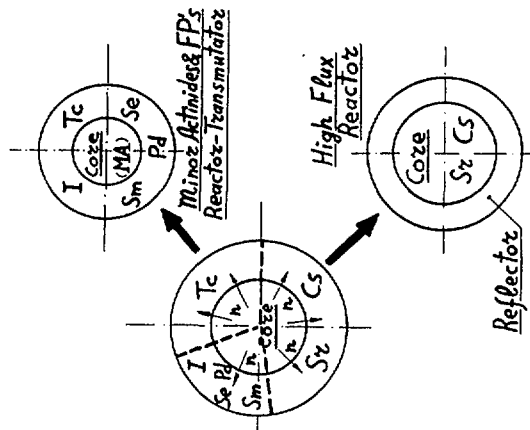


FIG. 2. Transmutation of MAAs and FPs in nuclear reactors.

TABLE 3. VOID EFFECT OF REACTIVITY (LINEAR PERTURBATION THEORY) AND COMPONENTS OF SENSITIVITY

Fuel	Coolant effect of reactivity	Void effect of reactivity (per one nucleus)	Components of sensitivity (per one nucleus)					
			Total	Capture				
(U-Pb) nitride	Na	-0.0198	1.28	-92.23	88.87	-3.36	-0.21	4.86
	Pb	-0.0194	0.91	-34.44	31.10	-3.04	-0.66	4.62
	K	-0.0047	0.61	-24.39	23.04	-1.56	-3.34	5.70
(U+Th) ²³⁵ + minor actinides	Na	+0.0619	-4.11	-34.72	46.55	-8.17	-0.08	4.15
	Pb	+0.0437	2.06	-25.56	20.40	-5.16	-0.36	3.47
	K	-0.00097	0.11	-16.84	14.46	-2.38	-3.33	5.83
75% Na + 25% K + 85% K + 15% Pb	Na	-0.0130	-2.19	-60.97	51.37	-9.61	-0.08	7.5
	Pb	-0.0051	-0.87	-16.72	14.37	-2.35	-3.28	4.76
	K	-0.0051	-0.51	-17.05	14.63	-2.41	-3.24	5.14
	Pb-208	-0.0299	6.82	-30.48	24.43	-6.05	-0.32	13.19

Safety of fast reactor-burner of minor actinides.

Applying of fuel containing minor actinides in FBRs leads to degradation of safety characteristics, namely to decreasing of effective fraction of delayed neutrons β_{eff} and to high positive sodium-void effect of reactivity /6/. to upgrade the value of β_{eff} it is supposed the addition of the fissile nuclides characterized by high fraction of delayed neutrons (U-235, U-233, Th-232) in fuel composition. The decreasing of sodium-void reactivity may be achieved by use of potassium as a coolant (or potassium alloys Na-K, K-Pb). The following coolants in core of fast actinide burner reactor were considered: alloy Na-K, alloy K-Pb and Pb enriched by isotope Pb-208. Table 3 presents the calculational results of void reactivity effect and its components for FBR with (U, Pu)-nitride fuel and with metal fuel containing the minor actinides. Void reactivity effect was evaluated by the formulae of linear perturbation theory. It may be seen that FBR with (U, Pu)-nitride fuel has negative void reactivity effect for any coolants investigated (Na, Pb, K). FBR with metal fuel containing the minor actinides is characterized by the significant positive void reactivity effect for Na and Pb coolants (+18%Bef and +12.8*Bef, respectively). FBR with potassium coolant has small negative void reactivity effect due to decreased density and slowing-down car-

bility. Also negative void reactivity effect was obtained by use of K-Pb alloy as coolant. Significant negative void reactivity effect was achieved for FBR with lead coolant enriched by the isotope Pb-208. It is caused by the high threshold of inelastic neutron scattering of Pb-208 (energy of the first excited level is 2.62 MeV) and small neutron capture cross-section. Application of linear perturbation theory for assessment of void coolant reactivity effect doesn't permit to analyse the broad ranges of coolant density decreasing. The results of direct calculations of Keff variations caused by coolant density decreasing are presented in Fig.3. One can see that coolant density decreasing leads to significant non-linear changes of Keff. When alloy 25 wt%Na + 75 wt%K is used as coolant the positive reactivity insertion caused by coolant density decreasing doesn't exceed the β_{eff} and may be compensated by the slight variation of FBR dimensions. So it may be concluded that the most preferable coolants for FBR-minor actinides burner are potassium, sodium-potassium alloy and lead enriched by Pb-208. As it is shown in Table 4 the effective fraction of delayed neutrons for FBR-actinide burner is comparable

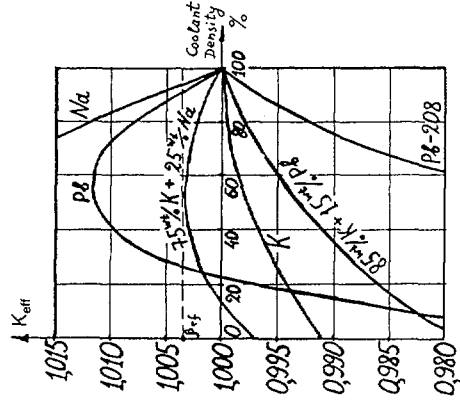


FIG. 3. K_{eff} as a function of coolant density in a fast reactor (fuel: ²³⁵U + MA + Th + 5 wt% Zr).

TABLE 4. β_{eff} , λ_{prompt} AND FISSION FRACTION OF MINOR ACTINIDES

Fuel	Coolant	Void effect of reactivity, %	Delayed neutrons β_{eff} , %	Prompt neutrons λ_{prompt} , 10^{-3} s	Fission fraction of MA, %
(U-Pu)-nitride	Na	-1.98	0.339	4.80	—
	Pb	-1.94	0.324	4.40	—
K	—	-0.47	0.352	2.90	—
	Na	+6.19	0.340	1.38	45.0
Pb	—	+4.37	0.341	1.50	46.9
	K	-0.09	0.340	1.20	48.5
25% Na + 5% Zr	—	+1.30	0.341	1.25	47.4
	85% K + 15% Pb	-0.51	0.341	1.23	48.2
Pb-208	—	-2.13	0.337	1.64	47.2

with β_{eff} of traditional fbrs and prompt neutron life-time λ_p is less by 3-4 times. In considered variants of actinide burner: the contribution of minor actinides in total fissions quantity is about 40%, so about 40% of reactor power is generated in burn-up process of these nuclides. Such FBRs are capable to perform the burn-up of minor actinides produced by 10 LWRs of equivalent electric power.

FPS transmutation in Fast Reactor.

In this section it will be considered the problem of transmutation of long-lived FPs with rather high neutron capture cross-sections (such as I-129, Tc-99, Sm-151, Se-79 and Pd-107). As mentioned above there is the principal ability to transmute these FPs in FBR blanket. If we want to increase the transmutation rate, to cut down the FPs volume in blanket and to reduce the neutron losses due to leakage then it is necessary to form the softened neutron spectrum by placing of moderator. In this case Fast Reactor intended for reducing of hazardous nuclides becomes to hazardous nuclear installation itself: fast core surrounded by blanket containing the moderator. Accidental penetration of moderator in core may lead to positive reactivity insertion. It is known that this fact was one of the main reasons to choose the 3-circuit scheme of heat removal in now operating FBRs. The

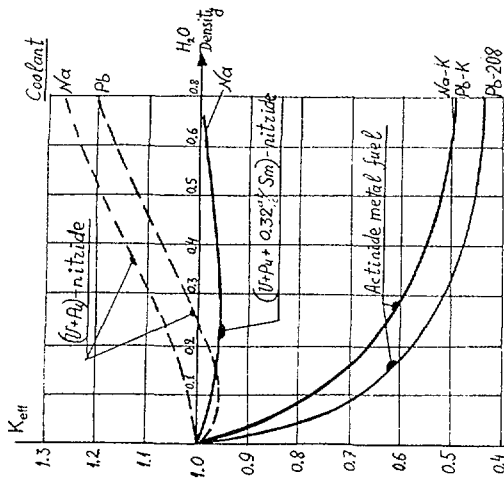


FIG. 4. K_{eff} as a function of H_2O density in the core of a fast reactor.

risk of moderator penetration in core is one of the main arguments of opponents to 2-circuit schemes of heat removal in advanced concepts of FBRs. In FBR-transmutator of FPs such combination (fast core as neutron source surrounded by blanket containing moderator and transmuted FPs) has the principal sense. In Fig.4 it is shown the K_{eff} changes when water penetrates in core. In the case of (U, Pu)-fuel reactor becomes super-critical. But if the minor actinides are inserted in fuel composition then the appearance of hydrogen in core decreases the reactor reactivity. This fact is determined by the threshold energy dependence of minor actinides fission cross-sections. Penetration of moderator in core softens the neutron spectrum, multiplying properties of minor actinides are degrading and K_{eff} is reducing. So it can be concluded that in FBR with sufficiently high fraction of minor actinides there is the principal ability to form the soft neutron spectrum in blanket by placing of moderator there. In order to limit the unfavourable influence of softened neutron spectrum in blanket on burn-up of minor actinides in core it was suggested that core is separated from blanket by coolant layer (see Fig.5)

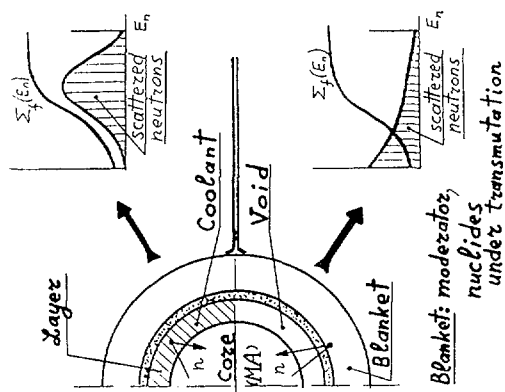


FIG. 5. Influence of layer coolant density on neutron spectrum.

Such technical solution was described in / 7 / at the first time by the author's opinion.

It can be noted that coolant layer separating core with hard neutron spectrum and blanket with soft neutron spectrum carries out two functions. At first, layer reflects fast neutrons leaking the core and prevents the penetration of resonance and thermal neutrons from blanket in core. At second, if the coolant density in layer decreases then reflection of fast neutrons leaking the core is reduced and the penetration of resonance and thermal neutrons from blanket in core is increased. These factors result in degradation of multiplying properties of actinide fuel and so in decreasing of K_{ef} (this effect may also be used in fast breeders by placing the fuel containing minor actinides in the outer part of core).

As an example, the reactivity effect was calculated in actinide burner (see Fig.5) :

- core surrounded by coolant layer (Na-K), thickness - 15 cm ;
- moderator in blanket - $ZrH_{1.85}$; blanket thickness - 100 cm ;

- nuclide under transmutation in blanket - Tc-99 (16 %) ;
- between coolant layer and blanket it is placed the zone (thickness - 3 cm) contained Tc-99 (100 %) .

The decreasing of coolant density in layer (10 %) results in degradation of reactivity $K_{ef} = - 0.162$ % . In the blanket it is realized the transmutation of 0.72 nuclei of Tc-99 per one fission in the core. Taking into account the data of the Table 2 it can be concluded that in the actinide burner there is a possibility for transmutation of Tc-99 generated in 10 reactors (of equivalent power). It is an evaluation leaving out of account the neutron losses.

Transmutation of Sr-90 and Cs-137 in nuclear reactor.

The most dangerous FPs are the isotopes Sr-90 and Cs-137 with half-life 27.7 years and 30.0 years, respectively. Due to low value of neutron capture cross-section of these nuclides it is necessary to upgrade the neutron flux approximately by 1000 times comparing with achieved ones in modern nuclear installations. Obviously in such flux the transmutation of Sr-90 and Cs-137 will be effective.

The main difficulty of this way is the necessity of transmuted nuclides irradiation in high neutron flux with simultaneous requirement of very rational neutron utilization. Non-economical neutron utilization for RAW transmutation in facilities with high neutron flux may result in the negative power balance: power demanded for RAW transmutation to harmless form may be higher than power produced by the reactors where RAW is generated. RAW transmutation in low neutron flux can't result in significant decreasing of RAW amount compared with natural radioactive decay.

For creation of high neutron flux and conditions when neutrons are mostly utilized in RAW transmutation with minimal losses in construction materials it is necessary to use the nuclides with specific properties. These nuclides must have the low neutron capture cross-sections and rather heavy atomic weight for ability to form the desired spectrum of neutrons. One of the possible materials is lead enriched by Pb-208. This isotope of lead is the double-magic nuclide (neutron and proton shells in Pb-208 nucleus

are completed) and so its neutron capture cross-section is very small (less than 0.0005 barn even in thermal region). Heavy atomic weight ($A=208$) determines the small slowing-down capability of this nuclide. This fact in turn may be reason of slowing the chain reaction development in such nuclear system compared with operating nuclear power reactors. It is evaluated that neutron slowing-down in Pb-208 is carried out with time duration by 10-100 times higher than in traditional moderators.

The low slowing-down capability and low neutron absorption probability of Pb-208 are the prerequisites for creation of facility characterized by large volume to place the transmuted radioactive nuclides and by high neutron flux.

So the rapid and effective transmutation of Sr-90 (or Cs-137) requires the high neutron flux and application of materials with low neutron capture cross-sections. In such conditions the dominant fraction of captured neutrons will be related with Sr-90 (or Cs-137). Low concentration of fissile materials in core is needed for the rational limitation of heat generation due to high neutron flux. Rapid fissile materials burn-up in such core demands the really continuous refueling. For instance, such core may consist of the molten Pb-208 mixed with fissile material (preferentially with U-233 because of low σ_c/σ_f in resonance and thermal neutron spectrum) both as fuel and as coolant. Sr-90 may be placed in graphite tubes arranged in regular lattice (see Fig.6).

As example the core with following parameters was considered:

Average value of the core specific power - 130 kWt/l,
 Graphite tubes: outer diameter/thickness - 7.5 cm/1.0 cm,
 pitch of graphite tube lattice - 15.5 cm,
 Fissile material - U-233/U-234/U-235/U-236 = 0.6/0.212/0.048/0.14.

The dependence of K-effective, Fluence ($E_n > 46.5$ KeV), Displacements per graphite atom (DPA-graphite) and Neutron Absorption by nuclides during irradiation in the core are presented in Table 5. It is considered that reactor operates on stationary fuel composition with continuous fuel feeding by U-233. Graphite tubular elements containing Sr-90 are placed in core and irradiated during cycle of predetermined duration (0.5, 1.0 and 1.5 year).

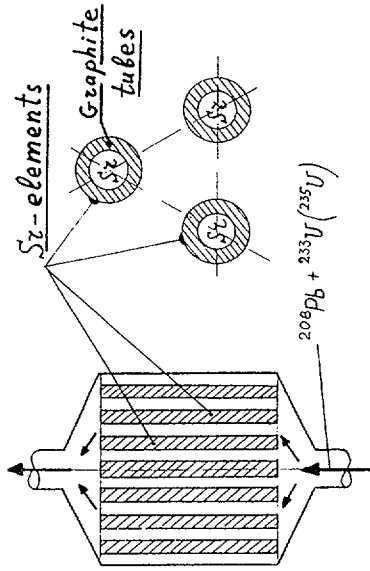


FIG. 6. Principal scheme of liquid metal nuclear reactor for Sr transmutation.

One can see that main fraction of absorbed neutrons is related with neutron capture in Sr-90. Appreciable fraction of neutrons was lost in graphite and FPs which content in lead alloy is supposed to be about 7 ppm. During cycle of irradiation the fraction of neutrons absorbed in Sr-90 decreases due to redistribution of neutron absorptions in produced daughter isotopes: Y-91, Zr-91, 92 and Zr-93. Factors limiting the FPs exposure time are the reduction of useful fraction of neutrons absorbed in Sr-90 and the graphite integrity after high fluence irradiation. Table 5 contains the calculated assessments of neutron fluence ($E_n > 46.5$ keV) and displacements per atom in graphite.

Publications on integrity of different graphites as function of neutron fluence /8,9/ predict the permissible fluence up to $5 \cdot 10^{22}$ n/(cm²). For the considered reactor it means that duration of irradiation cycle for tubular elements containing transmuted Sr-90 may be about 0.6 year. After exposure the tubular elements are discharged, Sr-90 is separated from daughter nuclides and graphite element is changed by the fresh one. Then the cycles of irradiation may be continued.

In the Table 5 it can be seen: at the beginning of irradiation K-trans(Sr-90) = 1.00 (K-trans - the number of acts of neutron

TABLE 5. NEUTRON ABSORPTION IN NUCLIDES OF THE CORE*

	Time of irradiation, years			
	0	0.5	1.0	1.5
FPs	4.08	3.92	3.87	3.90
C-12	8.26	7.92	7.85	7.95
Pb-208	4.74	4.61	4.58	4.61
Sr-90	75.07	59.77	49.49	41.91
Y-91	0	6.48	5.91	5.07
Zr-91	0	8.99	18.55	24.61
Zr-92	0	0.47	1.59	3.08
Zr-93	0	0.05	0.35	0.96
Leakage	7.0	6.78	6.71	6.75
(n,2n)	0.85	0.83	0.83	0.83
K _{eff}	1.000	0.978	0.973	0.979
K _{trans} (⁹⁰ Sr)	1.00	0.829	0.693	0.580
Fluence, $\frac{10^{20}}{(E_0 > 0.5 \text{ MeV})}$	0	4.4 \cdot 10^{22}	8.9 \cdot 10^{22}	15.3 \cdot 10^{22}
DPA(^{graphite})	0	36.6	75.4	113.

* Total absorption (excluding fuel) in the core plus neutron leakage and (n, 2n) reactions equal to 100%.

captures in Sr-90 per one act of fission in the core). Accounting the yield of Sr-90 per fission (about 0.06, or 0.03 after 30-year decay time) it can be concluded that one fission in such reactor leads to transmutation of Sr-90 produced in 14 (or 28) fissions in other reactors. The value of (1 - K-trans) characterizes here the fraction of transmuted Sr-90. If the specific power in that reactor is ~ 130 kwt/1 then neutron flux is $2.65 \cdot 10^{16} \text{ n}/(\text{cm}^2 \cdot \text{sec})$. Such neutron flux provides the rate of Sr-90 transmutation by 12.5 times higher than rate of natural radioactive decay (half-life - 27.7 years). Neutron spectrum in core is close to Fermi-spectrum since the multiplying medium is extremely diluted system. The core is characterized by following average cross-sections: $\sigma_c(\text{Sr-90})=0.41 \text{ barn}$; $\sigma_c(\text{C-12})=1.4 \text{ mbarn}$; $\sigma_c(\text{Pb-208})=0.26 \text{ mbarn}$; prompt neutron life-time $\text{lp}=0.0047 \text{ sec}$. So it can be concluded that the considered nuclear reactor is capable to provide high neutron flux and transmutation effectiveness

of Sr-90 and other FPs with low neutron capture cross-section. However the technical realization of such reactor construction requires the comprehensive analysis of technology to purify and feed the fuel based on Pb-208 alloy. Also it must be considered the important safety problems related with release of delayed neutron predecessors from core in circulating fuel alloy which serves as coolant too. In this concern there is a certain analogue with successful experiment on reactor MSRE / 10 / where similar release of delayed neutron predecessors was performed in circulating fuel alloy used as fuel and coolant. This liquid-fuel reactor was characterized by the high negative temperature and power reactivity effects. The small core specific power (~ 130 kwt/1) and good hydraulic parameters of graphite tube lattice allow to hope that important role in heat removal belongs to natural coolant circulation.

On advantages of external neutron source for FPs transmutation. As shown above the quantity of excessive neutrons in FBR can be sufficient to provide the positive balance in transmutation and generation of long-lived FPs with high biological hazard. Poor neutron balance of thermal reactor doesn't permit to achieve analogous situation. However, estimating the principal ability of fission nuclear reactors in RAW transmutation into harmless forms and considering the use of external neutron sources for this purpose it may be noted the following points.

At first, the value of excessive neutrons suitable for transmutation purposes is not very large and so FBRs may be used for transmutation of its own long-lived FPs plus perhaps from one-two thermal reactors. It means the nuclear power system with small fraction of FBRs will suffer from deficit of neutrons needed for transmutation of hazardous long-lived FPs. It will be necessary to find new sources of neutrons.

At second, among the long-lived FPs there are nuclides with low neutron capture cross-sections such as Sr-90 and Cs-137. For its effective transmutation the high neutron flux is needed, much higher than in traditional nuclear reactors. However, even if such flux is achieved and effective Sr-90, Cs-137 transmutation

is realized then the more serious problem arises related with Sr-88. This nuclide accompanying Sr-90 has neutron capture cross-section less than Sr-90 by 150 times. Isotope separation of Sr-88 is the dangerous radiation procedure since radioactive strontium has to be converted to gaseous phase. Avoiding of isotope separation by using of the supplementary large neutron source is important argument for development of such facilities for FPs transmutation without long decay period.

At third, among the long-lived FPs there is nuclide Sm-151 (half-life \sim 87 years) with complicated scheme of transmutation. This nuclide has rather high neutron capture cross-section for effective transmutation in the neutron flux of nuclear reactor. But Sm-151 is component in the mixture of FPs-samarium isotopes (from Sm-147 to Sm-152). Neutrons will be consumed not only in Sm-151 but they will be uselessly absorbed in other samarium isotopes. To avoid this problem it is necessary to apply the isotope separation as mentioned above. Otherwise the mixture of all samarium isotopes must be irradiated. Since the most of samarium isotopes have the long half-lives (or are stable) the transmutation of mixture will finish by Sm-153 (beta-decay with half-life 46.7 h to Eu-153). Until exhaustion of Sm-147 - Sm-150 isotopes, the precursors of Sm-151, its inventory willn't sufficiently decrease. For transmutation of one Sm-147 nucleus to nuclide of another element Eu-153 it is needed the successive absorption of 6 neutrons. Even separated Sm-151 with frequent purification from daughter isotopes requires at least 2 neutrons for transmutation of one Sm-151 nucleus. So transmutation of Sm-151 in mixture of other samarium isotopes is very neutron-expensive process which may be realized with use of the external source of neutrons. At fourth, the more completed decontamination of RAW requires the extension of isotopes list to be transmuted. The external source of neutrons will be useful for transmutation.

At fifth, duration of exposure followed by the procedures of purification and feeding by portion of fresh transmuted nuclides is determined by the amount of excessive neutrons. Desire to increase the duration of irradiation cycle and to reduce the quantity of outer cycle operations per one transmuted nucleus will

result in problem of neutron consumption balance. Intense external neutron source would be very useful.

At sixth, effective transmutation of relatively short-lived radionuclides with low neutron capture cross-sections (Sr-90, Cs-137) requires high neutron flux. It is impossible to exclude the possibility that needed neutron flux can be successfully created in non-fission nuclear installations with less expensive and less complicated technology. It may be facilities based on nuclear reactions with smaller power output compared with reaction of fission; neutron sources based on fusion reaction or on interaction of intense beams of particles with heavy targets, for instance.

At seventh, one of the important advantages of high neutron flux is related with possibility to use a diluted nuclide systems. Then the transmutation process will deal with small quantities of hazardous radionuclides. It corresponds with the rational desire to reduce the amount of radiotoxic materials both in inner and outer parts of transmutation cycle.

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MINOR ACTINIDE TRANSMUTATION IN FISSION REACTORS AND FUEL CYCLE CONSIDERATIONS

T. MUKAIYAMA, H. YOSHIDA, T. OGAWA
Japan Atomic Energy Research Institute,
Tokai-mura, Naka-gun, Ibaraki-ken,
Japan

Abstract

The concept of minor actinide burner reactor is proposed as an efficient and effective way to transmute long-lived minor actinides in order to ease the burden of high-level radioactive waste disposal problem. Conceptual design studies of minor actinide burner reactors and the fuel cycle facilities were performed to obtain a reactor model with very hard neutron spectrum and very high neutron flux in which minor actinides can be fissioned efficiently. Two models of burner reactors were obtained, one with metal fuel core and the other with particle fuel core. Minor actinide transmutation by the actinide burner reactors is compared with that by power reactors from both the reactor physics and fuel cycle facilities view point.

1. Introduction

Various methods of minor actinide transmutation have been proposed in order to alleviate the burden of the high-level radioactive waste (HLW) depository problem. One of the practical methods is to recycle them in fast reactors since minor actinides such as Np-237, Am-241, Am-243 (hereafter referred as MA) are fissionable with fission threshold in about 700 keV range and capture cross sections of these nuclides rapidly decrease with neutron energy higher than this threshold.

In the early 1980's, the feasibility of MA transmutation in power reactors such as LWR or FBR was shown. In these reactors, however, neutron spectra are rather too soft for MA to directly undergo fission. In thermal reactors, these nuclides undergo fission after one or two neutron capture, for example, Np-237 undergoes fission as Pu-239. Even in FBR, the fraction of neutron of which energy is higher than MA fission threshold is rather small for effective fission of MA.

If a special reactor with a hard neutron energy spectrum and high neutron flux is technically feasible, it will be very efficient and effective for MA transmutation. In this context we have been studying the concept of MA burner fast reactors (ABR: Actinide Burner Reactor). The concept of the double strata fuel cycle consisting of the commercial fuel cycle and the Partitioning-Transmutation cycle is illustrated in Fig. 1. The final HLW from this double strata cycle contains only fission products.

2. Designing of minor actinide burner reactors

In the design study, the fuel property and the thermal hydraulic analyses as well as the nuclear analysis were carried out to obtain an ABR model. The guidelines for designing an ABR are as follows;

- MA as the major fuel material,
- very hard core averaged neutron spectrum,

- very high neutron flux,
- burnup reactivity swing less than 3% $\Delta k/k$ per cycle,
- long fuel residence cycle length within the maximum allowable neutron irradiation of cladding material.

In Table 1, the composition of MA used in this study is shown. Two types of ABR design were obtained, namely MA metal fuel ABR and MA particle fuel ABR. The followings are the brief explanation of ABRs designed. The details of these ABR designing are described elsewhere^{1,2,3,4} For design study, a computer code system "ABC-SC" was developed to analyze actinide transmutation in fast reactors.⁵⁾

2.1 Na cooled MA alloy fuel ABR(M-ABR)

To design a core with a very hard neutron spectrum, a metal fuel core is the first choice. The pyrochemical reprocessing of metal fuel is also attractive from an economic view points of fuel cycle facilities because of the compactness.

Experimental MA fuel property data which are essential for designing fuel and a reactor core are very scarce. Theoretically estimated data, therefore, were used in design study. The followings are the result of the estimation⁴⁾:

- 1) Similar to U-rare earths, Np and Am are not mutually soluble,
- 2) to improve low melting point of MA metal, e.g. 640°C of Np, MA elements are to be alloyed with thermal diluent such as Zr and Y,
- 3) existence of Pu would not significantly affect the solidus of alloys.

Table 1 Minor actinides generated in a 3410MWt-PWR per year

Nuclide	Cooling time (year)	
	3	6
²³⁷ Np	56.2%	47.6%
²⁴¹ Am	26.4	38.3
²⁴³ Am	12.0	10.1
²⁴³ Cm	0.03	0.02
²⁴⁴ Cm	5.11	3.83
²⁴⁶ Cm	0.28	0.23
Total	100.	100.
Total Weight	23.8kg	28.9kg

Calculation data : JENDL-2
 code : SRAC-FPGS
 Burnup of fuel : 33,000MWD/MT
 Recovery of U and Pu : 100%

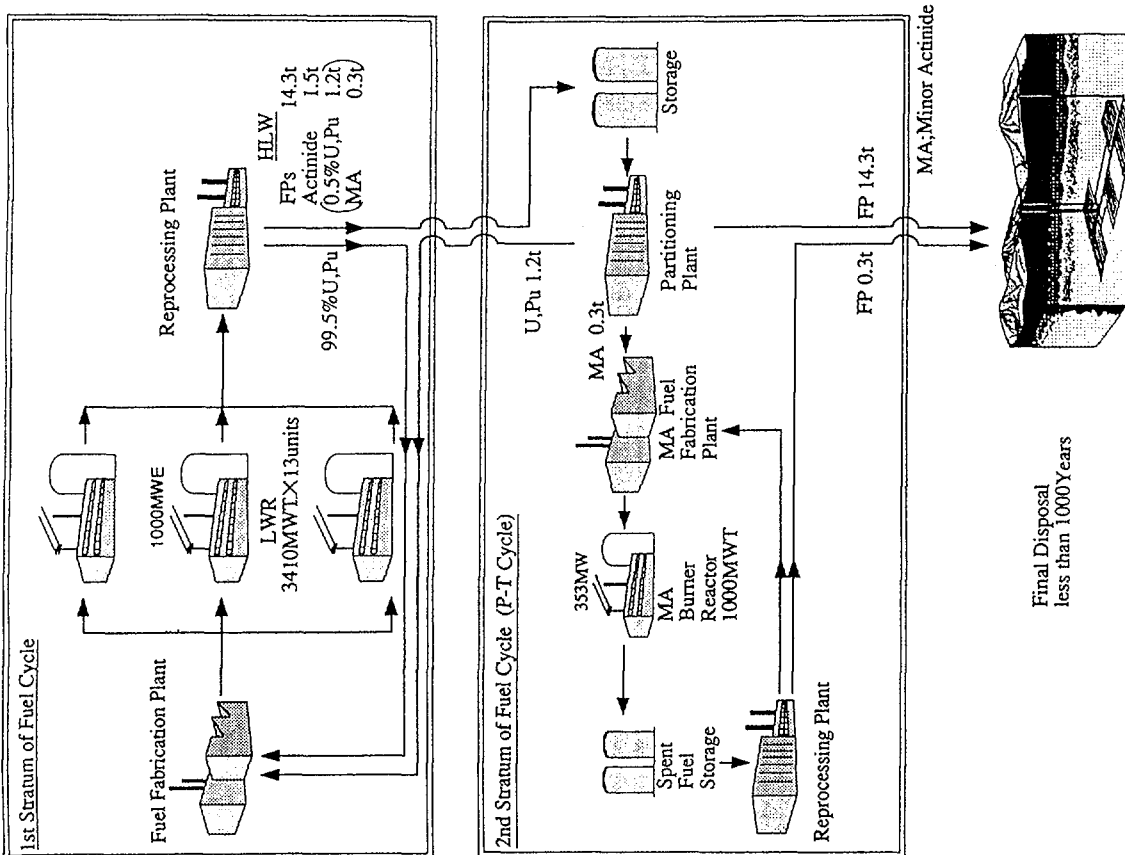


Fig. 1 Flow of high-level radioactive waste per year through double strata fuel cycle combined with partitioning and transmutation (MA burner reactor) cycle

As the result of these nature of MA metal property, we have to use two alloy systems, namely, Np-(Pu)-Zr and Am-Cm-(Pu)-Y. In these alloys, Pu is added because of two reasons; 1) To reduce critical mass, 2) To compensate for reactivity gain which is caused as a result of conversion of Np-237 to Pu-238, Am-241 to Am-242m introduces large reactivity gain and this should be compensated with burnup reactivity loss of Pu. Pu, however, is added only at the initial loading and after the first loading Pu converted from Np-237 plays this role.

2.2 He cooled MA particle bed ABR(P-ABR)

Low thermal conductivity and melting point of MA metal fuel are the limiting factors for the high MA burnup in M-ABR. Therefore, the particle bed reactor concept was applied as an alternative ABR, which has the high efficiency in heat transfer since small particle size produces a large heat transfer surface per volume.⁶⁾ The bed of coated fuel particle contained in double concentric porous frits is directly cooled by helium. The fuel is a microsphere of MA nitride which is coated with a refractory material such as TiN.

In a cold fuel concept, fuel temperature is to be kept lower than one third of its melting point to reduce mass transport. Since reduced mass transport results in smaller swelling and gas release, thickness of coating layer can be minimized to give large heavy-metal density in order to obtain hard neutron spectrum in a core. The kernel of the fuel particle is homogeneous mixture of Pu and MA nitride. The fuel concepts of M-ABR and P-ABR are shown in Fig.2.

2.3 Designing of ABR plants and fuel cycle facilities

Conceptual design studies of ABR plants and their fuel cycle facilities were also carried out to assess the feasibility of ABR concepts. In Fig. 3, the heat balance of M-ABR plant consisting of 6 modules of M-ABR is shown. In this plant, 300kg of MA generated in about 13 units of 3410MWt-PWR undergo fission yearly and 353MW electricity is generated.

In Fig. 4, the heat balance of P-ABR plant is shown. In this scheme, 355kg of MA from 15 units of PWR is fissioned yearly. Because of the cold fuel concept adopted for designing a P-ABR, the fuel temperature is restricted to lower than 1000K and the coolant output temperature is only 340°C which is low as He-cooled reactor. Under this low temperature condition, an applicable way of the generation of electricity will be by use of working fluid with low melting point, such as freon. To avoid the material corrosion problem by thermal decomposition of freon, the temperature of freon should be lower than 140°C. Owing to these low temperature, the electricity generation efficiency is only 15%.

In the designing of a fuel cycle, pyrochemical reprocessing of spent MA-nitride fuel was studied because of its compactness. The estimated mass flow of actinides (MA and Pu) in the fuel cycle facilities is only 19kg of actinides, 3.2kg of fission products, etc. per day for one unit of 1200MWt P-ABR plant when the plant load factor is 30% (Fig.5).

3. Characteristics of ABRs

3.1 Reactor performance

The reactor core design parameters of M-ABR and P-ABR at their equilibrium state are shown in Table 2, respectively. Comparison of core averaged neutron spectra is shown in Fig.6. In this

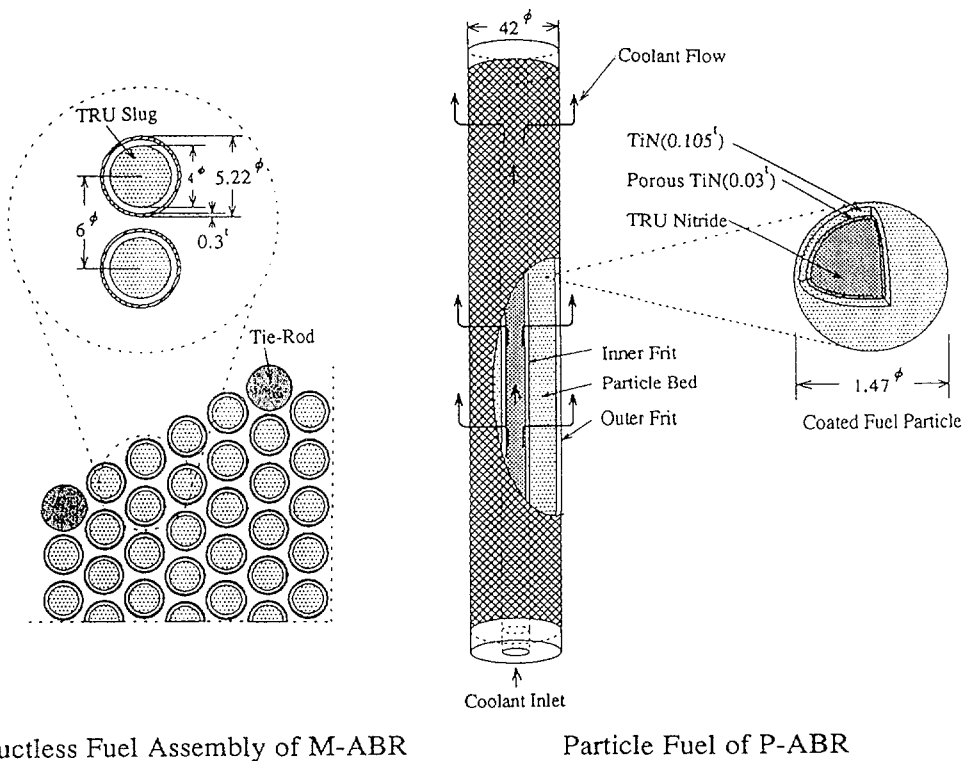


Fig. 2 Fuel Concept of ABR (unit mm)

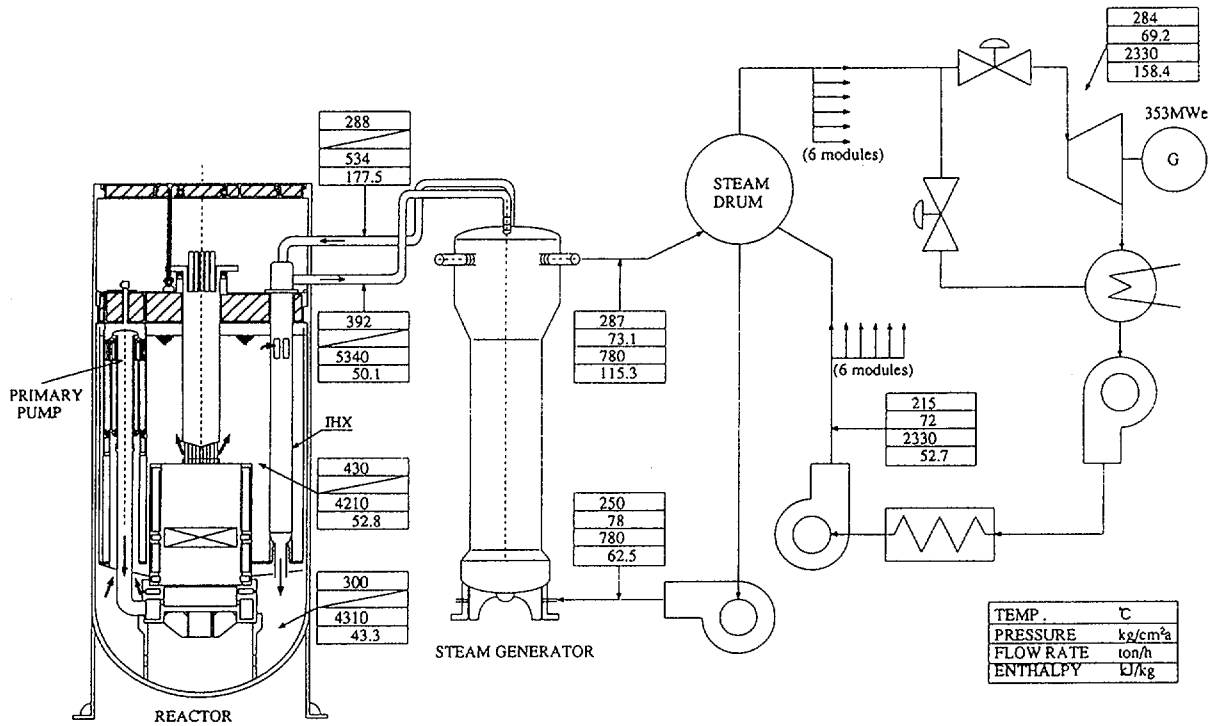


Fig.3 Heat balance of M-ABR plant

A plant which consists of 6 units of M-ABR burns 300kg of MA(minor actinides) per year and generates 353MW electricity

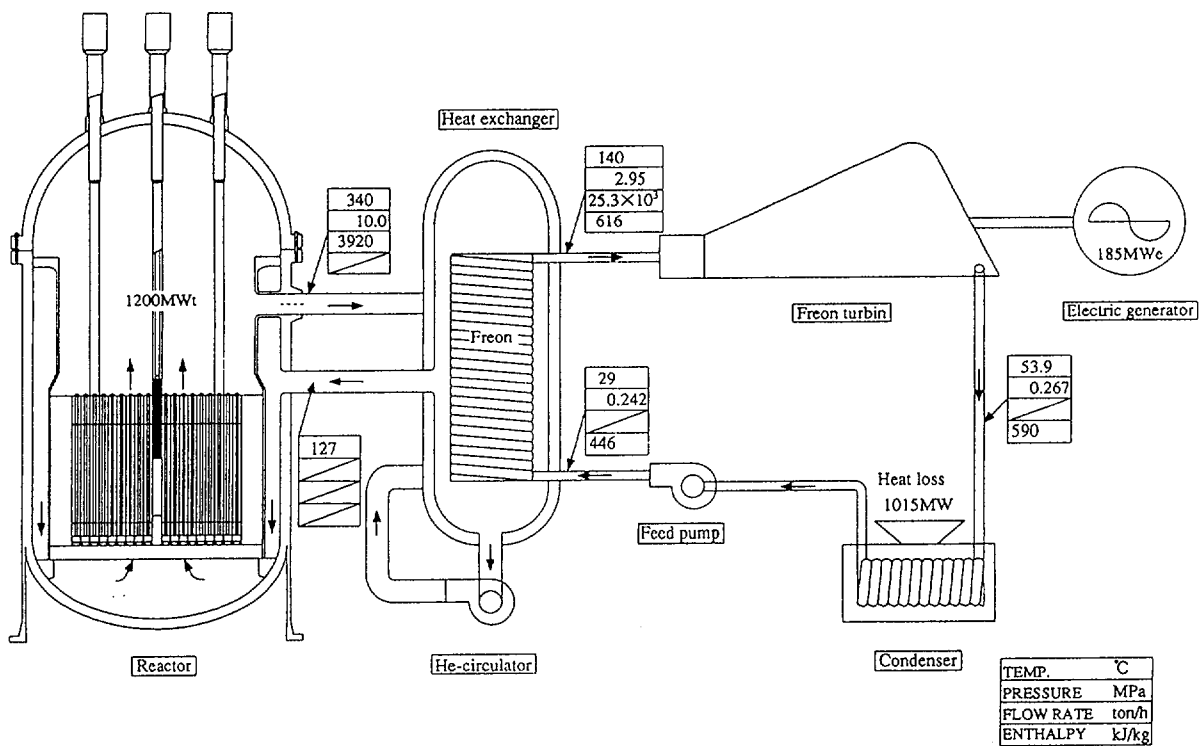


Fig.4 Heat balance of P-ABR plant

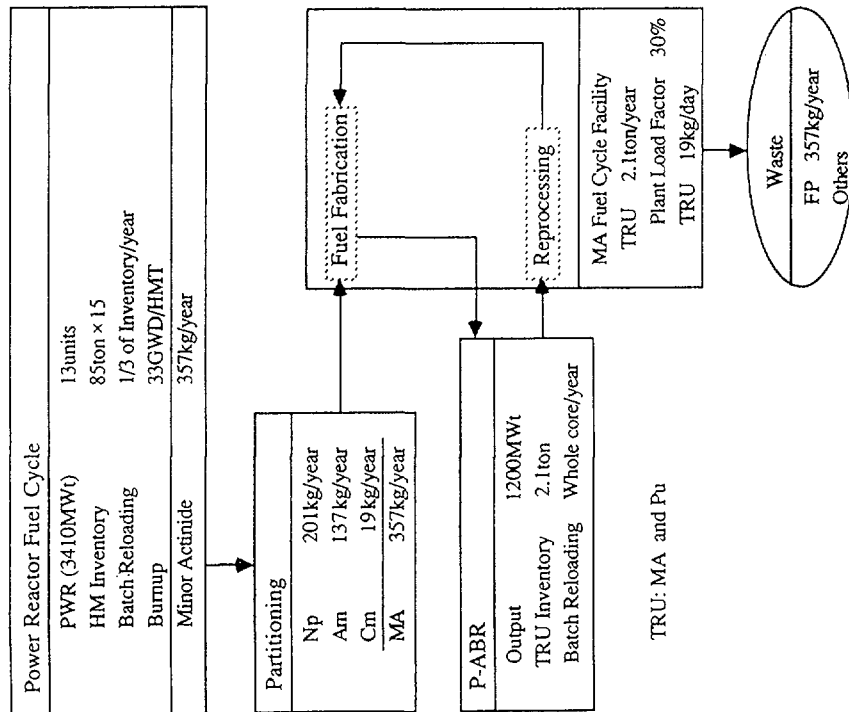


Fig.5 Mass Flow in P-T Fuel Cycle with P-ABR

figure, neutron spectrum of MOX-LMFBR is also shown for comparison. Significantly hard neutron spectra of ABRs are obvious. In the M-ABR, the neutron flux is not so high as opposed to the initial attempt to design a very high neutron flux reactor because of low melting point and low thermal conductivity of MA fuel. In the P-ABR, the neutron flux is very high and the MA burnup per year is larger than that of M-ABR owing to the efficient heat removal characteristics of particle fuel. One of the significant differences between ABRs and power reactors is that the fuel residence cycle of ABRs is limited by neutron fluence while those of power reactors are limited by burnup reactivity loss. This is due to the fact that in ABRs the burnup reactivity swing is small as the result of compensation for burnup reactivity loss by reactivity gain from the conversion of MA to fissionable material (eg Np to Pu, Am-241 to Am-242/Am-242m).

Table 2 Reactor design parameters of Actinide Burner Reactors

M-ABR ¹⁾		P-ABR ²⁾	
Fuel concept material	pin-bundle IC ³⁾ :Np-22Pu-20Zr OC:AmCm-35Pu-5Y	coated particle (66NpAmCm-34Pu) ₁₀ N _{1.0}	
MA initial loading ⁴⁾ kg Np/Am,Cm/Pu	666 255/199/212	2065 765/598/702	
Reactor power, MWth	170	1200	
Coolant material	Sodium	Helium	
velocity, m/s	8	1088	
inlet temperature, °C	300	10	
outlet temperature(core max), °C	IC:484 OC:440	13	
Fuel temperature, °C max ⁵⁾	IC:834 OC:809	127	
Clad temperature, °C max ⁶⁾	IC:517 OC:484	340	
Neutron flux, 10 ¹⁵ n/cm ² .sec	IC:4.1 OC:3.4	722	
Neutron fluence (E>0.1MeV), 10 ²³ n/cm ²	IC:2.2 OC:1.7	560	
Core averaged mean neutron energy, keV	IC:766 OC:785	8.4	
Reactivity (% Δk/k)		2.2	
Na-void reactivity/core	2.52	743	
Doppler reactivity/core (Δt=300°C)	-0.01		
Kinetic parameters			
β _{eff}	1.55 × 10 ⁻³	1.72 × 10 ⁻³	
L _p , sec	6.84 × 10 ⁻⁸	10.8 × 10 ⁻⁸	
Cycle length, full-power days ⁷⁾	730	300	
MA transmutation, %/cycle	26.0	25.3	
MA burnup, %/cycle	17.8	17.3	

- 1) M-ABR:MA metallic fuel actinide burner reactor
- 2) P-ABR :MA particle fuel actinide burner reactor
- 3) IC:Inner Core, OC:Outer Core
- 4) After 1st cycle,only Np,Am,Cm are added.
- 5) Predicted melting point of fuel 900°C for M-ABR
Max. allowable temp. of fuel 727°C (1/3 of M.P. 3000K) for P-ABR
- 6) Max. allowable temp. of cladding/frit (HT-9) 650°C
- 7) Fuel irradiation time

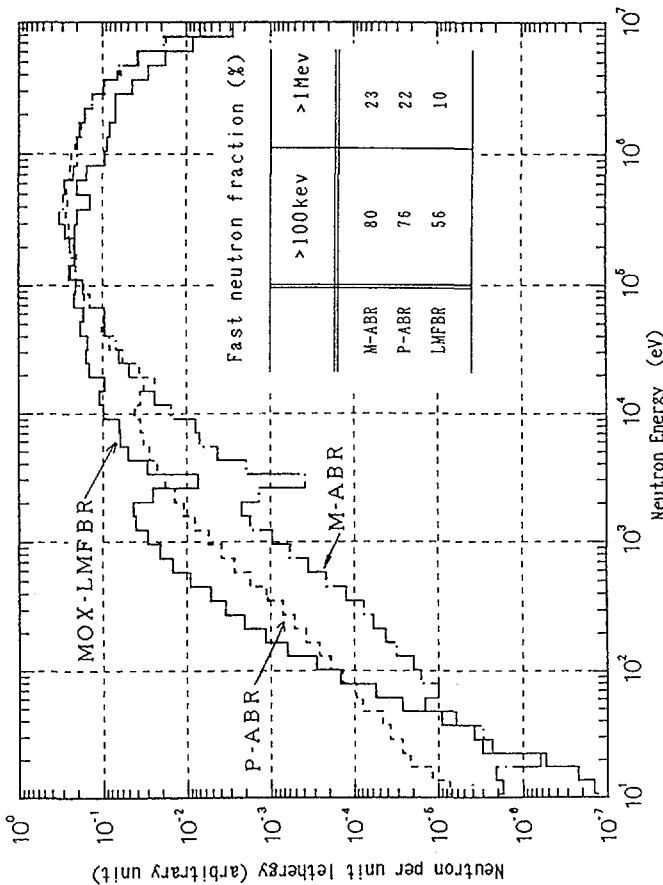


Fig.6 Comparison of core averaged neutron spectra of minor actinide burner reactors and MOX-FBR

In the current design of ABRs, Doppler reactivity coefficient and the delayed neutron fraction are very small and the positive sodium void coefficient is large owing to the lack of U in a core and also to the very hard neutron spectrum

3.2 Effect of Nuclear and Fuel Property Data Uncertainty on the ABR Design

Nuclear and fuel property data of MA strongly affect the design of ABRs because their major fuel material is MA. Also, the composition of MA will be full of variety depending on the spent fuel of different irradiation history. The effect of nuclear data uncertainty was checked by two sets of calculation for M-ABR, one using JENDL-2 data and the other using ENDF/B-V. The summary of this comparison is shown in Table 3. The differences of each parameters are quite large but that of MA burnup is rather small.

As for the fuel property data, if our theoretical prediction of non solubility of Am, Cm in Np is not correct, the fuel design of M-ABR will be very different. The thermal conductivity of MA metal fuel was assumed as the half of that of U-Pu-Zr alloy. If this number is one third, the MA burnup is 26% smaller. The 100°C lower melting point of MA fuel than the estimated 900°C results in 18% lower MA burnup.

Table 3 Comparison of JENDL-2 based nuclear calculation and ENDF/B-V based one for M-ABR

Item	Library	
	JENDL-2	ENDF/B-V
One group cross section(barn)*		
²³⁷ Np capture	0.711	0.678
fission	0.575	0.577
²⁴¹ Am capture	1.04	0.960
fission	0.587	0.531
Core averaged neutron mean reaction energy in the inner core(keV)	790	757
k _{eff} of BOC at 1st cycle	1.0313	1.0558
MA bump per cycle in the inner core at 10th cycle(%)	12.91	13.08

*) collapsed using the core averaged neutron spectrum

4. Comparison of MA transmutation in ABRs and in power reactors

In Table 4, the transmutation characteristics are compared between two types of ABRs together with thermal and fast reactors. For the MA transmutation in power reactors, the concentration of MA is assumed as 0.2% and 5% of heavy metal for U-PWR and fast reactors, respectively so that the addition of MA will not affect major reactor design parameters such as enrichment, coolant void coefficient etc.

4.1 MA burnup rate

The transmutation rate is usually defined as the ratio of MA weight at the end of cycle to that of the beginning of cycle. In this definition, any nuclear reaction such as fission, neutron capture, (n,2n), etc. can be used for transmutation and the conversion of Np-237 into Pu is the transmutation. Pu in the transmutation chain of Np-237 is mostly Pu-238 and these dirty Pu is not favorable for the reactor physics and the ex-core fuel handling. Therefore, the MA burnup rate defined as the weight ratio of MA fissioned during the irradiation to that at the beginning of cycle is the real index of transmutation effectiveness and efficiency because only fission is a real transmutation reaction to solve the problem of long-lived MA.

In Table 4, the burnup rate per cycle (transmutation effectiveness index) is 15 to 18% in all reactors except in MOX-FBR. The burnup rate per year(transmutation speed) is 17% in P-ABR and 4 to 7% in other reactors. High burnup rate in P-ABR is due to its hard neutron spectrum and very high neutron flux. In the MA burnup calculation for power reactors, MA generation from their fuel should be accounted for. MA generation is larger than MA burnup in U-PWR and about the half of burnup in fast reactors. The net MA burnup per 1GWt a year of ABRs is significantly larger than that of power reactors.

Table 4 Comparison of MA transmutation in various reactors

	MA Burner Reactors			Power Reactors		
	M-ABR	P-ABR	U-PWR	MOX-FBR	LMR	
Output (MWt)	170	1200	3410	2600	2632	
Cycle length ¹⁾ (FPD)	730	300	850	750	900	
Core averaged Neutron flux ($\times 10^{15}$)	3.6	8.4	0.37	3.3	5.0	
Mean neutron energy (keV)	780	750	thermal	480	490	
MA loaded (kg)	666	2065	180 ²⁾	1450 ²⁾	1200 ²⁾	
MA generated ³⁾ (kg/cycle)	-----	-----	71.4	83.5	90	
MA transmutation ratio ⁴⁾ (%/cycle)	26.0	25.3	54.1	27.6	38.7	
MA burnup ratio ⁵⁾ (%/cycle)	17.8	17.2	15.0	9.4	16.8	
Net MA transmutation (kg/1GW · year)	407	435	2.5	62	47	
Net MA burnup (kg/1GW · year)	279	296	-4.3	8.1	14	

1) Fuel irradiation time

2) Concentration of MA in fuel ; 0.2% for U-PWR, 5% for MOX-FBR and LMR

3) MA generation in fuel, i.e U and Pu

4) MA transmutation ratio = (MA(BOC) - MA(EOC)) / MA(BOC)

5) MA burnup ratio = (MA fissioned) / MA(BOC)

4.2 Influence of MA transmutation in power reactors on fuel cycle

For the MA transmutation using power reactors, not only reactor performance and fuel manufacturing but also the influence of transmutation on the fuel cycle facilities should be taken into account. The large difference between the transmutation rate and the burnup rate of power reactors shown in Table 4 indicates the larger conversion of MA to heavier nuclides than fission of MA. To evaluate the effect of MA addition to the fuel the analysis was made to calculate the increase of decay heat, neutron emission, and γ -ray intensity. In Table 5, the increase of neutron emission in spent fuel caused by the increase of neutron emitters such as Cm, Bk and Cf when 0.2wt% MA is added to PWR fuel is shown. In Table 6, the effect of MA addition to power reactors of U-PWR, MOX-PWR and FBR is summarized. The increase of decay heat is caused by the generation of Cm-244 and the increase of neutron emission is caused by the generation of higher Cm isotopes and Cf-252.

In the case of fast reactors, the Tl-208 build up will be also the problem because of its high energy γ -ray (2.6MeV). Tl-208 builds up in the Th decay chain starting from Np-236 which is formed by (n,2n) reaction of Np-237. When 5wt% MA (composition shown in Table 1) is added to the MOX-FBR fuel, the Tl-208 build up is about 50 times as much as that of without MA addition. At this level of the concentration, Tl-208 build up will pose a shielding problem.

As a result of increase of these Cm, Cf and strong γ -emitter, for all of the MA transmutation scheme in power reactors shown in Table 4, the shielding design change will be needed not only for the fresh fuel handling (manufacturing and transportation) but also for the spent fuel handling (transportation and reprocessing). This may cause the increase of cost in the generation of electricity.

Table 5 Effect of MA addition in PWR fuel (values : per ton of HM)

Item	Reference PWR	MA-PWR	(MA-PWR)/(Ref.PWR)
Nuclide (g)			
Np	469	918	2.0
Am	162	276	1.7
Cm	38	296	7.7
Bk	3.4×10^{-7}	1.7×10^{-4}	510
Cf	3.5×10^{-7}	2.4×10^{-4}	690
α -activity (10^5 Ci)	0.29	1.3	4.6
(α, n) (10^6 n/s)	6.3	36	5.8
Spnt. fission (10^9 n/s)	0.49	3.5	7.1

MA addition : 0.2% of HM
 U enrichment : 3.2%
 Burnup : 33000MWD/T
 Irradiation : 847 days
 Cooling : 150 days

In the case of ABRs, the shielding and the decay heat removal are much severer problem than the MA transmutation in power reactors since the concentration of MA is very high in ABRs. As it is mentioned in the section 2.3 of this report, the fuel cycle facilities for ABRs are very compact and the number of these facilities is also limited. Therefore, the economy of MA transmutation may be favorable for the ABRs even if the resources required to develop ABRs is larger than that for MA transmutation in power reactors.⁸⁾

5. Conclusions

Two types of ABR are designed. In these burner reactors, the MA burnup rate per cycle is significantly higher than those in power reactors. The small Doppler reactivity coefficient, delayed neutron fraction and the large positive sodium void coefficient of the present ABR designs is less favorable from the reactor safety point of view. MA transmutation in power reactors (L-WRs and FBRs) will require the design change of the radiation shielding in the whole fuel cycle facilities because of the increase of strong neutron and γ -emitting nuclides. Cost evaluation is required to

Table 6 Effect of MA addition to power reactor fuel handling

Reactor/Fuel	Ratio of value of MA added fuel to that of normal fuel	
	Decay heat	Neutron emission Photon intensity
U-PWR (0.2wt%) ^a fresh fuel(U235:4wt%) ^b spent fuel(45GWD/t) ^c	3.6×10 ³ 1.5	8.3×10 ⁴ 4
MOX-PWR (0.5wt%) fresh fuel(Pu:6.5wt%) spent fuel(45GWD/t)	1.4 1.5	4.8 1.7
MOX-FBR (5wt%) fresh fuel(Pu:30wt%) spent fuel(80GWD/t)	2.2 2.8	1.0×10 ² 19

a: minor actinides(MA) fraction in fuel (HM weight %)

b: fuel enrichment

c: fuel burnup (cooling time : 10 years)

choose the cost effective transmutation system based on the reliable data base which is not yet available.

The ABR concept will enable the confinement of troublesome MA in one closed site. From the economics and safety view point, the confinement of MA may be desirable.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the significant contributions of Y.Gunji of Nuclear Fuel Inc.in the numerical analysis.

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HOMOGENEOUS RECYCLING OF MINOR ACTINIDES IN AN EFR TYPE FAST REACTOR

H.W. WIESE, B. KRIEG

Institut für Neutronenphysik und Reaktortechnik,
Kernforschungszentrum Karlsruhe,
Karlsruhe, Germany

Abstract

The capability of the European Fast Reactor (EFR) as a typical large fast reactor to incinerate the minor actinides Np and Am, produced in LWRs, has been investigated for the case of homogeneous recycling of Np, Am and Pu.

Detailed account is taken for a three-region reload scheme in which every two years 1/3 of the core is refuelled. The fraction of admixed minor actinides is varied from 2.8 to 7.5%.

Results are given in terms of the number of clean-up LWRs by one burner, the nuclide inventories during recycling, and the risk potential of the waste from the incineration system compared to the non-incineration case.

It is concluded that an enforced research should be done in chemistry to prove that a satisfactory separation especially of Am and also of Cm from the rare earths in the waste is possible on large scale.

1. INTRODUCTION

KfK and the French CEA are investigating in close cooperation the transmutation of hazardous nuclei as Np, Am, Cm and the long-lived fission products Tc99 and I129 in fast and thermal reactors.

Since in fast reactors fission products cannot be destroyed efficiently - except to a certain degree in moderated subassemblies in outer core or blanket regions - this paper is restricted to the incineration of actinides.

The results presented here are part of a common CEA/KfK paper [1] prepared for the ANP Conference in Tokyo in October 1992. However, here also the risk potentials in terms of the ingestion hazard from the minor actinide waste of the incineration system and the LWR without actinide burning is compared. Since for storage times up to about 1000 years the fission products dominate the risk potential, this comparison mainly is performed for medium- and long-term storage times larger 1000 years.

In order to account for a realistic recovery of Am, i.e. 5-40% during reprocessing using the PUREX process, as a parameter a loss of Am of 70% is assumed in this work.

The incineration capability of the core of a large fast reactor in terms of the number of LWRs which may be cleaned-up by one FBR is determined. Special atten-

tion is paid to the evolution of minor-actinides contents during recycling, and to Pu238 as a transmutation product of Np237 and Am241.

2. EFR AND LWR SPECIFICATIONS

For the EFR, a core mass of 40.3tHM, an average core burn-up of 130GWd/tHM in 3 cycles, a cycle length of 2 years (640 fpd, 90 d for discharge and reloading), a cooling time of 3 years, and 1 year for reprocessing plus refabrication are assumed.

Fuel management model : The core is subdivided in 3 equal-mass load regions R1, R2 and R3. At BOC1, all three load regions are charged with fresh fuel. At EOC1,2,3, the regions R1,2,3 are discharged, and recharged with fresh fuel and Np, Am from external LWR waste, respectively, assuming that Np and Am can be separated from the waste. The irradiated fuels from R1 and R2 are only partly burnt and are withdrawn from the process of recycling. At EOC4,5,6, again R1,2,3 are discharged, with reloading of R1 and R2 with fresh fuel and with Np, Am from external LWRs. For R3, the reload now consists of fresh fuel, of recycled Pu, Np, Am from the reprocessed fuel discharged at EOC3 and of Np, Am from external LWRs. This process is continued, always recycling in cycle n the Pu, Np and Am discharged at the end of cycle n-2. Reprocessing losses of 1% are assumed for Pu and Np. Since Am is closely bound to the rare earths and hence can be recovered to only 5-40%, in the calculations estimated losses of Am of 70% are used.

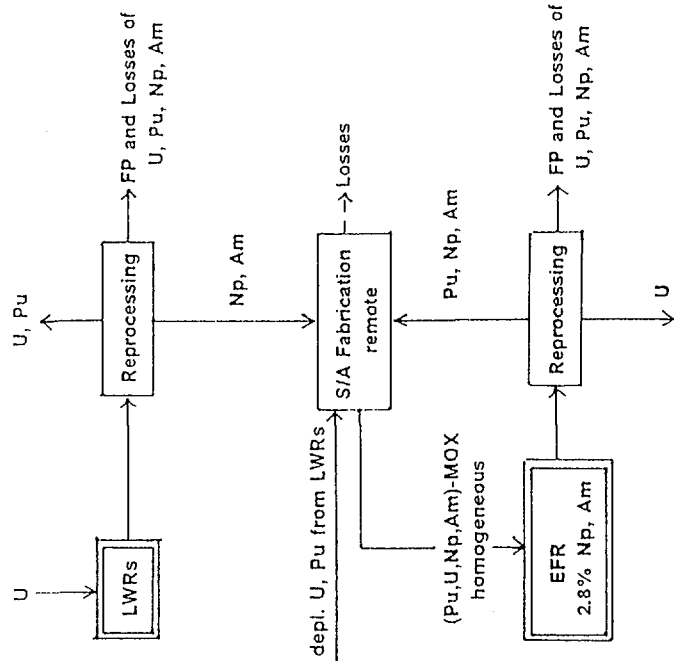
As a parameter, the amount of admixed Np and Am is set to 2.8, 5.0 and 7.5% of the total reload mass per cycle. Reactivity control is performed by an appropriate change of the Pu/(U + Pu) ratio of the reload fuel at each cycle. At BOC 2 and 3, Pu/(U + Pu) is increased according to the reactivity loss due to the Np and Am charge from LWRs only.

The burn-up reactivity loss in this phase is assumed to be compensated by withdrawing absorber rods. From cycle 4, k_{∞} at BOC is kept constant to that of cycle 3. Due to the deterioration of the recycled Pu, for 2.8% admixture Pu/(U + Pu) increases from 20.5% at BOC 1 to 27.5% at BOC 19. Since the equilibrium core inventory depends on the addition of fresh plutonium, refined 2d reactor calculations to determine the needed Pu from k_{eff} for the full reactor will be used for further investigations.

The Np and Am to be burnt in the EFR originates from a reference 1300MW_e PWR of 40GWd/tHM average burn-up, a reload of 24.44 tHM per year, a cooling time of 3 years before reprocessing, and 1 year for fuel element fabrication. The flow of the actinides during recycling can be seen from Fig. 1.

3. CALCULATIONAL MODEL

One recycle period comprises 5 EFR cycles (10 years). At each cycle from cycle 6 on, the reload fuel consists of several streams : of fresh U, Pu fuel and of por-



Reprocessing Losses : U, Pu, Np 1 %, Am 70 % (1 %)
 Np, Am Loading in the EFR 2.8 - 7.5 %

Fig. 1 : Scheme of Minor Actinide Incineration Including Recycling of Pu

tions of Pu, Np and Am which have run through one or more recycle periods. Since the burning behaviour of the homogeneously in relatively small amounts admixed materials approximately is determined by the neutron spectrum of the dominating fresh-fuel part, the relative variation of the isotopes of Np, Am and Pu from LWRs and the EFR during recycling can be pre-calculated by using only two sets of effective EFR neutron cross sections for the inner and outer core, namely that of the fresh fuels.

To account (a) for the different compositions of the external LWR waste and the EFR self-generated Np, Am and Pu, and (b) for the different spectral conditions in the inner and outer EFR core, for both wastes and for the inner and outer core separate pre-calculations for 1 HM LWR and EFR reference fuels were performed with the KORIGEN code [2].

The set of recycling equations, based on the space and time fuel mass balances, is solved with respect to the nuclide concentrations at begin of each cycle using actual EFR and LWR fuel masses and the above given recycling scheme.

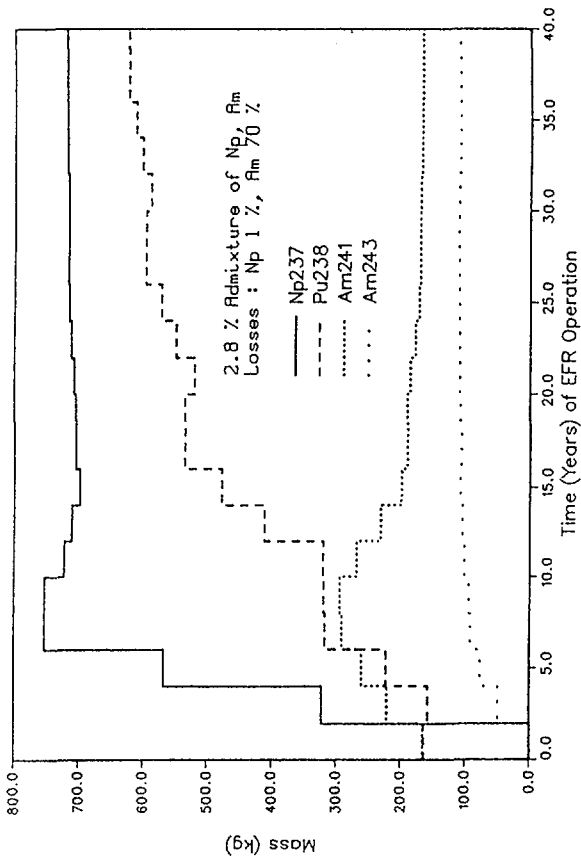


Fig. 2 : In-Pile BOC Nuclide Masses During Recycling

4. RESULTS

4.1. IN-PILE CHARACTERISTICS

From Fig. 2, showing the evolution of the masses of Pu238, Np237, Am241 and Am243 during recycling, a tendency to reach equilibrium concentrations after about 36 years (18 cycles) is stated. In the first ten years when there is no self-generated Np, Am available, LWR Np, Am, mainly consisting of Np237, is dominating. In the course of recycling when, because of incomplete burning, less LWR material can be charged to the EFR, the content of Np decreases whereas the self-generated part is gaining importance. The strongly reduced external load depicted versus the time of EFR operation.

In Tab. 1, the equilibrium in-pile masses of the minor actinide nuclides and of Pu238, the maximum Pu238/Pu, and the number of reference LWRs which may be cleaned-up by the EFR in equilibrium, are listed for 2.8, 5.0 and 7.5% minor-actinides admixtures and for reprocessing losses of 1% for Pu and Np and of 70% for Am.

For 2.8% minor-actinides admixture Pu238 is 6% of Pu. For larger admixtures Pu238 increases to more than 10% of Pu. The number of cleaned-up LWRs significantly increases with the admixture of Np and Am : for 5% admixture already 11 LWRs could be cleaned-up. Then, however, a Pu238 content of 9% would have to be dealt with.

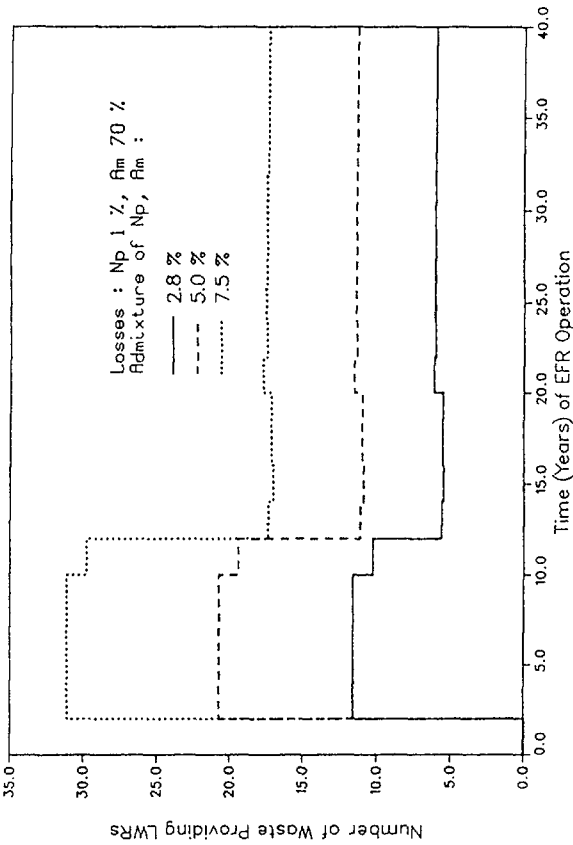


Fig. 3 : Incineration Capability of the EFR

TAB. I : EFR CORE EQUILIBRIUM CHARACTERISTICS DEPENDENT ON NP, AM ADMIXTURE

Quantity	Unit	Parameters and Calculated Values
Np, Am Admixture	%	2.8 5.0 7.5
Np237	kg	720. 1329. 2020.
Am241	kg	168. 236. 315.
Am243	kg	109. 148. 193.
Pu238	kg	613. 1036. 1518.
Pu238/Pu	%	6.0 9.0 11.6
Cleaned-Up LWRs		6.0 11.3 17.4

4.2. MEDIUM- AND LONG-TERM ACTINIDE RISK POTENTIALS

In this work, the risk potential (or hazard) is determined in units of the maximum allowed annual radioactivity ingestion as given in Ref. [3] from 1989. Probabilities of migration of the hazardous nuclei from the final depository to the biosphere are not accounted for if the risk potential is measured in this way.

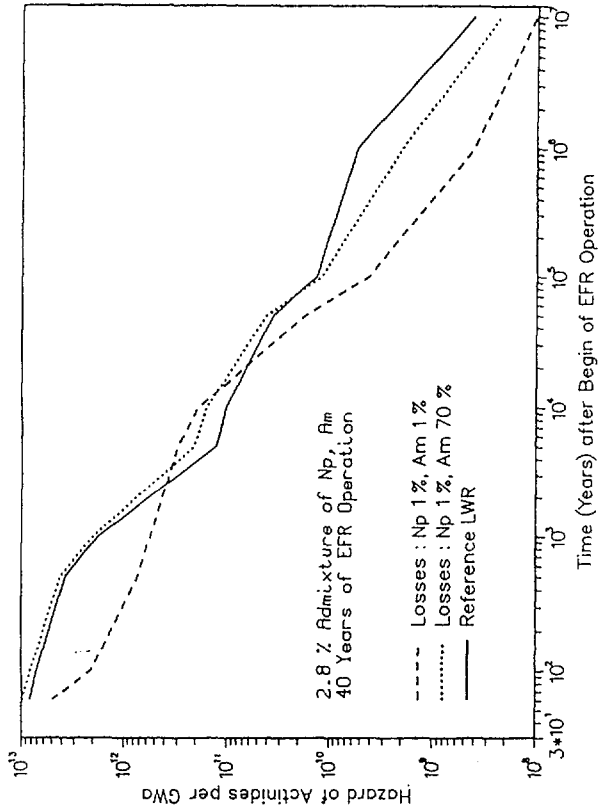


Fig. 4 : Waste Ingest. Hazard without Equilib. Contribution

In a recent work of Küsters and Wiese [4], former toxicity indices had been used giving rise to a marked contribution of Ra226 after about 10⁵ years of storage. This peak is decreased if the toxicity values from Ref. [3] are applied.

Figures 4 and 5 show the risk potential of the waste actinides per Gwa electrical output dependent on the time after begin of the EFR operation in the case of 2.8% admixture of Np and Am. The waste of the reference LWR comprises all the minor actinides produced during LWR operation and the reprocessing losses of U and Pu. The other curves show the hazards of the waste actinides from the LWR-EFR system for 70 and 1% losses of Am. These waste actinides consist of the accumulated losses from reprocessing

- of the recycle burner fuel, including U and Pu losses
- of the LWR fuel, the Np and Am of which is inserted into the EFR, including U and Pu losses

U recovered from burner fuel reprocessing, and U and Pu recovered from LWR fuel reprocessing, are assumed to be used for later fabrication of fuel - Fig. 1.

Excluding the burner inventory from the hazard calculation - as done in the calculations for Fig. 4 - means that burner reactors are continuously installed and are acting as interim storage for the hazardous nuclei. In this case only the reprocessing losses have to be finally disposed of.

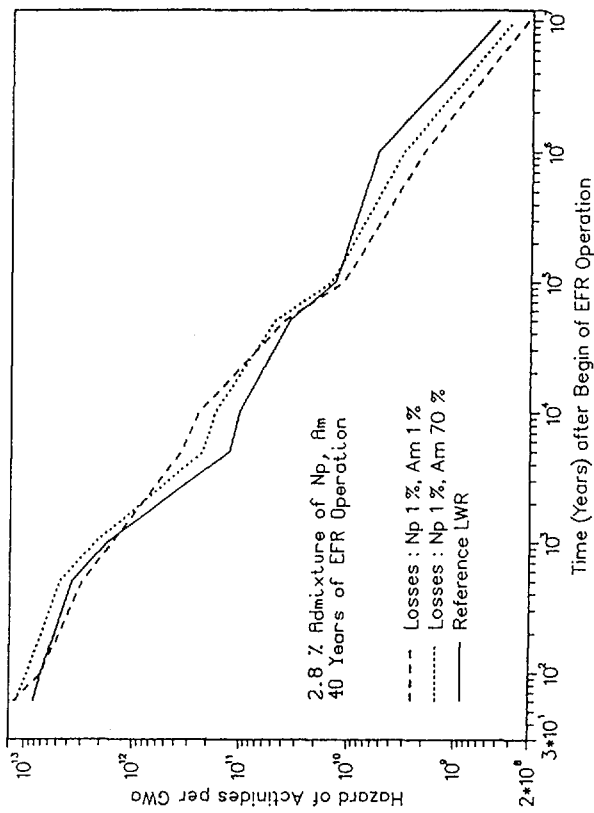


Fig. 5 : Waste Ingestion Hazard with Equilibr. Contribution

If, however, on the other hand the operation of burners is stopped, then the equilibrium inventory will have to be finally disposed of and will then contribute to the waste hazard, too. The hazards in this case are shown in Fig. 5. From Fig. 4, the strong effect of the losses of Am with respect to the hazards can be seen. In the case of probably unrealistic Am losses of 1%, a reduction of up to one order of magnitude is calculated in comparison to the hazard of the reference LWR except in the time region around 5000 years. For estimated losses of Am of 70%, a reduction of the hazard for storage times larger than 10^2 years, i.e. for the long-term hazard, of about 50% is stated. The increase of the hazard around 5000 years is mainly due to Pu240 and, less pronounced, to Pu240 originating from reprocessing losses and from decay of Am243 and Cm244.

Fig. 5 shows that in the case of stopping the operation of burner reactors a smaller reduction of the long-term hazard is achieved. A more drastic representation of the incineration ability is given in Figures 6 and 7, showing the ratios of the actinide hazards of the LWR-EFR system and the hazards of the reference LWR.

In the case of large losses of Am the generation of Cm244 from Am243 and hence the build-up of Pu240 is reduced. This leads to a decrease of the peak at 5000 years compared to the case of low losses of Am. In the long-term range where

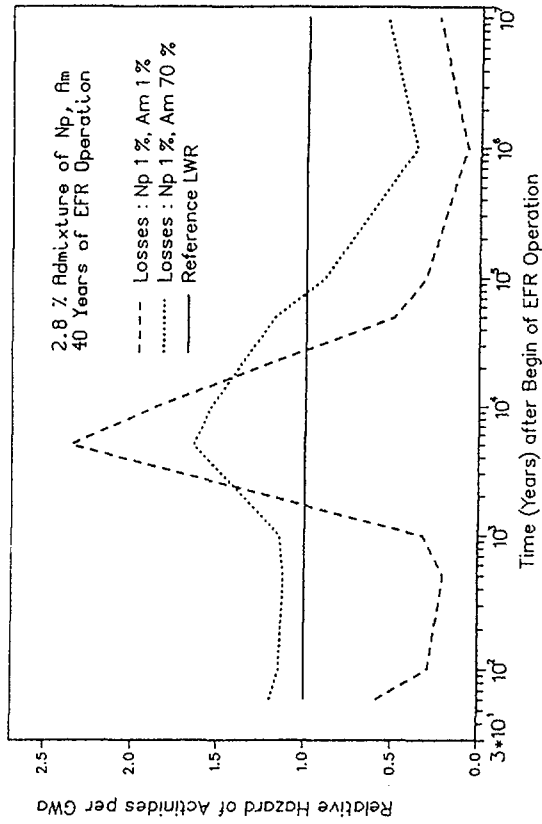


Fig. 6 : Rel. Waste Ingest. Hazard without Equilibr. Contr.

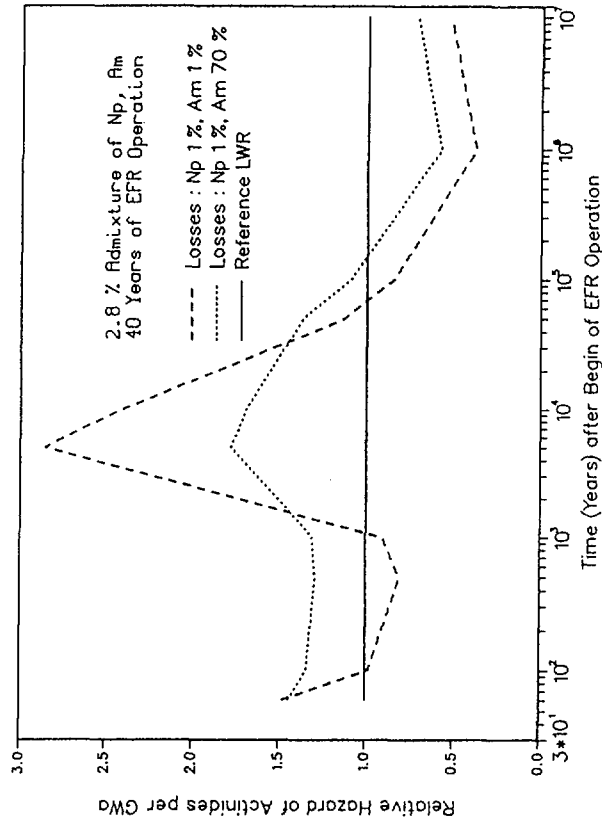


Fig. 7 : Rel. Waste Ingestion Hazard with Equilibr. Contrib.

Np237 and its daughters dominate the hazard, the relatively large amount of recycled Np in the case of large Am losses causes a worsening of the burning ability.

To improve the incineration capability in the medium-term range, a separation of Cm during reprocessing and a subsequent use of the daughter Pu as fuel would be helpful. This is also concluded by Zaetta in Ref. [1]. However, also Cm is closely bound to the rare earths and in the PUREX process its separation is as difficult as that of Am.

The plutonium peaks around 5000 years were also found by Corcuera [1] who investigated the incineration in LWRs as burners. However, since LWRs contain less Pu and hence less transplutonium elements, this peak is less pronounced in LWR-LWR burner systems.

5. SUMMARY

The investigations show that for estimated realistic losses of Am of 70% in comparison to the non-incineration case

- the long-term actinide hazards, i.e. the hazards for storage times larger than 10^5 years, can be reduced to about 40-50%,
- for the medium-term range between 1000 and 50000 years of storage, the Pu in the burner fuel and the Pu generated via decay of Cm244 gives rise to an increase of the actinide hazard of 50%.

The incineration ability could be improved

- in the long-term range by a reduction of the reprocessing losses of Am,
- in the medium-term range by a separation of Cm and a subsequent use of the daughter Pu as fuel.

It is concluded that an enforced research should be done in chemistry to prove that a satisfactory separation especially of Am and also of Cm is possible on large scale.

One large fast reactor of type EFR is able to clean-up six 1300MW_e PWRs in equilibrium in case of 2.8% LWR and recycled Np and Am homogeneously admixed to the core fuel. Then the fraction of Pu238 in the core Pu is limited to 6%. Larger admixtures seem to be possible in smaller cores with respect to safety parameters [4], but then the content of Pu238 will strongly increase causing problems with heat and radiation during fabrication of recycle fuel.

ACKNOWLEDGEMENT

The authors gratefully acknowledge the numerous fruitful discussions with Dr.H.Küsters.

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CHARACTERISTICS OF TRU TRANSMUTATION IN AN LMFBR

T. WAKABAYASHI, M. YAMAOKA

Oarai Engineering Center,
Power Reactor and Nuclear Fuel
Development Corporation,
Oarai-machi, Ibaraki-ken, Japan

Abstract

Feasibility studies of TRU transmutation in an LMFBR has been performed to establish TRU (exactly minor actinides) transmutation technology by in an LMFBR. Systematic parameter survey calculations were performed for a conventional 1000MWe LMFBR core to investigate basic characteristics of TRU transmutation in an LMFBR core and also to establish TRU loading method which has no serious influence on core design. TRU loading resulted in a decrease of the burnup reactivity loss, which is a desirable feature for the extension of reactor operation period. TRU transmutation rate reaches approximately 11% per cycle and the amount of the TRU transmutation with TRU ratio of 5% is almost six times as much as that of the TRU from a 1000MWe-class LWR. Homogeneous TRU-loading has almost no serious influence on core performances. However, it is desirable to limit the TRU loading ratio to several percent from the aspect of reactor operational safety. Heterogeneous TRU-loading method resulted in a larger power swing. Further optimization is necessary to clarify the feasibility of the method.

Study on an innovative core concept for TRU transmutation was also carried out taking into account the results of systematic parameter survey calculations. Neutronic feasibility was studied on a 1000MWe Super Long-Life Core (SLLC) with no need of fuel exchange during plant life. The SLLC is an attractive option for transmuted TRU nuclides since it can transmute them with confining them into the reactor during plant life. It was found that remarkable reduction of both reactivity change and power variation during burnup is possible by optimizing the amount and zoning of TRU loaded. The reactivity change and the power swing during 30 years of the optimized core are less than half of those of the SLLC with no TRU loaded. The amount of TRU transmuted is about 10ton which corresponds to the amount of TRU produced by 13 LWRs. The SLLC loaded with TRU fuel was found feasible from the neutronic point of view.

R&D programs are being pursued in PNC-Japan to establish the transmutation technology of TRU nuclides in LMFBRs.

I. INTRODUCTION

There is a strong social requirement not to leave the radiological hazardous material form the use of nuclear power to future generations. Some of the transuranic (TRU) nuclides contained in residual waste from reprocessing have extremely long-term radiotoxicity(1). There are some means of reducing radiotoxicity of the TRU nuclides under investigation(2),(3),(4). The TRU nuclides produce useful energy when converted into short-lived fission products by neutron bombardment. From this standpoint, a nuclear reactor provides an extremely rational means for transmutation of TRU nuclides. Among the various nuclear reactors, the sodium-cooled liquid metal fast breeder reactor (LMFBR), now under development, can be used for transmutation of many TRU nuclides, because of the possible nuclear fission generated by high-energy neutrons.

The following studies are implemented to establish TRU (exactly minor actinides) transmutation technology by LMFBRs in PNC-Japan:

- Feasibility studies of TRU transmutation by LMFBR and evaluation of TRU material balance
- Nuclear data evaluation of TRU nuclides in sample irradiation experiments
- Measurement and evaluation of physical and chemical properties of TRU compounds
- Development of fabrication technology of TRU fuel
- Evaluation of fuel behavior by TRU pin irradiation experiments

This paper shows the results of the following two feasibility studies of TRU transmutation in an LMFBR:

- (1) Study on core characteristics of an LMFBR loaded with TRU fuel.

In this study, systematic parameter survey calculations have been performed for a conventional 1000MWe-class LMFBR core. The purpose is (i) to investigate basic characteristics of TRU transmutation in an LMFBR core and (ii) to establish TRU loading method which has no serious influence on core design.

- (2) Study on feasibility of a Super-Long-Life-Core (SLLC) loaded with TRU fuel which has no need of fuel exchange during plant life

Taking into account the advantage of the TRU-loaded core, a 1000MWe SLLC with no need of fuel exchange during plant life is proposed as an LMFBR core for TRU transmutation. The SLLC is an attractive option

transmuting TRU nuclides since it can transmute them with confining them into the reactor during plant life. A feasibility of the SLLC was studied form the neutronic view point.

The subsequent two section describes the main results of the studies. The last section shows the R&D program in PNC for the establishment of TRU transmutation technology in an LMFBR.

II. STUDY ON CORE CHARACTERISTICS OF AN LMFBR LOADED WITH TRU FUEL

1. Calculational method

First, a 1000MWe-class LMFBR core with MOX fuel was defined as the reference whose main parameters are shown in Table 1 and Fig. 1. The nuclear characteristics of TRU-loaded core was calculated by two-dimensional diffusion

Table 1 Design Parameters of Conventional 1000MWe LMFBR Core

Parameters	Data
Thermal Power	2520(MW)
Operation Cycle Length	456(EFPD*)
Fuel Material	PuO ₂ /UO ₂
Number of Subassemblies	
Inner Core/Outer Core	175/180
Radial Blanket	72
Control Rods	24
Core Height	100(cm)
Equivalent Core Diameter	368(cm)
Axial Blanket Thickness	30(cm)
Number of Fuel Batch (Core/Blanket)	3/4
Fuel Pin Diameter	0.83(cm)
Assembly Lattice Pitch	17.98(cm)
Volume Fraction	
Fuel/Coolant/Structure	41.6/37.5/20.9(%)

*Effective Full Power Days

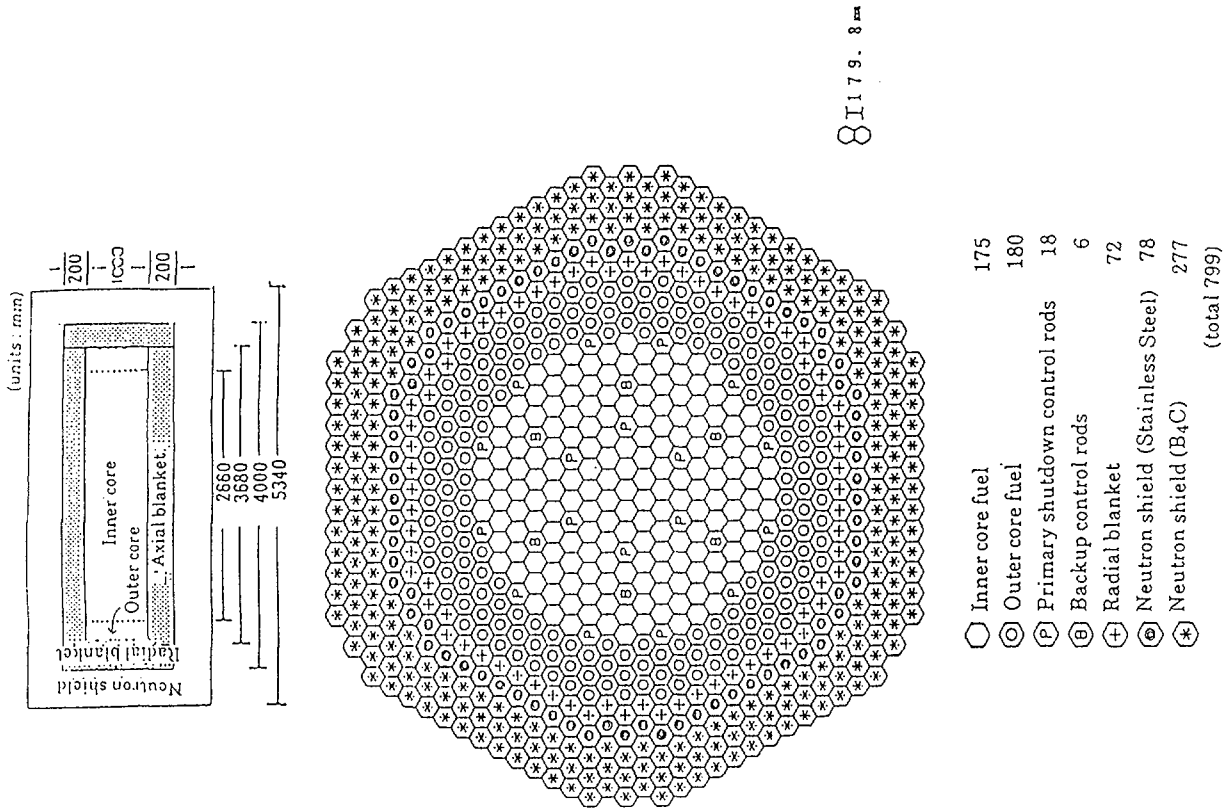


Fig. 1 Core Layout of Conventional 1000 MWe LMFBR

theory with depletion chain. The cross sections used were effective seven group constants condensed from the Japanese standard 70-group constant set, JFS-3-J2(5), which is based on an evaluated nuclear data library, JENDL-2(6).

TRU fuel was assumed to come from LWR spent fuel with five-year cooling time before reprocessing. The isotopic composition of the TRU shown in Table 2 was calculated by the ORIGEN2 code(7).

2. Survey of TRU loading ratio

Influence of TRU loading ratio was analyzed with TRU homogeneously dispersed in the core. The main results were shown below.

Figure 2 shows the relationship between the amount of TRU transmuted and that of loaded TRU in the FBR core. At least, one percent of TRU loading to fuel would be needed to eliminate TRU, and the amount of TRU transmuted increases linearly with that of loaded TRU. Since a 1000MWe-class LWR produces about 26 kg of TRU per year, an LMFBR with 5% TRU-loading can transmute the TRU mass from six LWRs in rough estimation.

As seen in Fig. 3, TRU loading to core results in a significant decrease of burnup reactivity loss mainly due to ^{238}Pu build-up. However, the burnup reactivity loss becomes positive when the amount of loaded TRU exceeds 10%.

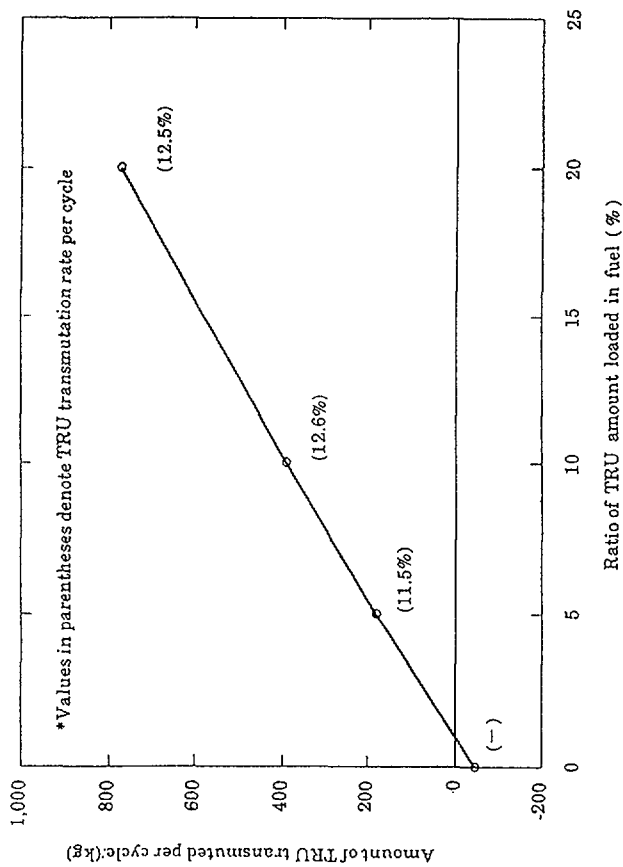


Fig. 2 Relationship between Amount of TRU Transmuted and Ratio of TRU Amount loaded in Fuel

Therefore, the maximum amount of loaded TRU would be limited to several percent from the aspect of reactor operational safety. A proper amount of TRU might be advantageous to the extension of reactor operation period.

It was found that TRU loading tends to increase the burnup change of the global power distribution. However, it could be possible to minimize the power change by optimize the ratio of TRU amount between the inner core and the outer core.

3. Survey of TRU loading methods

We consider here two typical TRU-loading methods in the LMFBR core. One is the method dispersing TRU homogeneously throughout the entire core (homogeneous TRU-loading method), which is expected not to affect the core characteristics seriously. The other possible method is the use of small number of subassemblies (target S/As) which concentrate TRU in MOX fuel (heterogeneous TRU-loading method), which can have the advantage of the fuel cycle because of handling the small number of TRU-included fuels in fabrication factory.

Table 2 Composition of TRU Fuel *

Nuclide	Weight Fraction (%)
Np-237	49.1
Am-241	30.0
Am-242m	0.08
Am-243	15.5
Cm-243	0.05
Cm-244	5.0
Cm-245	0.26

* Discharged from PWR (35GWd/t) and Cooled for 5 Years Before Reprocessing

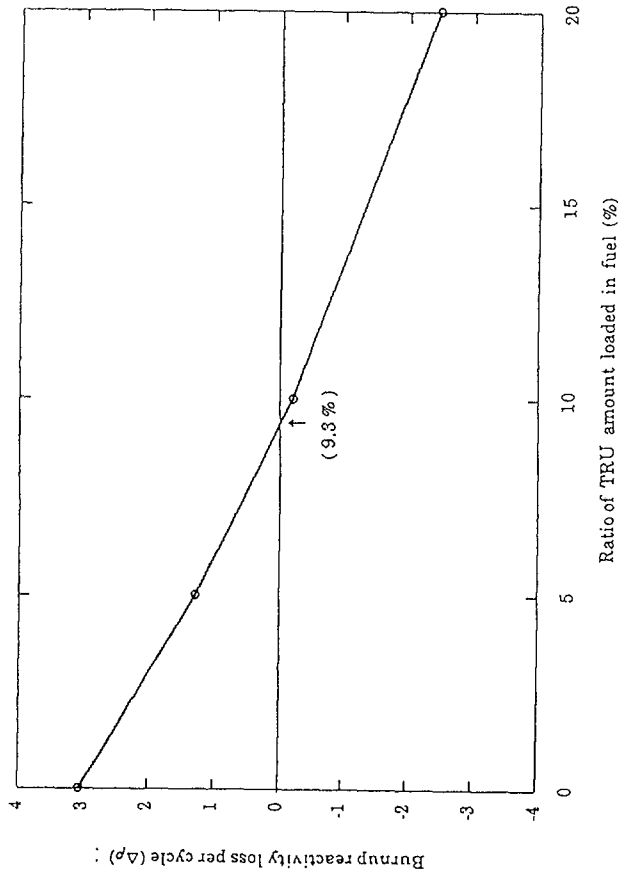


Fig. 3 Relationship between burnup reactivity loss and ratio of TRU amount loaded in fuel

A typical TRU-loading core was assumed for each of the homogeneous and heterogeneous TRU-loading methods. The total mass of loaded TRU is set to be approximately identical between them, that is, 5% TRU in the whole core fuel in the case of the homogeneous method, and 37 target S/As which have 50% of TRU in fuel in the case of the heterogeneous method. Figure 4 shows the loading pattern of TRU-loaded S/As in the heterogeneous method. A core with no TRU loaded was also analyzed as the reference case.

Table 3 compares the calculated results about the nuclear characteristics of the two TRU-loaded cores and the reference core.

The maximum linear heat rate of each TRU-loaded core is not so much different with that of the reference core. The radial power distribution of the heterogeneous TRU-loaded core, however, is quite different from that of the other cores as shown in Fig. 5. At beginning of the equilibrium cycle, the power of TRU-loaded region is very depressed compared with the reference core, while they get close at end of the

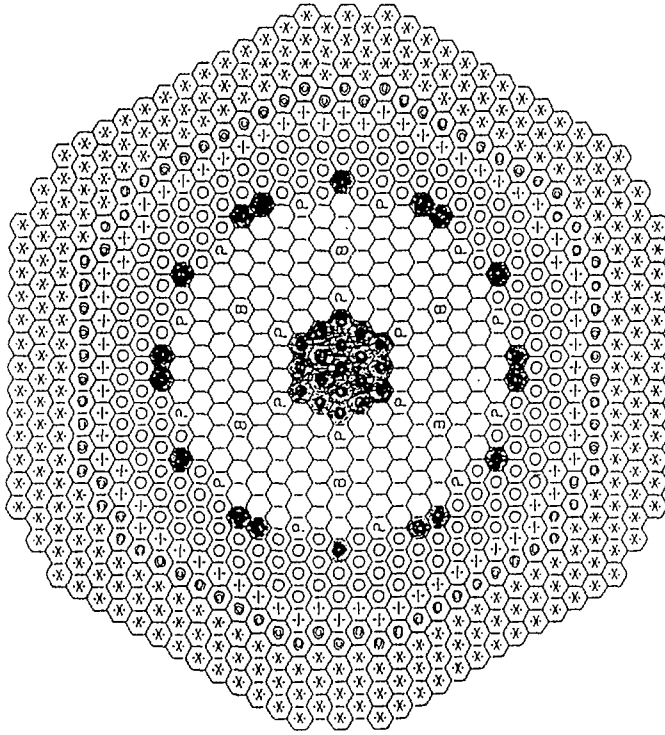


Fig. 4 Loading pattern of target S/As (Heterogeneous TRU-loaded core)

equilibrium cycle. This power swing would be a great obstacle for thermal characteristics. Slight decrease of the maximum linear heat rate in the heterogeneous TRU-loaded core is attributed to TRU-loading in the region where the peak power occurs if no TRU is loaded.

The control rod worth of the TRU-loaded cores decreases from the reference core by 10~20%. This may be caused by the hardening of neutron spectrum.

The burnup reactivity in the TRU-loaded cores is 40% smaller than that of the reference core because of the production of ^{238}Pu from ^{237}Np in TRU fuel.

The Doppler coefficients of the TRU-loaded cores are 20~30% smaller in absolute value, and the sodium density reactivity coefficients are 50% larger than the reference core because of the spectrum hardening.

Table 3 Comparison of Nuclear Characteristics of TRU-Loading Cores

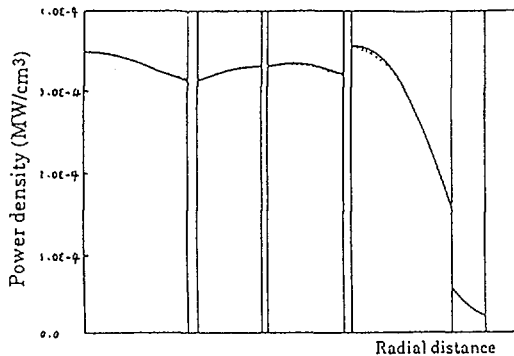
Item	Reference Core (No TRU-loaded)	Homogeneous TRU-Loading Core	Heterogeneous TRU-Loading Core
Pu Enrichment (Inner Core/Outer Core)	15.3/19.3 wt%	16.2/19.6 wt%	15.7/19.7 wt%
Max Linear Heat Rate Inner Core Outer Core	(BOEC/EOEC) 380/419 w/cm 420/357 w/cm	(BOEC/EOEC) 376/431 w/cm 416/355 w/cm	(BOEC/EOEC) 386/428 w/cm 439/342 w/cm
Burnup Reactivity Loss	3.3%Δk/kk'	1.9%Δk/kk'	1.8%Δk/kk'
Control Rod Worth (BOEC 33cm Insertion of Primary Rods)	1.7%Δk/kk' (1.00)*	1.5%Δk/kk' (0.88)*	1.5%Δk/kk' (0.88)*
Doppler Coefficient	-1.1×10 ⁻² Tdk/dT	-7.1×10 ⁻³ Tdk/dT	-7.4×10 ⁻³ Tdk/dT
Coolant Density Coefficient (Δp/p/100%Density Change)	-1.7×10 ⁻²	-2.5×10 ⁻²	-2.7×10 ⁻²
β _{eff}	3.7×10 ⁻³	3.5×10 ⁻³	3.3×10 ⁻³
Amount of TRU transmuted per cycle	-46kg	184kg (11.7%)	176kg (10.6%)

*) Values in Parentheses Denote Relative Control Rod Worth

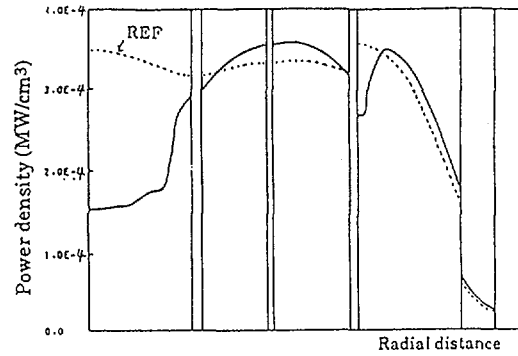
The values of prompt neutron life time and effective delayed neutron yield in the TRU-loaded cores is smaller by 20% and 10%, respectively, than those of the reference core.

Both TRU-loaded cores can transmute TRU by 11~12%/cycle (about 180kg/cycle), and there is no difference between the two TRU loading methods.

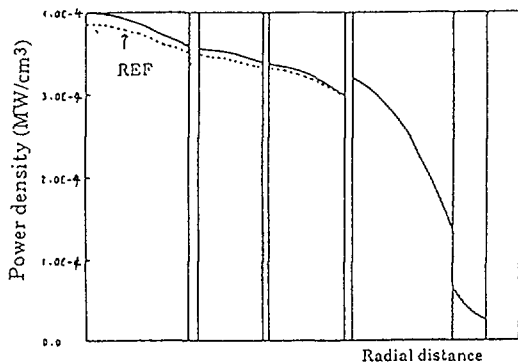
The maximum cladding temperature of each TRU-loaded core was evaluated on condition that the coolant flow distribution in the core is the same as that of the reference core. There is no thermal problems about the homogeneous TRU-loaded core, since the power distribution hardly changes from the reference core. On the



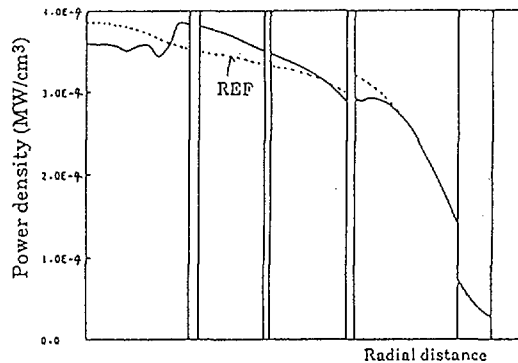
Radial power distribution of homogeneous TRU-loaded core at BOEC (Primary control rods partially inserted)



Radial power distribution of heterogeneous TRU-loaded core at BOEC (Primary control rods partially inserted)



Radial power distribution of homogeneous TRU-loaded core at EOEC (Primary control rods fully withdrawn)



Radial power distribution of heterogeneous TRU-loaded core at EOEC (Primary control rods fully withdrawn)

Fig. 5 Radial power distribution of TRU-loaded cores

other hand, the position where maximum temperature of cladding occurs in the heterogeneous TRU-loading core moves from the reference core, and the hot spot temperature of the cladding is 40 degrees-C higher than that of the reference core. Although there might be rooms for optimization of the flow distribution, the significant power swing is a disadvantage to thermal characteristics.

However, this problem may be overcome by using target S/As with a smaller amount of TRU or loading them in a more scattered manner. Further optimization is necessary to clarify the feasibility of the method.

4. Conclusion

TRU loading results in a decrease of the burnup reactivity loss, which is a desirable feature for the extension of reactor operation period. TRU transmutation rate reaches approximately 11% per cycle and the amount of the TRU transmutation with TRU ratio of 5% is almost six times as much as that of the TRU from a 1000MWe-class LWR. Homogeneous TRU-loading has almost no serious influence on core performances. However, it is desirable to limit the TRU loading ratio to several percent from the aspect of reactor operational safety. Heterogeneous TRU-loading method resulted in a larger power swing. Further optimization is necessary to clarify the feasibility of the method.

III. STUDY ON POSSIBILITY OF A SUPER-LONG-LIFE-CORE (SLLC) LOADED WITH TRU FUEL

1. Calculational model

The feasibility of the 1000 MWe Super-Long-Life-Core (SLLC) loaded with TRU fuel was studied. The design parameters are shown in Table 4. The core reactivity life should be 30 years (effectively) in order to accomplish reactor operation without fuel exchange during plant life time. In this study, the amount and the region of TRU loading were surveyed so that burnup change of reactivity and power distribution could be minimized assuming that the dimensions of the core and the fuel are the same as those of the core as an SLLC with mixed oxide fuel(8). The core layout is shown in Fig. 6. A large fuel pin of 12mm diameter is employed in order to enhance the internal breeding, which resulted in a fuel volume ratio of 50%. The core volume is chosen to be about 4 times larger than that of the conventional core with the same power output so that the average fuel burnup during life time is about 200GWd/t.

Table 4 Design Parameters of 1000MWe Super long-Life LMFBR Core

Parameters	Data
Thermal Power	2520(MW)
Core life	30(EFFY*)
Fuel Material	PuO ₂ -UO ₂ -(Np,Am,Cm)O ₂
Number of Subassemblies Core/Blanket/Control Rods	462/84/49
Core Height	180(cm)
Equivalent Core Diameter	494(cm)
Axial Blanket Thickness	15(cm)
Fuel Pin Diameter	1.22(cm)
Assembly Lattice Pitch	20.8(cm)
Volume Fraction Fuel/Coolant/Structure	53.0/24.7/22.3(%)
Pu Isotopic Ratio (²³⁹ Pu/ ²⁴⁰ Pu/ ²⁴¹ Pu/ ²⁴² Pu)	58/24/14/4(w/o)

*Effective Full Power Year

Core neutronics calculations were done by the two-dimensional RZ burnup code based on the diffusion theory using 7-group effective cross sections collapsed from JFS-3-J2 library(5) based on JENDL-2(6). The isotopic composition of the TRU is the same as that used in the study of section II (Table 2). The core performances of the optimized core were compared with those of the SLLC with mixed oxide fuel.

2. Results of parametric survey

Figure 7 illustrates the effect of the TRU loading ratio on the variation of the effective multiplication factor (k_{eff}) during burnup with uniform TRU-loading for whole core (See (1) of Fig. 7). If no TRU is loaded, the reactivity change during 30 effective full-power years (EFFY) is over 10%Δk and the reactivity life is about 25 EFFY. (The reactivity life could be extended more by increasing the plutonium

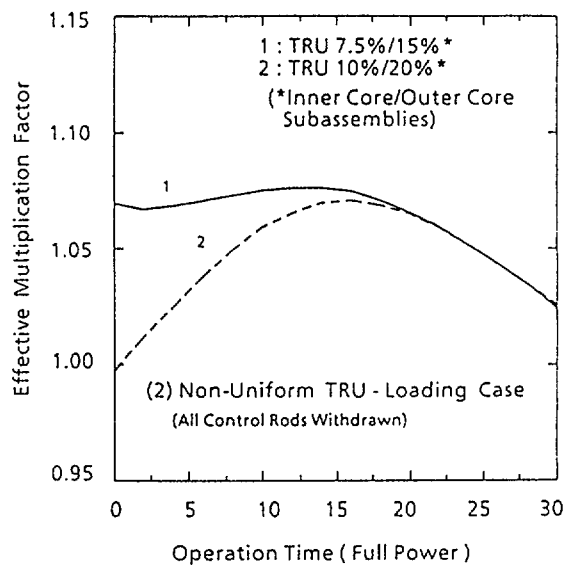
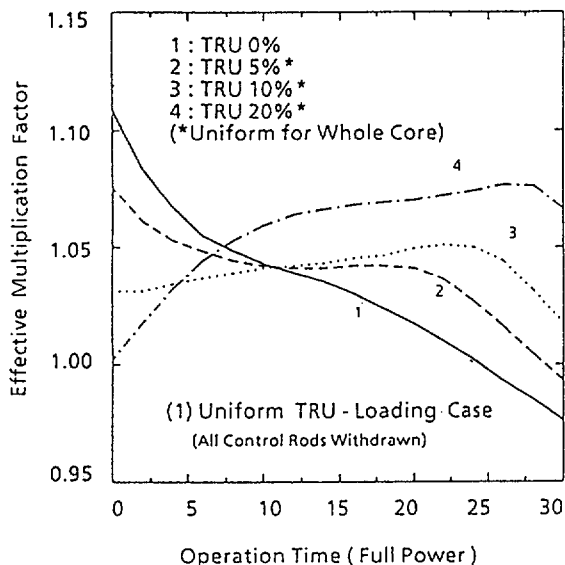


Fig. 7 Reactivity Change of Super Long Life Core due to Burnup

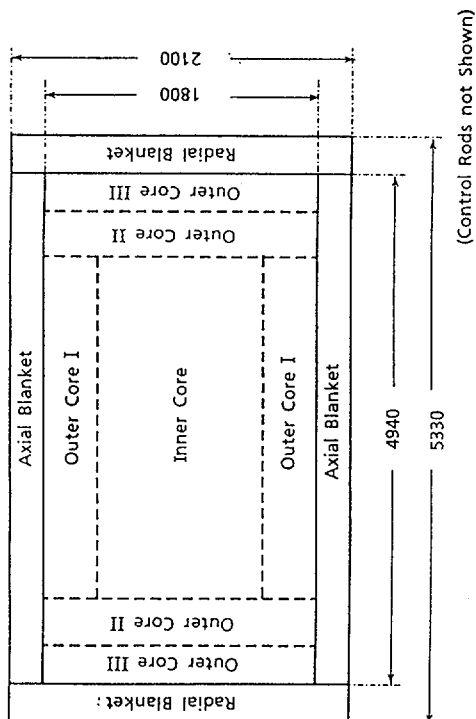


Fig. 6 Core Layout of Super Long Life Core (Vertical View)

enrichment. However, it would increase reactivity change.) TRU loading in the core resulted in significant reduction of reactivity change. The figure shows that TRU loading ratio of about 10% makes reactivity change of about $5\% \Delta k$ possible.

Figure 8 illustrates the variation of the power distribution during burnup for the core with no TRU loaded and that with uniform TRU loading of 10% for whole core. (See cases 1 and 2 of Fig. 8) The figure shows that uniform TRU-loading for whole core resulted in significant variation of the power distribution during burnup. This means that the uniform TRU-loading for whole core accelerates the power shift due to burnup from the outer core to the inner core. It seems that this effect became conspicuous due to the very long core operation time.

Loading TRU in the outer core subassembly more than in the inner core sub-assembly is considered one of the effective method for suppressing variation of the power distribution during burnup. Figure 8 illustrates the variation of the power distribution during burnup for the core with TRU loading ratio of 7.5% and 15% for the inner core and the outer core, respectively. (See case 3 of Fig. 8). This non-uniform loading of TRU fuel resulted in very constant power distribution during burnup. Figure 7 illustrates the reactivity change of the core with non-uniform TRU-loading core (See (2) of Fig. 7). From the results shown above, we finally

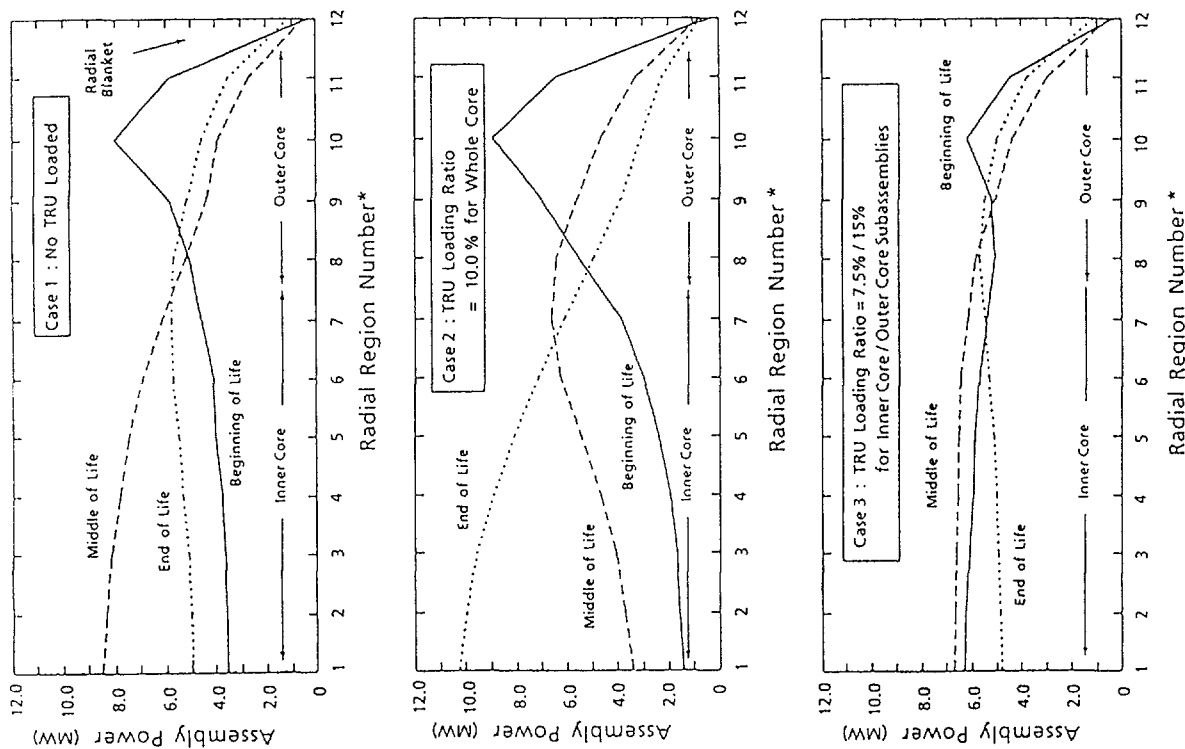
Table 5 Core Performances of Super Long-Life Core

	No TRU Loaded Core	TRU Loaded Core
Loading ratio of TRU [w/o]	-	7.5/15.0*
Reactivity Change [%Δk]	13.3	5.2
Average Power Density [w/cc]	79/75**	80/75**
Max. Linear Heat Rate [w/cm]	268	266
Power Swing*** [%]	21.4	47.2
Fuel Burnup [GWd/t]	188/262 (Average/Max.)	187/292 (Average/Max.)
Max. Fast Neutron Fluence [nvt]	6.9 × 10 ²³	6.6 × 10 ²³
Max. Cladding Temp. [°C]	not calculated	655
Sodium Void Reactivity [Δk]	0.014/0.030**	0.026/0.031**
Doppler Coefficient [-1 × 10 ⁻³ Tdk/dt]	8.3/4.7**	4.6/2.7**

*Inner core assembly / Outer core assembly
 **Beginning of life / End of life
 *** Average of maximum power change of each assembly during life time

selected the TRU loading ratio of 7.5% and 15% for the inner core and the outer core, respectively. The reactivity change during 30 EFPY is about 5%.

The results of parametric survey show that remarkable reduction of both reactivity change and power variation during burnup is possible by optimizing the amount and zoning of TRU loaded.



(*Control rod regions not included)

Fig. 8 Variation of Assembly Power Distribution of Super Long Life Core due to Burnup

3. Analysis of nuclear characteristics

Nuclear characteristics for the optimized core were analyzed and compared with those of the core with no TRU fuel. The results are summarized in Table 5. In this study, no control rod strategy during burnup was considered and the reactor calculations were performed with all the control rods withdrawn from the core.

The reactivity change of the TRU-loaded SLLC during 30 years (EFPY) is about 5% and it is less than half of that of the SLLC with no TRU loaded. The values of the maximum linear heat rate for both of the SLLCs are about 270w/cm. They would increase to some extent if control rods are inserted in order to suppress the excess reactivity. The average fuel burnup is about 190GWd/t and the maximum fast neutron fluence is about 7×10^{23} nvt. This irradiation condition is almost the same as the target of a commercial FBR core.

The maximum cladding temperature was evaluated on condition that the reactor outlet and inlet temperatures are 530 and 375 °C, respectively. Although it is preliminary since no control rod insertion effect is considered in the calculation of the power distribution, the maximum cladding temperature is low enough compared with its limit value (typically, 700 °C).

Due to TRU loading, the sodium void reactivity increased and the absolute value of the Doppler coefficient decreased. The safety aspect of the SLLC loaded with TRU is one of the problem to be investigated, although a lot of parameters other than these parameters also contribute to the core safety.

Table 6 shows mass balance of TRU. Total loading amount of Np, Am, and Cm is about 17ton and 10ton of it (about 60% of loading amount) is transmuted during the 30-year reactor operation. Transmuted amount of Np, Am, and Cm is about 340 kg per year, which is nearly equal to the amount of those from 13LWRs of the same power output. Total amount of ²³⁹Pu and ²⁴¹Pu is slightly increased during operation. Thus, the SLLC has an advantage of transmuted large amount of TRU with preserving plutonium fissile material.

4. Conclusion

It was found that remarkable reduction of both reactivity change and power variation during burnup is possible by optimizing the amount and zoning of TRU loaded. The reactivity change and the power swing during 30 years of the optimized core are less than half of those of the SLLC with no TRU loaded. The amount of TRU transmuted is about 10ton which corresponds to the amount of TRU produced

Table 6 TRU Mass Balance of Super Long-Life Core
(unit: ton)

	Loaded amount (A)	Discharged amount (B)	Difference (B-A)
U	159	133	-26
Pu	20.4	25.5	5.1
(²³⁸ Pu)	(0)	(3.5)	(3.5)
(Pu Fissile)*	(14.5)	(14.8)	(0.3)
Np	8.4	2.6	-5.8
Am	7.8	3.5	-4.3
Cm	0.9	1.0	0.1
Np+Am+Cm	17.2	7.1	-10.1

*²³⁹Pu + ²⁴¹Pu

by 13 LWRs. The SLLC loaded with TRU fuel was found feasible from the neutronic point of view.

IV. R&D ISSUES FOR TRU TRANSMUTATION IN AN LMFBR

Shown in Fig. 9 is the R&D programs in PNC-Japan to the transmutation technology of TRU nuclides in LMFBRs.

Through design study of an LMFBR transmuted TRU and evaluation of TRU mass balance, it is planned to establish appropriate core design concepts for a large-size LMFBR and strategy of their introduction. Basic researches including nuclear data evaluation, and fuel property measurement and evaluation are also programmed.

"Joyo" an experimental FBR, is planned to be used both for initial transmutation experiments irradiating fuel samples, and for oxide fuel irradiating tests and post-irradiation tests. In addition, basic tests are planned on fuel preparation of TRU obtained from the partitioning of nuclides, the results of which will be reflected in fuel fabrication technology.

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JAPANESE FISCAL YEAR ITEM	1991	1992	1993	1994	1995
(1) Design Study and Material Balance	Feasibility Study	Material Balance Evaluation	Conceptional Design		
(2) Nuclear Data Evaluation	Irradiated Np Foil Analysis	Sample Preparation (Np, Am, Cm)	Sample Irradiation in Joyo	Analysis	
(3) Fuel Property Measurement and Evaluation	Survey	Preparation		Measurements (Np)	
(4) Fabrication Technology Development	Survey		Design	Cell Modification	Trial Fabrication
(5) Irradiation Test	Preparation	Fuel (contained ²⁴¹ Am)	PIE	Analysis	Preparation of TRU (Np, Am) Fuel Pin

Fig. 9 R&D Schedule of TRU Transmutation by LMFBR in PNC-Japan

EXPERIMENTAL INVESTIGATIONS CONCERNING THE PROBLEM OF MINOR ACTINIDE TRANSMUTATION

S.M. BEDNYAKOV, V.A. DOULIN, I.P. MATVEENKO, A.M. TSIBULYA, A.V. ZVONAREV
 Institute of Physics and Power Engineering, Obninsk, Russian Federation

Abstract

A systematic investigation of MA transmutation started at BFS assemblies in 1991. The first experiments were carried out using absolute calibrated fission chambers and reactivity perturbations by a small MA sample.

INTRODUCTION

The problem of minor actinides (MA) transmutation can be solved by recycling of spent nuclear fuel in fast reactors. The evaluation of period and parameters of recycle requires validation fission and capture cross-sections of MA in a wide range of neutron energies. The achieved and required (in parenthesis) accuracies of neutron cross-sections are:

	$\delta\sigma_f, \%$	$\delta\sigma_c, \%$	$\delta\sigma_{in}, \%$
Np-237	7(5)	15(5)	30(10)
Pu-238	10(5)	25(10)	40
Pu-240	10(10)	20(4)	20(15)
Pu-241	8(5)	15(7)	20
Am-241	10(5)	10(5)	30(10)
Am-243	10(5)	30(5)	30
Cm-244	10(5)	30	30

One of the first wide set of experiments on a critical assemblies on this problem was carried out in FCA (JAERI) /1/. Analysis of these experiments /2/ showed an agreement of micro- and macroexperiments for Np-237, Pu-238, Am-241 and Am-243

fission cross-sections (~5% in the neutron energy range of 0.5-10.0 Mev). But the central reactivity worth (CRW) measurements were not adequate to identificate capture cross-sections to a required accuracy.

A systematic investigation of MA transmutation problem started at 50C assemblies in 1991. The first experiments were carried out using absolute calibrated fission chambers and reactivity perturbations by a small MA samples.

ANALYSIS OF FCA (JAPAN) AND 50C (RUSSIA) EXPERIMENTS

The FCA results on CRW of Pu-240 were compared with similar 50C ones. Parameters of assemblies are given in Table I.

TABLE I. PARAMETERS OF ASSEMBLIES

Assembly	Fuel	Enrichment %	Neutron percentage below 10 keV in spectrum	$\frac{\sigma_{10}^{10}}{\sigma_{235}^{235}}$
E0G-45A-I	UO ₂	21	4,7	-1,10
E0C-49-2	(Pu-U)O ₂	12	13,1	-0,92
E0C-49-4	(Pu-U)O ₂	12	6,7	-0,72
E0C-51-I	U	36	0,4	-0,72
E0C-55-I	Pu-U	10	2,4	-0,60
KEP-10	UO ₂	90	16,4	-1,54
F CA-IX-I	U	93	17,6	-
F CA-IX-2	U	93	9,1	-
F CA-IX-3	U	93	5,0	-
F CA-IX-4	U	93	0,9	-
F CA-IX-5	U	93	0,4	-
F CA-IX-6	U	93	0,1	-
F CA-IX-7	U	20	0,04	-
E0G-61	Pu-U	15	4,0	-0,73
E0C-54-4	UO ₂	22	4,6	-1,08
E0C-56-IA	(Pu-U)O ₂	16	6,6	-0,76
E0C-56-IB	(Pu-U)O ₂	14	5,5	-0,70

a) Calculational CRW ratios of ¹⁰B to ²³⁵U

TABLE II. ASSESSMENT OF EXPERIMENTS ON NP-237 TO PU-239
CRW RATIOS FOR SOME ASSEMBLIES (ρ^{237}/ρ^{239})

Assembly	Experiment (E)	Calculations, EHAE-90 (C)			C-E	
		NL-C		NL ₂₃₇ -0		
		hom.	het.	het.		
FCA-IX-1	-0.865	-0.896	-1.05	-0.811	-0.744	0.121
FCA-IX-2	-0.242	-0.184	-0.248	-0.196	-0.183	0.059
FCA-IX-7	0.117	0.188	0.187	0.194	0.187	0.070

B. EOC-54-4:

	ρ^{237}/ρ^{239} (I=0)	Corrections		Medium heterogeneity
		Self-shielding for $NI(b^{-1})$	0.00456	
Experiment	-0.220 [±] ±0.007	0.00220	0.00283	-
Calculation	-	-0.005	-0.006	-0.027

The experiments carried out at EOC are shown in Table IV. The comparison of experiments and calculations is presented in Table V.

The results on CRW and fission cross-sections ratios for Np-237 to Pu-239 ($\sigma_f^{237}/\sigma_f^{239}$, ρ^{237}/ρ^{239}) of EOC are close to similar results of FCA. There are also contradictions for Pu-238 and Am-243 to Pu-239 fission cross-sections ratios ($\sigma_f^{1}/\sigma_f^{239}$).

EXPERIMENTAL RESULTS ON EP-1 AND EH-350

A set of MA cross-sections ratios to U-235 fission cross-sections was measured in the center of EP-1 plutonium critical as-

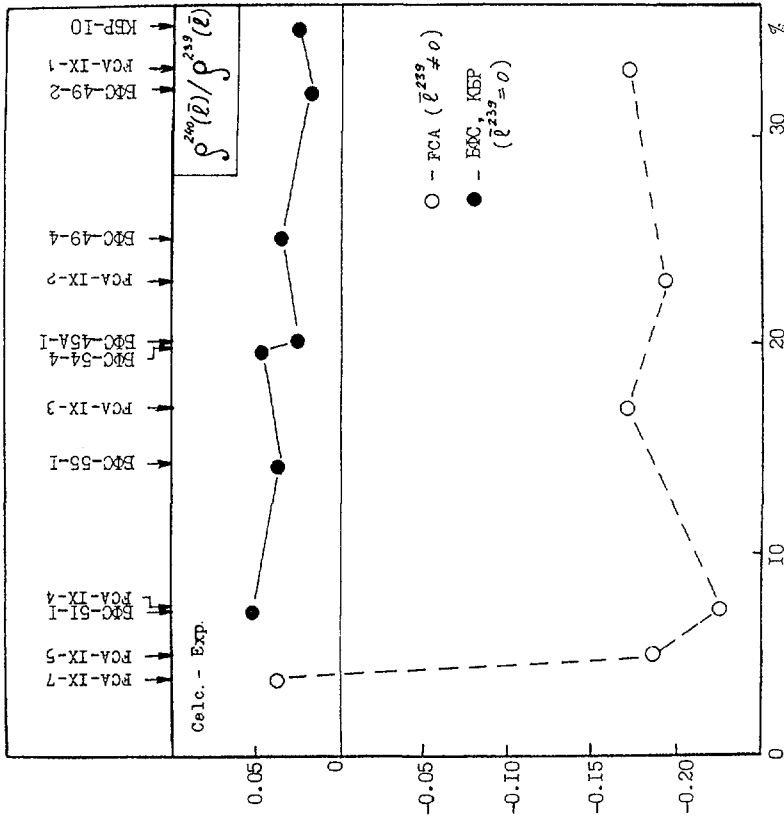


Fig. 1. Central reactivity worths of ²⁴⁰Pu in FCA, EOC, KEP.
Horizontal axis - percentage of neutrons below 46.5 keV in assembly spectrum

Sets of absolute discrepancies between calculations and experiments on these assemblies contradict each other (see Fig. 1). Two causes could clarify the discrepancy. The first is a hydrogen inside FCA dioxide sample. The second is an uncertainty of self-shielding corrections for thicker EOC metallic sample. An illustrative information on this analysis is given in Tables I-III and Figures.

TABLE III. ASSESSMENT OF EXPERIMENTS ON PU-240 TO PU-239 CRW RATIOS FOR SOME ASSEMBLIES (ρ^{240}/ρ^{239})

Assembly	Experiment (E)	Calculations, EHA5-90 (C)			C-E	
		N1-C		N1-90 = 0.0052 b ⁻¹		
		hom.	het.	N1 ₂₃₉ = 0 N1 ₂₄₀ = 0.0052 b ⁻¹		
FCA-IX-1	0.253	-0.112	-0.400	0.071	0.065	-0.188
FCA-IX-2	0.472	0.271	0.189	0.236	0.221	-0.251
FCA-IX-7	0.307	0.338	0.331	0.346	0.333	0.026

B. E8C-54-4 and E8C-45A-1:

E8C-54-4	C ²⁴⁰ / C ²³⁹ (t=0)	Corrections			Medium heterogeneity
		Self-shielding for N1(b ⁻¹)	0.0297	0.0548	
E	0.209± ±0.006	-0.023	-0.029	-0.046	-
C	-	-0.015	-0.016	-0.020	-0.008
E8C-45A-1:					
E	0.232± ±0.010	-	-	-	-
C	-	-	-	-0.058	-0.015

assembly (average neutron energy 1 Mev). The experimental techniques used glasses, activation detectors and layers. Some results of these experiments and comparison with Monte-Carlo calculations on the basis of EHA5-78 library are given in Table VI.

There were many measurements at E8C-350 aimed to check neutron cross-sections of MA in spectra of low enrichment zone,

TABLE IV. EXPERIMENTS AT E8C ASSEMBLIES NORMALIZED BY U-235 AND PU-239

Assembly Nuclide	E8C-54-4	E8C-56-1A	E8C-56-1B	E8C-61
238 Pu	frr ^a	frr	-	frr
240 Pu	frr crw ^b	frr, crw	crw	frr
241 Pu	frr, crw	crw	crw	crw
242 Pu	frr	-	-	frr
237 Np	frr, crw	frr, crw	crw	frr, crw
241 Am	frr	crw	crw	-
243 Am	frr	frr	-	frr
244 Cm	frr	frr	-	frr

a) Reaction rate ratio $G_f^i/G_f^{235, 24239}$
 b) Central reactivity worth ratio ρ^i/ρ

high enrichment zone, plutonium subassembly between them and metallic uranium subassembly in low enrichment zone. The results are presented in Table VI.

Other samples of Am-241, Cm-244, Pu-238, Pu-240 are irradiated at E8C-350 now.

THE PROGRAMME OF EXPERIMENTS PLANNED FOR NEAR FUTURE

We plan a set of measurements directed to validation of fission and capture cross-sections of MA at critical assemblies. A set of small samples, fission chambers and other detectors with MA will be used. A wide range of neutron spectra from the hardest (EP-1) to more softer than E8C-350 is planned to be arranged.

TABLE V . COMPARISON OF EXPERIMENTS AND CALCULATIONS

Ratio	C/E	
	50C-54-4	50C-61
$\frac{\rho_{f,238}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	0,67	0,62
$\frac{\rho_{f,240}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	1,02	1,01
$\frac{\rho_{f,241}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	1,00	0,98
$\frac{\rho_{f,242}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	0,98	0,95
$\frac{\rho_{f,237}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	1,02	1,01
$\frac{\rho_{f,244}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	1,20	-
$\frac{\rho_{f,243}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	0,69	0,65
$\frac{\rho_{f,244}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	0,96	0,96
$\frac{\rho_{f,240}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	0,084	-
$\frac{\rho_{f,241}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	1,01	1,03
$\frac{\rho_{f,237}}{\rho_f} / \frac{\rho_{f,235}}{\rho_f}$	0,126	0,074

a, b) For the marked ratios the difference C-E is given

Some experiments carried out with these chambers and samples at 50C-54-1 (with plutonium inner core) and 50C-61 (a mock-up of fast reactor with Pb coolant) assemblies are analysed. A next step of research is connected with active cores containing a large quantity of MA. It will permit to validate capture and inelastic cross-sections. It will also give a possibility to found parameters of cores of reactors for burning MA in homogeneous and heterogeneous compositions.

We plan to have at 50C 10-15 kg of Np-237 dioxide at the end of the year. This amount is expected to increase to 50-100 kg.

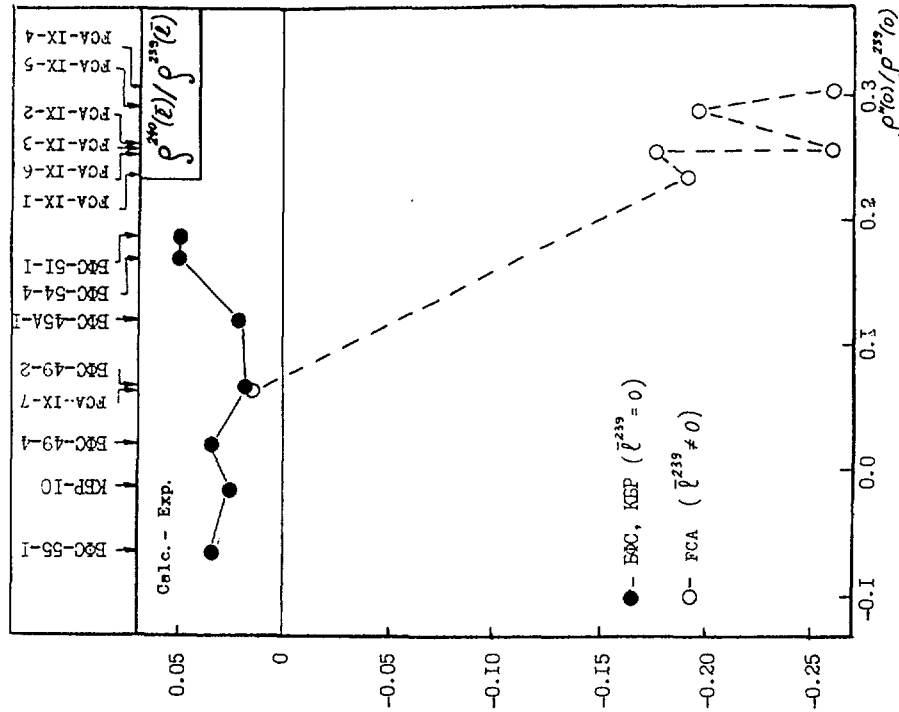


Fig.2. Correlation of ^{240}Pu central reactivity worths with hydrogen ones. $\rho'(0)/\rho'(0)$ - calculated (KEP, FCA) and measured (B5C).

TABLE VI. COMPARISON OF CALCULATED AND MEASURED AVERAGE CROSS-SECTIONS NORMALIZED BY U-235
 $(\sigma_{fc}^i / \sigma_f^{U235})$

Reaction rate	BP-I		BH-350				Pu-subassembly (ZLE center)metal U.21E		Depleted metal U.21E	
	E	C/E	Zone of low enrichment (ZLE) E	C/E	Zone of High enrichment (ZHE) E	C/E	E	E	E	C/E
$^{240}\text{Pu}(n,f)$	0.827±0.026	0.91±0.03	0.200±0.010	1.02	0.242±0.012	1.04				
$^{241}\text{Pu}(n,f)$	1.29±0.04	1.02±0.3	1.11±0.05	1.16	1.12±0.05	1.15				
$^{242}\text{Pu}(n,f)$	0.66±0.02	0.98±0.03								
$^{237}\text{Np}(n,f)$	0.77±0.023	1.04±0.04	0.182±0.006	0.994	0.249±0.012	0.93	0.227±0.011	0.978	0.190±0.07	1.01
$^{241}\text{Am}(n,f)$	0.825±0.025	1.00±0.04								
$^{237}\text{Np}(n,\gamma)$	0.240±0.012	1.08±0.07			0.73±0.04	0.95			0.68±0.03	1.09
$^{241}\text{Am}(n,\gamma)^a$			3.51±0.28	1.018			4.02±0.20	0.92		

a) Normalized by $^{239}\text{Pu}(n,\gamma)$

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A CONCEPT OF SPECIALIZED FAST REACTOR FOR MINOR ACTINIDE BURNING

V.I. MATVEYEV, A.P. IVANOV, E.M. EFIMENKO
Institute of Physics and Power Engineering,
Obninsk, Russian Federation

Abstract

The paper presents a concept of specialized sodium cooled fast reactor core designed for minor actinide burning. A special feature of the core at issue is utilization of free from fertile uranium-233 fuel material based on inert matrix with zirconium. The selection of this material enables to rise the portion of minor actinides in a mixture with uranium-235 or plutonium within the framework of the restrictions SVE and this ensures their efficient burn-up.

The paper contains the results of physical calculations of two cores of this type: for modular type reactor of 170 MWe and for BN-800-type reactor.

Here we shall confine ourselves to considering physics aspects of this problems. One of the main impediments is associated with the drastic rise in a positive value of sodium void reactivity effect (SVE) compared with conventional cores. Note, that "Nuclear Safety Guides on Reactor Installation of Atomic Power Plants" (NSG RP AS-89), valid within the borders of the Russian Federation contain the requirement to ensure a negative value of SVE. Another problem is connected with an essential reduction in the value of β_{eff} which originates from the decrease in the delayed neutrons importance in the system with a threshold nature of fission probability. The investigations conducted have demonstrated that the following principles inherent in the core concept can be applied:

1. The use is made of uranium-233 fertile material-free fuel, and fuel cores are made of special material representing an inert matrix based on zirconium with minor actinides inserted into it. This solution allows for not only a capability of more efficient burning of minor actinides, but a feasibility of significant decrease in SVE due to higher neutron leakage from the core.

2. The use is made of a mixture of minor actinides and uranium-235, which results in a noticeable rise in β_{eff} . Coming from these principles the reactor designs have been developed, with the data on their calculations stated below.

I. MODELS AND INITIAL DATA INVOLVED

The models of two reactors were selected for calculational investigations: a relatively small-power reactor (modular-type)-170 MWe reactor and a BN-800 type reactor. The both reactors adopted the similar fuel subassemblies (face to face dimension 96mm) and fuel elements ($d=6.9$ mm) with the maximum heat load $q_{1max}=450$ w/cm.

The fuel represents a zirconium matrix with minor actinides (neptunium, americium and curium mixed with uranium-235) doped into it. For the BN-800 type reactor consideration is also given to an alternative composition of fissile material—a mixture of minor actinides with plutonium from the VVER-1000 reactor spent fuel. With the notion of "enrichment" inapplicable in the event of the core of the type involved, the critical parameters are characterized by the density (g/cm^3) of fissile materials amount homogenized over the core volume.

The first reactor core represents a single-zone version, the second reactor is furnished with 3 sub-zones of dissimilar composition (analogous to three enrichments) to flatten the power density field. A sodium cavity is used in the both reactors for additional reduction in SVE, it is located between the core and the upper axial blanket—similar to 1/3 the lateral and bottom axial blankets are made of steel and sodium (the Tables I, II). The calculations were made in the traditional (diffusional) techniques in the two-dimensional (r, z) geometry.

INTRODUCTION

Use of fast reactors for minor actinide transmutation is likely to be an effective way of solving the general problem of long-lived nuclear waste activity reduction. The analysis of fast reactor capabilities for this purpose demonstrates the expediency to consider two trends:

- use of serial fast power reactors whose fuel contains minor actinides (3-4% weight) in the form of minor admixture; -adoption of specialized cores (or specialized fast reactors), whose fuel contains the maximum feasible amount of minor actinides.

It is assumed that in the first case no further serious problems in fuel cycle organization or in reactor design are assumed to emerge, except the necessity of the spent fuel deep purification from minor actinides. The latter circumstance represents the main condition of transmutation expediency in fast reactors in general. In the first case the efficiency of minor actinides transmutation is attained at the expense of fairly large amount of fast reactors in nuclear power by the estimates made their portion should be not less than 30%.

The second case is peculiar in the ability of neutronics properties of minor actinides—americium, neptunium, curium—in principle) to establish critical load on each of the above actinides including their mixtures. In this case their more complete and rapid burn-up could be attained with an efficiency 5-10-fold higher than that in the first case. Evident are the difficulties encountered in designing such a reactor.

TABLE I. SPECIFICATIONS OF THE REACTOR BN-300.

VERSION	VER. 1	VER. 2
Type of fuel	U+MA	Pu+MA
Composition of fresh fuel	Np/Am/Cm 59.26/37.41/3.33 ^a	Pu/Np/Am/Cm 59.82/6.67/4.18/0.37 ^a
Fuel matrix density (g/cm ³), accommodated over the core	0.48	
Volume fraction of fuel (%)	38.28	
Number of core sub-zones	3	
Core height (cm)	100.00	
Core sub-zone radius (cm)	82.00/105.24/128.84	
Sodium cavity thickness (cm)	30.00	
Compositions of lateral and bottom axial blanket (st./Na)	0.36/0.64	
Thickness of lateral and bottom blanket (cm)	100.0/100.0	
Number of burn-up compensators	12	
Burn-up compensator material	BAC (boron carbide 60% enrichment)	

^a Spent fuel of WWER-100 (5 year Decay)

2. RESULTS OF CALCULATIONS

The calculations of critical parameters were made proceeding from the requirement of the SVE zero value; for the BN-300-type reactor the additional demand was for the provision of power density field flattening over the core radius. In this case SVE was determined by the space incorporating the core, sodium cavity and upper axial blanket. In this case two critical parameters were determined: homogenized density of fissile materials and weight ratio of the total amount of minor actinides to the general amount of fissile isotopes.

TABLE II. SPECIFICATIONS OF THE REACTOR BN-170.

Type of fuel	U+MA
Composition of fresh fuel	Np/Am/Cm 59.26/37.41/3.33 ^a
Fuel matrix density (g/cm ³), accommodated over the core	0.37
Volume fraction of fuel (%)	30.00
Number of core sub-zones	1
Core height (cm)	100.00
Core radius (cm)	57.00
Sodium cavity thickness (cm)	30.00
Compositions of lateral and bottom axial blanket (st./Na)	0.36/0.64
Thickness of lateral and bottom blanket (cm)	43.0/60.
Number of burn-up compensators	6
Burn-up compensator material	BAC (boron carbide 60% enrichment)

^a Spent fuel of WWER-100 (5 year Decay)

The values of β_{eff} , reactivity variation resulting from fuel burn-up (within 30 days), efficiency of burn-up compensators, fuel composition under its burn-up over 30, 90 or 150 days were also calculated. The principal calculational results are given in Tables III-VI and in fig. 1.

The results presented testify to the feasibility of SVE zero value and acceptable value of β_{eff} in a specialized core of fast reactor with an adequately high amount of minor actinides in fuel- 45-50 %.

TABLE III. CALCULATIONAL VALUES OF MAIN CHARACTERISTICS

TYPE OF REACTOR	BN-170	BN-800	VER.1	VER.2 ^a
critical density of fuel material (g/cm ³ core)	1.75	zone1 0.86 zone2 1.10 zone3 1.25	zone1 0.31 zone2 0.40 zone3 0.45	
weight fraction of MA in fuel material (%)	53.0	47.0	11.2	
β_{eff} % (1-k)/k	0.484	0.526	0.301	
efficiency of fuel				
burn-up compensators % (1-k)/k	3.2	2.8	7.7	
continuous reactor operation (days)	160	110	65	
reactor reactivity variation during the 1-st month of operation: % (1-k)/k	-0.55	-0.78	-3.38	

^a Plutonium to minor actinides ratio for this version is unchanged, therefore SVE is -7.5 % (1-k)/k

TABLE IV. VARIATION OF FUEL COMPOSITION (KG) IN THE BN-170 REACTOR DURING BURN-UP TIME (DAYS)

ISOTOPES	0.0	30.0	90.0	150.0
am41	201.00	193.00	192.00	186.00
am2m	0.15	0.51	1.18	1.80
am43	70.20	69.50	68.20	66.50
cm42	0.13	1.57	4.01	5.81
cm44	22.50	22.70	23.00	22.90
cm45	1.51	1.56	1.66	1.76
np37	430.00	423.00	412.00	397.00
u235	651.00	640.00	617.00	598.00
u238	73.30	73.10	72.70	72.30
u236	0.00	2.30	6.85	11.36
pu38	0.00	3.68	12.20	21.12
pu39	0.00	0.16	0.53	1.13
fp35	0.00	12.20	36.50	60.40

TABLE V. VARIATION OF FUEL COMPOSITION (KG) IN THE BN-800 REACTOR DURING BURN-UP TIME (DAYS) /FUEL:U+MA VER.1/

ISOTOPES	0.0	30.0	90.0	150.0
am41	759.00	743.00	711.00	679.00
am2m	0.57	2.54	6.15	9.38
am43	264.00	261.00	254.00	248.00
cm42	0.48	8.54	21.90	31.90
cm44	84.80	86.20	88.80	91.10
cm45	5.69	6.00	6.60	7.22
np37	1620.00	1590.00	1530.00	1470.00
u235	2460.00	2390.00	2270.00	2150.00
u238	276.00	276.00	273.00	270.00
pu38	0.00	20.80	68.00	116.00
pu39	0.00	0.89	3.44	7.02
fp35	0.00	63.20	188.00	314.00

TABLE VI. VARIATION OF FUEL COMPOSITION (KG) IN THE BN-300 REACTOR DURING BURN-UP TIME (DAYS) / FUEL: PU+MA VER. 2/

ISOTOPES	0.0	30.0	90.0	150.0
am41	60.90	58.30	53.00	48.10
am2m	0.05	0.50	1.23	1.74
am43	21.30	22.30	24.40	26.90
cm42	0.04	1.96	5.14	7.36
cm44	6.81	7.39	8.70	10.13
cm45	0.46	0.57	0.82	1.12
pu38	21.60	26.00	35.90	44.60
pu39	931.00	921.00	803.00	693.00
pu40	428.00	434.00	438.00	434.00
pu41	212.00	205.00	193.00	181.00
pu42	103.00	104.00	105.00	110.00
np37	130.00	123.00	110.00	93.00
fp37	0.00	62.80	188.00	316.40

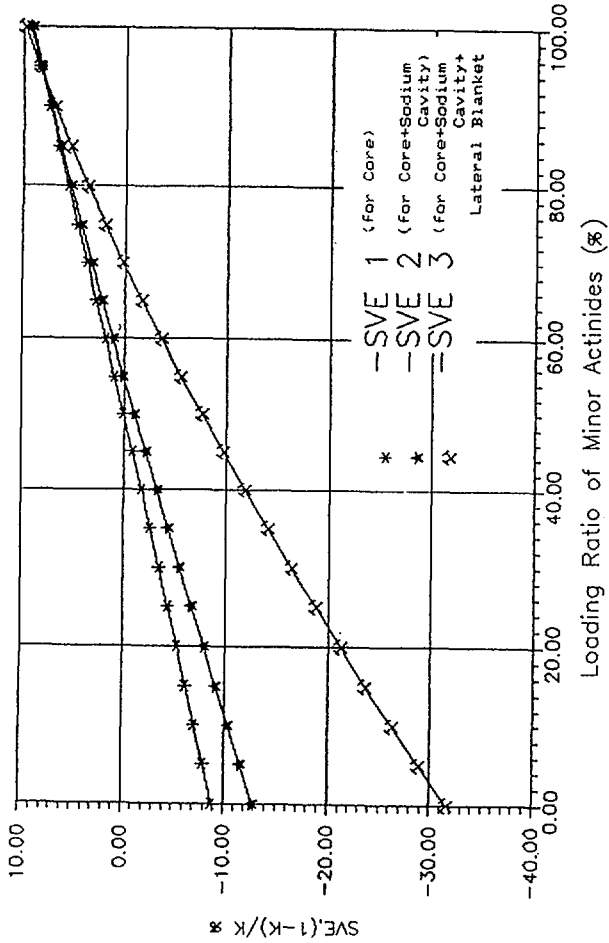


Fig.1 Relationship between loading ratio of minor actinides and sodium void effect (SVE).

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CONCLUSION

The problem of specialized core development (or specialized reactor) for efficient burning of minor actinides directly involves the problem of required safety parameters ensurance, primarily SVE and β_{eff} . This problem can be solved with the use of uranium-238-free fuel with inert diluent. The discussed capabilities will evidently be essentially determined by the specific features of the external fuel cycle, primarily by the feasibility to fabricate fuel containing a great amount of actinides. This needs a special consideration.

In the BN-170 and BN-300 reactors over a year 150 and 450 kg of minor actinides (Np, Am, Cm), respectively, are burnt.

THE PROPOSED FUEL CYCLE OF THE ACTINIDE BURNING FAST REACTOR DOVITA

(Summary)

A.V. BYCHKOV, A.A. MAYORISHIN, O.V. SKIBA
V.I. Lenin Research Institute of Atomic Reactors,
Dimitrograd, Russian Federation

The existing conceptions and programmes on transmutation of long-lived minor actinides (Np, Am, Cm) do not consider practically the question of fuel cycle development of actinide burner fast reactors (ABFR) or accelerator.

Our program of fuel cycle for ABFR is based on the following requirements:

- TRU waste content does not exceed 10^{-4} ;
- minimum wastes quantity is mainly in the solid form;
- minimum operations under fuel reprocessing;
- carrying out of all manufacturing process stages in the remotely controlled hot cells facility.
- To realize of our program on the ABFR closed fuel cycle (DOVITA) some unconventional and familiar approaches are used;
- (D) Dry reprocessing and preparation technologies of fuel composition, containing actinides will provide the minimum quantities of technological stages and wastes;
- (O) Oxide fuel application as the most studied will permit to use the familiar engineering decisions for FBR involving promising fuel elements and steel for achievement of maximum burn-up;
- (V) Vibropacking technology of fuel element production will permit to introduce the different compositions containing minor actinides into fuel and automate completely the remotely controlled production process;
- (I) Integral disposition of fuel reprocessing and fuel elements refabrication facilities together with reactor will permit in solving of ecological safety problems since such complex will need only supply of actinide mix from other reactors;
- (TA) The whole complex of approaches will permit to create the compact plant for Transmutation of Actinides.

The proposed program is based on the research developments:

- pyroelectrochemical production and reprocessing of fuel with high neptunium content;
- partial reprocessing of Am and Cm contained composition before a repeat irradiation;
- Am and Cm pyroelectrochemical separation from the main fusion products involving REE;
- introduction of the different additions into vibropac fuel elements involving compositions for the minor actinide burn-up;
- high burn-up of vibropac fuel elements.

Engineering basis for DOVITA program demonstration realization at RIAR:

- complex of facilities on pyroelectrochemical reprocessing and production of high activity nuclear fuel in the hot cells;
- automated facility for vibropac fuel elements production from high active fuel;
- BOR-60 fast reactor;
- complex of hot cells for postirradiation investigation;
- analytical and radiochemical laboratories.

The obtained experience which can be applied in realization of the program:

- BOR-60, BN-350 and BN-600 uranium-plutonium fuel and subassembly production;
- an experience of FBR irradiated uranium and uranium-plutonium fuel reprocessing by "dry" methods;
- an experience of development and operation of fuel elements and assemblies automated-production facilities.

The possible dates and stages for DOVITA program demonstration under financing:

1. Production of compositions containing the minor actinides; experimental fuel elements production and their irradiation in the BOR-60 reactor - during 3 years.
2. Postirradiation investigation of fuel elements; development of minor actinides contained fuel reprocessing; investigations on optimization of the closed fuel cycle - during 3 years.
3. Creation of demonstration complex on the BOR-60 minor actinides transmutation on RIAR basis; preparation of the suggestion for commercial facility design - during 3 years.

Considering the flexibility of pyrochemical technology of actinide fuel reprocessing and vibropac fuel technology the program can be changed taking into account the requirement for physics of definite FBR type and base fuel whatever uranium-thorium fuel.

REDUCTION OF MINOR ACTINIDES IN NUCLEAR WASTE VIA MULTIPLE RECYCLING IN FAST REACTORS

A. F. RENARD, S. PILATE, A. LA FUENTE

Belgonucléaire,
Brussels, Belgium

J. JOURNET, G. VAMBENEPE, J. VERGNES

Electricité de France,
Paris and Lyon,
France

Abstract

Seven successive re-irradiations in a fast reactor of the EFR type have been explicitly represented, with realistic out-of-pile times. This covers a period of time of about one century. Minor Actinides are assumed to be homogeneously recycled, i.e. mixed with the (U, Pu) oxide fuel. The advantage of the Pu + M.A. recycling strategy is to reduce the radio-toxicity of the actinides.

1. INTRODUCTION

The problem of long-ranging toxicity in nuclear waste due to alpha-emitting actinides is now clearly identified. While beta- and gamma-emitting fission products dominate the activity of nuclear waste over some hundred years, beyond that time the residual activity is nearly entirely due to the actinides.

The idea has thus emerged to recycle the quantities of generated actinides in nuclear reactors: one would recycle not only the plutonium, as in the present MOX recycling programmes, but also neptunium, americium and curium, the so-called minor actinides. Many recent studies, like [1] and [2] for example, have considered as well thermal as fast neutron reactors to that aim.

In the studies presented below, fast reactors are considered for recycling. Realistic schemes are assumed for the in-pile and out-of-pile times, and successive recycling operations are explicitly represented over a total period of about 100 years.

Minor actinides are assumed to be mixed homogeneously with the usual mixed oxide (U,PuO₂), in the same stoichiometric fraction as at the preceding discharge.

Reactors and schemes are defined in section 2.

The method of calculation is mentioned under section 3.

The impact on the toxicity of nuclear waste from recycling, either plutonium only, or plutonium and minor actinides, is compared and discussed under section 4.

Section 5 draws conclusions.

2. REACTORS AND SCHEMES

The initial fuel irradiation is assumed to take place in a 900-MWe, UO₂-fuelled PWR, analogous or close to 34 reactors running in France and 5 in Belgium. The U235 fuel enrichment is 3.7 %, so that a burnup of about 43,000 MWD/tonne can be achieved over an operation time of 4 years.

While the total PWR fuel inventory amounts to 72 tonnes of heavy metal, the fate of one tonne of uranium initially loaded is followed in a multiple recycling scheme.

The reactor used for recycling is a 1450-MWe fast reactor, similar to the first consistent design of the European Fast Reactor (EFR), already considered for such studies in [2]. Its average plutonium enrichment is close to 21 %, and one will associate one twelfth of such a reactor to the LWR above, as far as fuel quantities are concerned. In other terms, the actinides (either Pu alone, or Pu, Np, Am, Cm together, according to the case), produced from 4 years of operation of 12 PWRs can be recycled in one core load of such a fast reactor (FR).

Seven successive cycles of the Pu and minor actinide (MA) flows are explicitly represented, covering nearly a period of 100 years. The detailed time schemes are given in Table I below.

Table I
Recycling time schemes

Steps	Duration (years)
Initial PWR irradiation	4
Storage+cooling	3
Reprocessing+fabrication	2
FR irradiation n°1	5.5
Subsequent Storage+cooling	5
Reprocessing+fabrication	2
FR irradiations n°2 to n°7	5.5

As the fissile zones alone of the FR reduce progressively the Pu quantities, because they have a negative breeding gain, use is made of part of the Pu generated in blanket to restore criticality at reloading; the hypothesis of a FR just self-sufficient in Pu is thus made.

In view to establish the benefits of FR recycling on waste toxicity, the comparison is made with respect to a reference case without recycling, corresponding to the once-through strategy: after the PWR irradiation, all spent fuel assemblies are stored and constitute waste.

A first recycling strategy consists of re-irradiating successively in FR the Pu alone, supposed to be recovered with 99.5 % efficiency at reprocessing; minor actinides are rejected as waste.

In the second recycling strategy, both the Pu and the M.A. are recovered, with 99.5 % efficiency at reprocessing, and re-irradiated successively in FR ; only the 0.5 % residues go to waste.

Sensitivity studies have been added on the influence of the separation yields of Pu and of the M.A., and of the out-of-pile times.

3. METHODS OF CALCULATION

The computer programme used for this multiple recycle scheme is ORIGEN-2 [3]. It treats neutron flux irradiation periods as well as natural decay. It allows to calculate the evolution of activity for a very large variety of actinides (130 isotopes) and of fission products (850 isotopes). Among the actinides, one follows in particular all isotopes of interest for uranium, neptunium, plutonium, americium (10 isotopes) and curium (11 isotopes). The version of the programme used was released in 1990.

The cross-section library retained for thermal reactor irradiation is that referred to as '50,000 MWd/t'; the fast reactor library corresponds to the 'advanced' one. Parallel calculations with more detailed methods have served to check the validity of the burnup results. The LWR calculations were backed by a comparison with results produced in an OECD expert group [4], while the FR burnup calculations had been checked with respect to EFR design methods in [2].

The activities have been converted in relative toxicities, by dividing them by the values of Annual Limits of Intake (ALI), for an ingestion, recommended for the workers by the International Committee of Radiological Protection (ICRP) in 1990 [5]. These limits, defined in Becquerels, take the biological effect of the radiations into account.

It is worth mentioning that the 1990 ICRP recommendation has modified the ALI values for Pu, Np, Am, Cm with respect to the former 1986 recommendation : the ALI are now lower (thus more limitative) for Pu (by a factor 10), and for Am241, Am243 and Cm (by a factor 2), while they have been relaxed by a factor 10 for Np237.

A potential toxicity value can be calculated in the same way for the quantity of natural uranium ore (7.6 t) which is used to prepare one tonne of enriched uranium feeding the PWR.

Dividing the toxicity of the waste by the toxicity of the initial uranium ore allows to know after how long a time waste will become as harmless as the uranium ore is ; this is analogous to the method of the 'risk factor', as used in [1].

Maximum potential hazards only are considered here : no attempt is made to evaluate the possible release rates of fission products and actinides out of waste repositories.

Table II

Evolution of M.A. Quantities over one Century of Recycling Operations (Pu + M.A.) (in g/t of uranium initially loaded in PWR)

The first line always refers to the reactor loading (begin/end of irradiation), the second one to the waste (underlined figures)

	PWR	FR n°1	FR n°2	FR n°3	FR n°7	Sum of Waste
Np 237	627 3	627/297 <u>1.5</u>	300/155 <u>0.8</u>	158/93 <u>0.5</u>		51/46 <u>0.24</u>	9
Am 241	77 <u>1.5</u>	429/310 2	465/258 <u>1.7</u>	370/214 <u>1.5</u>		318/199 <u>1.46</u>	26
Am 243	169 <u>0.8</u>	168/229 <u>1.1</u>	228/243 <u>1.2</u>	241/230 <u>1.15</u>		169/156 <u>0.8</u>	8
(Cm 244)	62 <u>0.3</u>	51/89 <u>0.37</u>	68/107 <u>0.44</u>	81/112 <u>0.46</u>		69/82 <u>0.34</u>	1
Cm 245	2,8 <u>0.01</u>	2,8/16 <u>0.1</u>	16/23 <u>0.1</u>	23/27 <u>0.1</u>		24/22 <u>0.1</u>	0,7

4. RESULTS

4.1 Variation of M.A. quantities over 100 years of recycling

For the case of the recycle of Pu and M.A., the evolution of M.A. quantities over the 100-year period explicitly considered is given in Table II. Masses are shown in g per tonne of U initially loaded for four important M.A. isotopes : Np237, Am241, Am243 and Cm245. One observes the following :

- Np237 is reduced by about a factor 2 after each of the initial FR irradiations ; afterwards the reduction factor becomes

progressively smaller, leading to a minimum fraction of about 1/15th of the initial amount ;

- Am241 is growing in the two first recycles, because of the natural decay of Pu241 during the out-of-pile times ; then comes a slow, progressive decrease, which derives from the reduction of its predecessor Pu241 ;

- Am243 too is first growing, and then slowly decreasing ; this comes from the reduction by irradiation of its predecessor Pu242

- Cm245 requires some 7 cycles to become under control ; this isotope gives a small, long-term contribution to the build-up of Pu241 and Am241.

Table III gives for the 3 cases : no recycle, Pu recycle, Pu and M.A. recycle, the quantities of the same 4 isotopes which accumulate in waste after 100 years of recycling.

Table III
Quantities of Minor Actinides Accumulating in the Waste after one Century.
(in g/t of Uranium initially loaded in PWR)

	PWR once-through	PWR +7 Recycles in FR (Pu)	PWR +7 Recycles in FR (Pu + M.A.)
<u>Np</u> ²³⁷ PWR FR 1 to 7 Transmutations Total	629 - 170 800	629 169 125 922	3 4 2 9
<u>Am</u> ²⁴¹ PWR FR 1 to 7 Transmutations Total	299 - 1200 1500	299 1340 -112 1527	1.5 11 14 26
<u>Am</u> ²⁴³ PWR FR 1 to 7 Transmutations Total	169 - -1 168	169 572 -3 738	0.85 7.15 - 8
<u>Cm</u> ²⁴⁵ PWR FR 1 to 7	2.8 -	2.8 10	0.014 0.7

It appears from this comparison that, as far as waste is concerned, there is no advantage to recycle Pu only (except of the energy production). There is a large incentive to recycle M.A. in addition to the Pu, so that masses after 100 years can be reduced, with respect to the PWR case, by the following factors :

- 90 for Np237
- 60 for Am241
- 20 for Am243
- 4 for Cm245.

It should be stressed that the recycling programme cannot be stopped after this period, otherwise the reduction factors above would fall to about 14 for Np237 and 3 for Am241 ; there would be no reduction at all for the 2 last isotopes.

Such a recycling programme needs to be pursued. On the other hand, the M.A. can obviously be concentrated prior to recycling.

4.2 Long-term evolution of waste toxicity

In terms of toxicity, it is first useful to note what are the actinide isotopes contributing mostly to the total toxicity. This is done in Fig.1 for the waste from the initial PWR irradiation. The major contributors are, respectively :

- Am241 between 100 and 1000 years ;
- Pu239 and Pu240 between 1000 and 10,000 years ;
- Pu239 around 100,000 years ;
- Np237 and its successor Th229 beyond 100,000 years.

One notes that the importance of Np237 is smaller than in earlier work, like [1]. This is essentially a result of the re-evaluation of the ALI values for Np in the last ICRP recommendation. Now, successive re-evaluations in either sense let suggest that the results should be given a relatively large uncertainty.

Fig. 2 compares the time evolution of waste toxicity for the different cases considered : PWR alone, PWR plus 7 cycles in fast reactor, recycling Pu only, or M.A. in addition to Pu.

The total toxicity contained in waste is their maximum potential hazard, remaining trapped in packaged waste, should anyone suddenly ingest them. The effect of possible migration is not accounted for.

Fission products dominate first the waste toxicity. After a few hundred years their contribution is rapidly falling to a very low level, corresponding to the presence of Tc99 and I129. From that time on, the actinides dominate waste toxicity. Recycling M.A. in addition to Pu decreases their toxicity so that, with respect to the PWR case, the reduction is as follows :

- at 1000 years, by a factor 40.
- at 10,000 years, by a factor 25,
- at 100,000 years, by a factor 30.

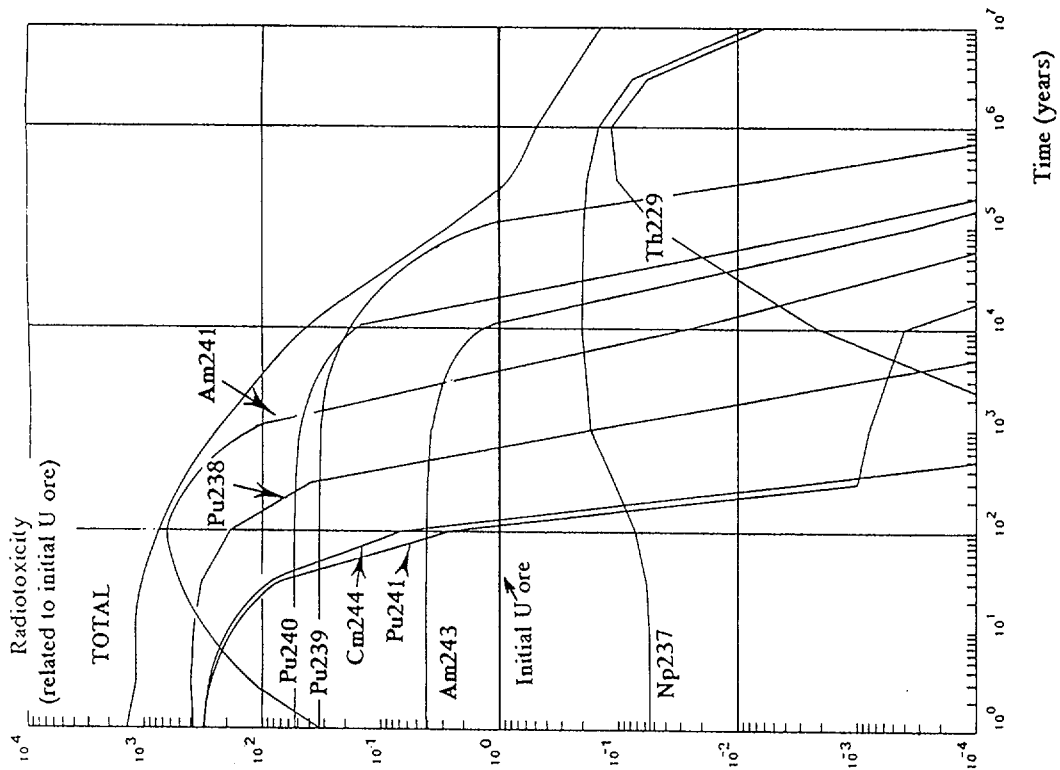


FIG. 1. Toxicity vs time for the actinides contained in waste (PWR, once-through).

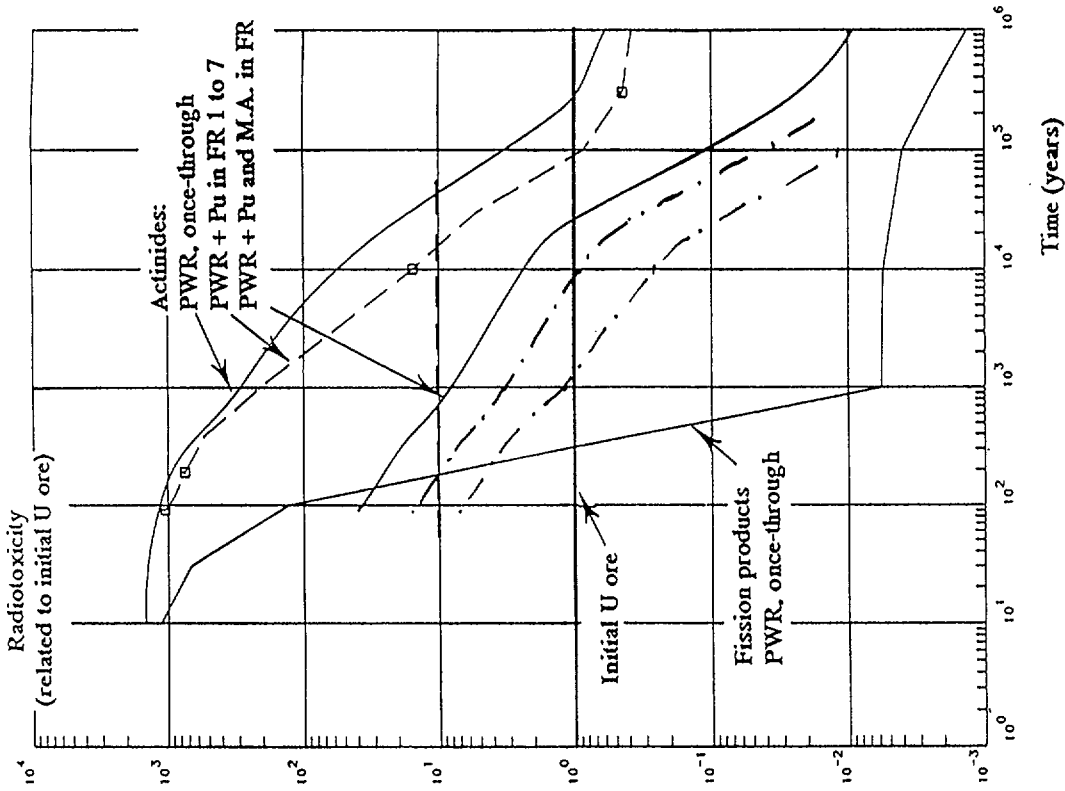


FIG. 2. Toxicity vs time for the actinides and fission products in waste.

These reduction factors are significant, although they do not reach the factor 200, theoretically associated with residues of 0.5 %.

4.3 First results in terms of acceptable toxicity levels

On Fig. 2, all toxicities are related to that of the quantity of uranium ore needed for the fabrication of the initial tonne of enriched uranium feeding the whole scheme. A first, simple criterion would thus be to declare acceptable (i.e. requiring no surveillance anymore) nuclear waste when their toxicity by ingestion has come down to this level.

According to this, fission products are acceptable after about 300 years. In contrast, actinides would need much longer times : 300,000 years (PWR, once-through), nearly 100,000 years (Pu recycle), or 20,000 years (Pu and M.A. recycle).

4.4 Toxicity versus energy production

In the FR recycling schemes considered here over a century, the total production of electrical energy is about 2.5 times higher than in the PWR irradiation alone. It appears thus justified to scale down by such a factor 2.5 the curves giving the toxicity of the actinides with recycles on Fig. 2.

4.5 Sensitivity studies

Two types of variants have been considered. In the first one the out-of-pile times were changed. In the second one, the separation yields of Pu and M.A. at reprocessing were varied.

If the out-of-pile times can be halved, this favourably affects the quantities of Am241 during the cycling operations themselves. For example, they are reduced by one third for the first two refabrication campaigns. On the other hand, the activity of Am241 in waste after 100, 200 or 1000 years is hardly influenced.

Assuming 80 % yield for the M.A. (what is already a hard challenge for Am) results in an increase of the actinide toxicity, given above for the 99.5 % assumption, by a factor 10 in the time range from 100 to 2,000 years. This stresses how important it is to develop high purification separation methods for the M.A., and first of all for Americium.

Improving the separation yield for the Pu isotopes from the 99.5% assumed above to 99.9% is beneficial, as it further reduces the actinide toxicity by a factor 3 around 1000 years and 4 between 10,000 and 100,000 years.

4.6 Acceptable toxicity levels : recapitulation

When one combines the findings of par. 4.4 and 4.5, assuming in particular recovery yields of 99.9% for Pu and 99.5% for M.A., the specific actinide toxicity is reduced by recycle down to the level of the natural uranium ore toxicity after, respectively, 15,000 years (Pu recycle) or about 1,000 years (Pu + M.A. recycle).

The latter time range can still be covered by human memory, so that a surveillance programme makes sense ; in contrast the former time ranges, much longer, escape from this scope.

(Simple criteria based on maximum potential toxicities are not meant here to be preferable to more elaborate criteria covering possible geological transfers by migration up to the groundwater. The point is rather to deduce orientations and priorities for further activities, based on parametric studies.)

5. CONCLUSIONS

Starting from an initial fuel irradiation in a PWR, the long-term potential radio-toxicity of the spent fuel, which becomes waste in the once-through option, has been compared to the toxicity of residual waste from multiple recycling in a fast reactor. Either the plutonium alone is recycled, or the plutonium and the minor actinides (Np, Am, Cm), all supposed to be recovered by reprocessing with a 99.5 % yield.

Seven successive re-irradiations in a fast reactor of the EFR type have been explicitly represented, with realistic out-of-pile times. This covers a period of time of about one century. Minor Actinides are assumed to be homogeneously recycled, i.e. mixed with the (U,Pu) oxide fuel.

The advantage of the Pu + M.A. recycling strategy is to reduce the radio-toxicity of the actinides by the following factors with respect to the PWR once-through case :

- at 1000 years, by a factor 40,
- at 10,000 years, by a factor 25,
- at 100,000 years, by a factor 30.

In addition, relating the actinide toxicity to the energy production scales down by a factor (e.g. 2.5) the curves for the actinides in the recycle cases.

The actinide isotope whose contribution is dominant from 200 to 1000 years is Am241.

Halving the out-of-pile times would reduce the quantities of Am241 effectively recycled, what is favourable to refabrication ; the toxicity of the waste itself is hardly influenced.

With respect to waste toxicity, recycling Pu + A.M. in fast reactors will be a success, provided that the recovery yield at separation can be brought high enough, as well for Pu isotopes as for M.A. isotopes and first of all for Americium : respective targets of 99.9% (Pu) and 99.5% (Am) should be aimed at in research work.

Under such conditions indeed, the actinide toxicity will be reduced to the level of toxicity which corresponds to the natural uranium ore initially used, after about 1,000 years ; in contrast to the PWR, once-through strategy (for which 300,000 years are needed) and also to recycling Pu alone (15,000 years are still required), this period of time makes sensible a surveillance programme, for waste repositories.

Note

The present results and orientations should be checked with regard to the long-term waste disposal assessments.

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LIST OF PARTICIPANTS

Bychkov, A.V.	V.I.Lenin Research Institute of Atomic Reactors 433510 Dimitrovgrad Ulyanovsk Region, Russian Federation	Mukaiyama, T.	Japan Atomic Energy Research Institute 2-4 Shirakata-shirane, Tokai-mura, Naka-gun Ibaraki-ken, 319-11, Japan
Burstall, R.F.	Physics Application & Development Dept. AEA Reactor Services AEA Technology Risley, Warrington, WA3 6AT, United Kingdom	Pilate, S.W.G.	Belgonucléaire S.A. Avenue Ariane, 4 B-1200 Brussels, Belgium
Cecilie, L.	Commission of the European Communities Rue de la Loi, 200 B-1049 Brussels, Belgium	Rabotnov, H.C.	Institute of Physics and Power Engineering 249020, Bondarenko Sq. 1 Obninsk, Kaluga Region, Russian Federation
Cockey, C.A.	General Electric Company Nuclear Energy 6835 Via de Oro, M/C S-72 San Jose, CA 95153-5354, United States of America	Sasahara, A.	Nuclear Engineering Department Komae Research Laboratory Central Research Institute of Electric Power Industry (CRIEPI) 11-1, Iwato Kita 2-Chome, Komae-shi, Tokyo - 201, Japan
Decressin, A.	Commission of the European Communities Rue de la Loi, 200 B-1049 Brussels, Belgium	Shmelev, A.N.	Moscow Institute for Physics and Engineering 31, Kashirskoe Shosse 115409, Moscow, Russian Federation
Ivanov, A.P.	Institute of Physics and Power Engineering 249020, Bondarenko Sq. 1 Obninsk, Kaluga Region, Russian Federation	Szatark, H.	Framatome/Direction Novatome Service NVTPC 10 rue Juliette Récamier BP 3087 F-69398 Lyon Cedex 03, France
Khalil, H.	Argonne National Laboratory Building 208, Argonne Illinois 60439, United States of America	Vasiliev, B.A.	Experimental Machine Building Design Bureau 603603, Nizhny Novgorod - 74 Burnakovskiy Proyezd, 15, Russian Federation
Koch, L.	Commission of the European Communities Joint Research Centre Institute for Transuranium Elements Postfach 2340 D-7500 Karlsruhe, Germany	Wakabayashi, T.	Orarai Engineering Center Power Reactor & Nuclear Fuel Development Corporation 4002 Narita, Orarai-machi Ibaraki-ken, 311-13, Japan
Kochetkov, L.A.	Institute of Physics and Power Engineering 249020, Bondarenko Sq. 1 Obninsk, Kaluga Region, Russian Federation	Wehmann, U.K.	Siemens AG Power Generation Group KWU Friedrich-Ebert-Strasse D-5060 Bergisch Gladbach 1, Germany
Ledgergerber, G.	Paul Scherrer Institute CH-5232 Villigen, Switzerland	Wiese, H.W.	Institut fuer Neutronenphysik und Reaktortechnik Kernforschungszentrum Karlsruhe Weberstrasse 5 Postfach 3640 D-7500 Karlsruhe 1, Germany
Matveyev, V.I.	Institute of Physics and Power Engineering 249020, Bondarenko Sq. 1 Obninsk, Kaluga Region, Russian Federation		
Matveenko, I.P.	Institute of Physics and Power Engineering 249020, Bondarenko Sq. 1 Obninsk, Kaluga Region, Russian Federation		

Zcykunov, A.G.

Institute of Physics and Power Engineering
249020, Bondarenko Sq. 1
Obninsk, Kaluga Region, Russian Federation

Arkhipov, V.

Scientific Secretary

International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400 Vienna, Austria

