

COMPARISON OF ACTINIDES AND FISSION PRODUCTS RECYCLING SCHEME WITH THE NORMAL PLUTONIUM RECYCLING SCHEME IN FAST REACTORS

by

Asif SALAHUDDIN¹ and Masood IQBAL²

¹Pakistan Atomic Energy Commission, Islamabad, Pakistan

²Pakistan Institute of Nuclear Science and Technology, Islamabad, Pakistan

Scientific paper

DOI: 10.2298/NTRP1301001S

Multiple recycling of actinides and non-volatile fission products in fast reactors through the dry re-fabrication/reprocessing atomic international reduction oxidation process has been studied as a possible way to reduce the long-term potential hazard of nuclear waste compared to that resulting from reprocessing in a wet PUREX process. Calculations have been made to compare the actinides and fission products recycling scheme with the normal plutonium recycling scheme in a fast reactor. For this purpose, the Karlsruhe version of isotope generation and depletion code, KORIGEN, has been modified accordingly. An entirely novel fission product yields library for fast reactors has been created which has replaced the old KORIGEN fission products library. For the purposes of this study, the standard 26 groups data set, KFKINR, developed at Forschungszentrum Karlsruhe, Germany, has been extended by the addition of the cross-sections of 13 important actinides and 68 most important fission products. It has been confirmed that these 68 fission products constitute about 95% of the total fission products yield and about 99.5% of the total absorption due to fission products in fast reactors. The amount of fissile material required to guarantee the criticality of the reactor during recycling schemes has also been investigated. Cumulative high active waste per ton of initial heavy metal is also calculated. Results show that the recycling of actinides and fission products in fast reactors through the atomic international reduction oxidation process results in a reduction of the potential hazard of radioactive waste.

Key words: fast reactor, recycling, actinide, fission product, plutonium, PUREX, AIROX

INTRODUCTION

The transmutation of long-lived nuclear waste nuclides to stable or short-lived nuclides is considered to be a possible way of reducing the long-term hazard of nuclear waste. The nuclear fuel cycle involves a number of processes, starting from uranium ore mining up to the disposal of nuclear waste. While the fuel fabrication process and energy production in nuclear reactors have attained a high state of technological development, an optimal solution for fuel reprocessing and waste disposal processes is still under discussion. With the growing contribution of nuclear energy in the generation of electric power, the accumulation of nuclear waste and the question of its disposal present an obstacle to public acceptance of the nuclear industry.

Radioactive waste appears at all steps of the fuel cycle. The tailings leftover from mining and milling of

uranium ores constitute the largest mass of low-level waste. But the main source of concern, apart from the tailings, is the high-activity waste generated during the reprocessing of the spent fuel discharged from nuclear reactors and the fabrication of fuel containing recovered plutonium or transplutonium actinides.

In the PUREX process, about 99% of U and Pu are separated from the spent fuel, while the rest is treated as waste [1]. The waste thus contains about 1% of U and Pu, in addition to minor actinides (all other actinides except U and Pu, *i. e.* mainly Np, Am, and Cm) and fission products. Some of these fission products and actinides such as ⁹⁹Sr, ⁹⁹Tc, ¹²⁹I, ¹³⁵Cs, ¹³⁷Cs, Np, Am, and Cm, have half-lives ranging from 30 to more than 10⁵ per years. Thus, the final storage of nuclear waste is a long-term problem. A safe storage of several hundred to thousands of years is required.

A considerable effort is being made in a number of countries to study the long-term storage of highly radioactive wastes after solidification, vitrification and packaging. Some of the proposals are: disposal

* Corresponding author; e-mail: asif_salahuddin@yahoo.com

into caverns, tunnels or holes drilled into geological formations of hard rock, salt, clay or the sea bed. Proposals have also been made to shoot the waste into space.

A method suggested as an alternative to the above stated ones is that of neutron burn-up of the long-lived nuclear waste nuclides in thermal and fast reactors. That is, of the conversion of long-lived nuclides into short-lived or non-radioactive nuclides. Other devices to be considered for this kind of nuclear transmutation are: accelerators, molten salt reactors, blankets of fusion reactors, special transmutation reactors, and nuclear explosions. Generally, accelerators require more power than is generated by the reactors producing the waste [2]. Molten salt reactors and fusion reactors are not available at present. Special fission transmutation reactors (*e. g.* high flux reactors, accelerator-driven reactors) would require a detailed reactor design before an analysis of the fuel cycle could begin. Further, the research, development, and demonstration costs of such a waste management reactor would be very high [3]. The feasibility and public acceptance of nuclear explosions is highly doubtful.

The transmutation of solely transuranium actinides is under consideration in a number of countries and a considerable amount of literature on the subject is available. Studies have shown that the transmutation of minor actinides is feasible both in thermal as well as in fast reactors. However, the main problem in actinide transmutation is actinide separation from nuclear waste. The technology of actinide separation is very complicated and expensive and a lot of research and development work is required [4]. The high capital costs of the processing facilities outweigh the possible benefits from the reduced final waste disposal charges [5].

Some studies have been carried out in South Korea on the use of PWR spent fuel in CANDU reactors directly through the oxidation reduction of oxide fuel (OREOX) process, known as DUPIC fuel. Economic analyses of DUPIC fuel handling, fabrication, cycle and disposal has proved it to be feasible, as well. It has been established that the DUPIC fuel cycle can save uranium resources by 20 to 23% and also reduce the spent fuel for up to 67% [6-9].

It is, thus, clear that the main obstacle in the recycling of minor actinides is their separation from high active waste. The removal of minor actinides does not, of course, eliminate the necessity of protecting the waste until major fission products have decayed. This means that a disposal of about one thousand years is still required. Therefore, it seems worthwhile to look for a possibility of recycling fission products in nuclear reactors *along with* actinides, in an attempt to avoid the necessity of perpetual storage.

To achieve this goal, it has been proposed to reprocess the spent fuel with a non-aqueous reprocessing fabrication process called atomics international re-

duction oxidation (AIROX) process [10-15]. This process is based on the oxidation of UO_2 fuel to U_3O_8 and, then, its re-conversion to UO_2 by reduction with hydrogen. The two reactions involved are:

- (a) oxidation $3\text{UO}_2 + \text{O}_2 \rightarrow \text{U}_3\text{O}_8$ (reaction takes place at $\sim 400^\circ\text{C}$), and
- (b) reduction $\text{U}_3\text{O}_8 + \text{H}_2 \rightarrow 3\text{UO}_2 + 2\text{H}_2\text{O}$ (reaction takes place at $\sim 600^\circ\text{C}$).

The phase change in the UO_2 lattice structure due to oxidation results in the pulverization of fuel pellets and release of volatile fission products (VFP). This property is utilized to separate the fuel from the cladding, *e. g.*, for LWR about 99.9% of the fuel is separated from the fuel cladding [16]. The declad material is then reduced to UO_2 by hydrogen. The repetition of the oxidation reduction process a number of times (about three times) will produce a fuel powder which has good sintering properties for the refabrication of fuel pellets.

This process removes the volatile fission products [like ^3H (100%), Kr (99%), Xe (99%), Cs (95%), and Ru (40%)] from the spent fuel, allowing the recycling of actinides as well as non-volatile fission products (NVFP) without going through the complicated and expensive methods of separating minor actinides from the spent fuel. After the removal of volatile fission products, the remaining fuel is used to re-fabricate new fuel elements, upon the addition of a certain amount of fissile material to it. The addition of fissile material is required so as to compensate for the loss of reactivity due to the burn-up of the fissile material in the previous cycle and the presence of neutron poisons in the form of fission products in the fuel. The re-fabricated fuel elements are then used to reload the reactor. In this way, each reactor accumulates and transmutes its own actinides and fission products.

This study suggests multiple recycling of NVFP and actinides by recycling them in fast reactors. With the increasing number of cycles, the in-pile time, as well as the concentration of certain nuclides, will increase and that, in turn, will cause an increase in the transmutation rate of those nuclides. An advantage of the multiple recycling concept seems to lie in the fact that the reactor itself offers a safe place for the temporary storage of fission products and actinides. Fission products and actinides with half-lives of the order of one to thirty years may decay to a large extent during the life of the reactor (approximately 30 years) and one does not need additional storage for them. In this manner, with the help of the multiple recycling of actinides and fission products, the problem of waste disposal should possibly decrease or, at least, postpone the time for its final storage.

After multiple recycling, the spent fuel contains U and Pu, as well as high concentrations of fission products and minor actinides. Either U or Pu are separated by the aqueous process from this fuel and the rest is placed into an ultimate storage (strategy I) or, all the

fuel materials are put into an ultimate storage without separating U and Pu (strategy 2).

Calculations for various in-pile and out-of-pile parameters (such as fuel inventory, fissile material requirements, activity, decay heat, ingestion hazard and radiation level) of different recycling schemes in a liquid metal fast breeder reactor (LMFBR) are presented in this paper. The investigated schemes are plutonium recycling (PuR) and actinides & fission products recycling (AFR). Plutonium recycling (PuR) is the normal scheme used in fast reactors. The main purpose of these calculations is to investigate the advantages and disadvantages of AFR over PuR with respect to long-term waste hazards.

REACTOR MODELLING

Method of calculation

Calculations for in-pile as well as out-of-pile parameters are done by means of a modified version of the Karlsruhe isotope generation and depletion code, KORIGEN [17]. This computer code has been used for burn-up, isotopic concentration and decay characteristics of spent fuel [18-20]. The modifications to the code are described below.

KORIGEN-M program for fast reactor calculations

In the modified version of KORIGEN, hereafter called KORIGEN-M; certain nuclides in the fuel can be removed or replaced by other nuclides. For example, in the AIROX recycling process (*i. e.* AFR) scheme, volatile fission products are removed from the spent fuel. The decrease in the density due to the accumulation of NVFP in the fuel is accounted for. The code also takes into account the fabrication and reprocessing losses. Furthermore, there is an option allowing the amount of fissile material added to the recycled fuel to be adjusted automatically, so that the infinite multiplication factor (k_{∞}) at the beginning of each cycle equals the k_{∞} at the beginning of the first cycle.

To increase the accuracy of the results of the fission product inventory in spent fuel, a novel fission product yields library for fast reactors has been created which has since replaced the old KORIGEN fission product yields library. In the old KORIGEN library, fission product yields for fast reactors were available only for three actinides ^{235}U , ^{238}U , and ^{239}Pu . The new KORIGEN-M library for fast reactors contains data for seven actinides, *i. e.* ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , and ^{241}Am [21]. In the library, fission product yields for other actinides (*e. g.* ^{237}Np , ^{243}Am , ^{244}Cm *etc.*) have been put equal to those of the nearest actinide isotopes in mass number.

Depletion calculations

A 1250 MWe liquid metal fast breeder reactor (LMFBR) is taken as representative of fast reactors. Such a reactor, the Superphoenix, is operating in France [22]. A detailed model of the 1250 MWe LMFBR is described by Croff *et al.* [23]. At reloading time, the LMFBR is refueled with a batch of core fuel elements containing 9.8 TIHM (tons of initial heavy metal) core fuel, 5.166 TIHM of axial blanket fuel and a batch of radial blanket fuel elements containing 9.059 TIHM radial blanket fuel. In an equilibrium cycle, the reactor core consists of 3 and the radial blanket of 6 batches, respectively. At reloading time, a batch which has reached its full burn-up at the core is removed and replaced by a fresh batch. At the same time, a batch from the radial blanket is removed and replaced by a fresh one. Both blankets are composed entirely of depleted uranium (0.2 wt.% ^{235}U) in the form of oxide pellets. The core fuel is comprised of the same depleted uranium enriched with 18.5% LWR Pu. The isotopic composition of LWR Pu is taken as 1.4/55/25.3/13.3/5 percent for ^{238}Pu / ^{239}Pu / ^{240}Pu / ^{241}Pu / ^{242}Pu , respectively. The core fuel is then irradiated for 3 years, resulting in a final burn-up of 101,000 MWD/TIHM. The axial blanket fuel, contained in the same assembly as the core, achieved a burn-up of 5660 MWD/TIHM at discharge. The radial blanket fuel irradiated for 6 years and then moved from the outermost to the innermost row of the radial blanket during this time, resulted in a burn-up of 7250 MWD/TIHM at discharge. In this paper, the specific power (MWD/TIHM) of the fuel and the number of irradiation days in the specified reactor are used as input parameters and kept constant in each cycle for depletion calculations of the different fast reactor recycling schemes. The startup period of the reactor is neglected and depletion calculations are based on the fuel that has obtained the required discharge burn-up, as mentioned above (for example, 101,000 MWD/TIHM for the core fuel and 7250 MWD/TIMH for the radial blanket fuel). Other general characteristics assumed for different recycling schemes in the said calculations are given below:

- ex-core time for the core fuel in PuR and AFR is taken to be one year,
- reprocessing and refabrication losses in the aqueous reprocessing scheme (*i. e.* PuR) are assumed to be 0.5% each, while in the AIROX process scheme (AFR), these losses are assumed to amount to 0.1% and 0.5% , respectively,
- in the recycling schemes considered in this paper, it is assumed that no part of the axial or radial blanket discharged fuel is used in the refabrication of blanket fuel elements; however, the Pu obtained after the reprocessing of the discharged fuel from axial and radial blankets is used for the re-enrichment in the refabrication of recycled core fuel elements in PuR and AFR; this re-enrichment is required as a guarantee for the criticality of the reactor; the cooling and reprocessing time of axial

and radial blanket elements have been assumed to amount to 9 months.

Calculations for the 10 complete cycles in each recycling scheme of the LMFBR have been done. The duration of a complete cycle is equal to the length of a fuel cycle which is defined as the time period between the fabrication of a given fuel batch to its refabrication after reprocessing (*i. e.* complete cycle length = fuel cycle length = in-core time + ex-core time). The method

and the cross section data used in the depletion calculations of both recycling schemes are described below.

PuR

A flow diagram for depletion calculations and for the adjustment of fissile material in the core of PuR is given in fig. 1. The cross-sections used are averaged one-group cross-sections, constant in time, calculated for

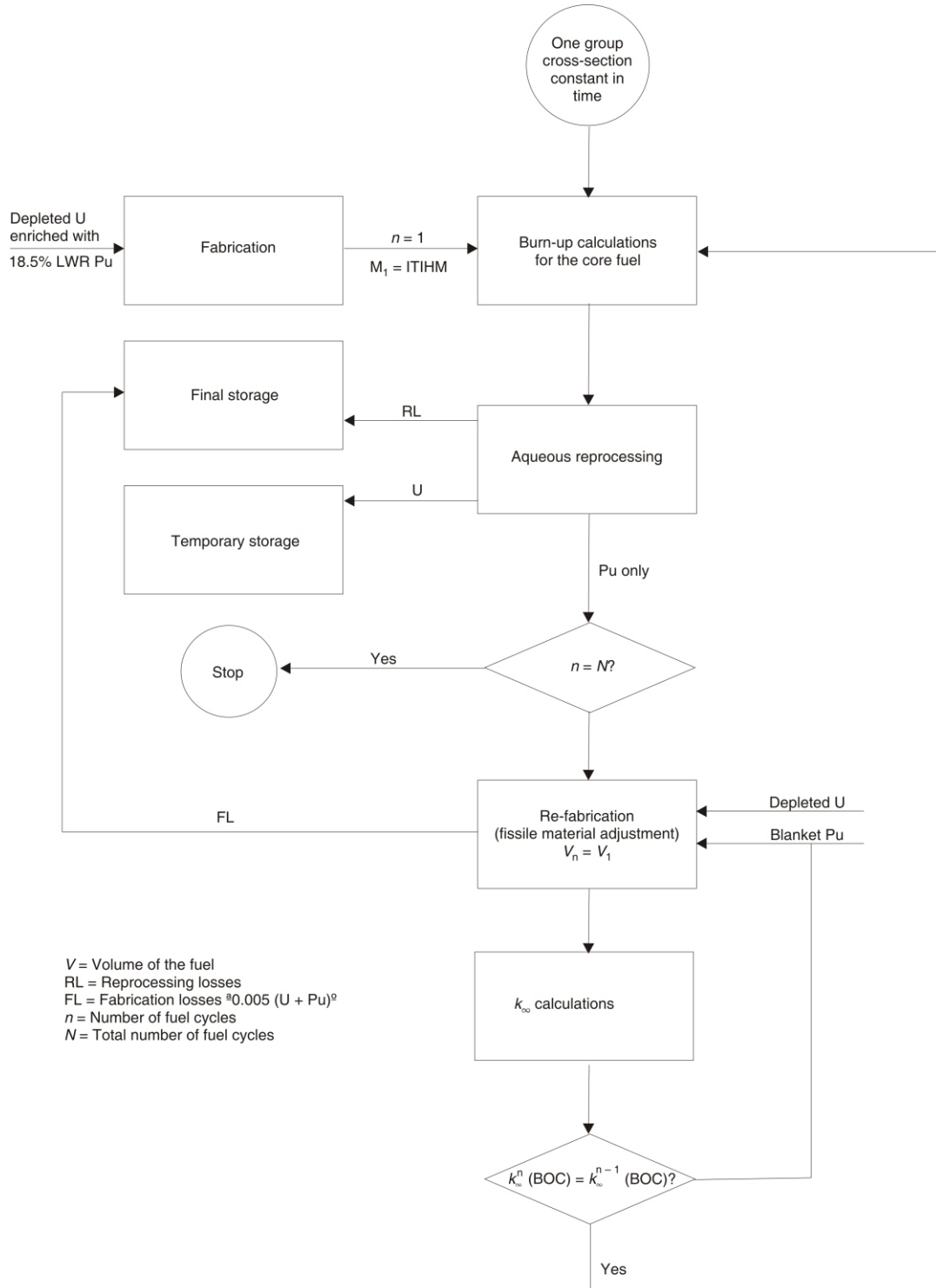


Figure 1. Flow diagram of depletion calculations and the adjustment of fissile material in PuR core fuel

the initial composition of the SuperPhoenix [22]. Reactor specific, zone-dependent, one-group effective cross-sections for the isotopes of actinides and fission products available in the 26 group extended KFKINR library are calculated by collapsing the 26 groups to a single group by using the 26 group energy spectrum obtained by 2-D calculations performed with a 2-D multi-group diffusion program DIXY [24]. For the purposes of this study, the standard 26 group data set, KFKINR [25], developed at the Forschungszentrum Karlsruhe, Germany, was extended. The KFKINR energy group structure is the same as the ABBN energy group structure [26], except for the thermal group. In the thermal group, the Maxwellian spectrum averaged cross-sections are used instead of the 2200 m/s ones. The KFKINR data set is supplemented with the following group cross-sections:

Actinides

The 26 group data for the 13 actinides, ^{233}Pa , ^{237}Np , ^{239}Np , ^{238}Pu , ^{241}Am , ^{242}Am , $^{242\text{m}}\text{Am}$, ^{243}Am , ^{242}Cm , ^{243}Cm , ^{244}Cm , ^{245}Cm , and ^{246}Cm , are added.

Except for the self-shielding factors, the normalized transfer matrices of elastic and inelastic scattering and (n, 2n) processes, all the data are calculated from the differential data library KEDAK-4[27]. The data are processed with the group constant generating code MIGROS-3 [28]. The self-shielding factors (f-factors) for the 4 actinides (namely, ^{233}Pa , ^{237}Np , ^{238}Pu , and ^{244}Cm and the transfer matrices for the 13 actinides mentioned above) are taken from the improved version of the KFKINR library [29]. The self-shielding factors for the remaining 9 actinides mentioned above have been taken as a unity. The KFKINR-2 library uses the weighting spectrum of a 1300 MWe LMFBR. Because of the exploratory nature of this investigation, the time dependence of the weighting spectrum is neglected.

Fission products

The cross-sections for the following 68 fission product nuclides are added to the KFKINR library in a 26 group representation: ^{81}Br , ^{83}Kr , ^{84}Kr , ^{85}Kr , ^{86}Kr , ^{85}Rb , ^{87}Rb , ^{88}Sr , ^{90}Sr , ^{89}Y , ^{91}Zr , ^{92}Zr , ^{93}Zr , ^{94}Zr , ^{95}Zr , ^{96}Zr , ^{95}Mo , ^{97}Mo , ^{98}Mo , ^{100}Mo , ^{99}Tc , ^{100}Ru , ^{101}Ru , ^{102}Ru , ^{103}Ru , ^{104}Ru , ^{106}Ru , ^{103}Rh , ^{105}Pd , ^{106}Pd , ^{107}Pd , ^{108}Pd , ^{110}Pd , ^{109}Ag , ^{111}Cd , ^{128}Te , ^{130}Te , ^{127}I , ^{129}I , ^{131}I , ^{131}Xe , ^{132}Xe , ^{133}Xe , ^{134}Xe , ^{136}Xe , ^{133}Cs , ^{134}Cs , ^{135}Cs , ^{137}Cs , ^{138}Ba , ^{139}La , ^{140}Ce , ^{141}Ce , ^{142}Ce , ^{144}Ce , ^{141}Pr , ^{143}Nd , ^{144}Nd , ^{146}Nd , ^{148}Nd , ^{150}Nd , ^{147}Pm , ^{149}Sm , ^{151}Sm , ^{152}Sm , ^{153}Eu , and ^{155}Eu . Additional material, such as the pseudo fission product (FPP9), is also added to the KFKINR set. The data for these materials have been calculated by ECN-Petten [30]. Here, again, a 1300 MWe LMFBR weighting spectrum is used.

It has been confirmed that these 68 fission products constitute about 95% of the total fission products yield and about 99.5% of the total absorption due to

fission products in fast reactors. The resonance self-shielding factors for all 68 fission products have been taken as unity.

The one-group cross-sections, constant in time, for the light elements and the rest of the isotopes of actinides and fission products, are taken from the KORIGEN standard data library for fast reactors [17].

In fast reactors, the cross-sections are not very dependent on burn-up because of a relatively high internal conversion; in addition, the neutron spectrum has a high average energy above the resonance region. Consequently, the mean free path of neutrons is large (about 5 cm), as compared to dimensions of the fuel pins and the distance between the pins. Due to this effect, the core can be treated as homogenous.

After a cooling time of about 200 days, the spent fuel is reprocessed through the PUREX process and U and Pu are separated from it. The recycling of uranium obtained from the spent fuel has not been considered and it is assumed that it is to be kept in a temporary storage until a further decision about its use is reached.

The Pu obtained from the core spent fuel is recycled again and mixed with depleted uranium to re-fabricate new fuel elements to be loaded into the next cycle. This Pu, however, is not sufficient to guarantee the criticality of the reactor because of its burn-up in the last cycle in the core. Therefore, a certain amount of blanket (axial and/or radial blanket) Pu has to be added, keeping the volume of the core fuel constant in each cycle. The ratio of Pu and depleted U is adjusted so that the K_{∞} at the beginning of each cycle is equal to the K_{∞} at the beginning of the first cycle. A total cycle loss of 1% of Pu and U during reprocessing (0.5%) and re-fabrication (0.5%) of the fuel elements has been considered.

AFR

A flow diagram of depletion calculations and the adjustment of fissile material in the AFR core fuel is given in fig. 2. Average one-group cross-sections, constant in time, used in these calculations are the same as mentioned in the case of PuR. In the fuel, the neutron spectrum will become harder with the increasing number of cycles. This increased average energy in a fast reactor is above the resonance region where the fission and capture cross-sections do not vary rapidly. Therefore, the change in cross-sections due to this effect is small. However, for exact calculations, one-group cross-sections averaged over the neutron spectrum for various burn-up steps in each cycle have to be used.

After a cooling time of about 200 days, the spent fuel is treated by the AIROX process which removes volatile fission products. The rest of the fuel is then available for the refabrication of fuel elements to be loaded in the next cycle.

The fissile material added to the recycled fuel for criticality is taken to be blanket Pu. If the Pu from axial

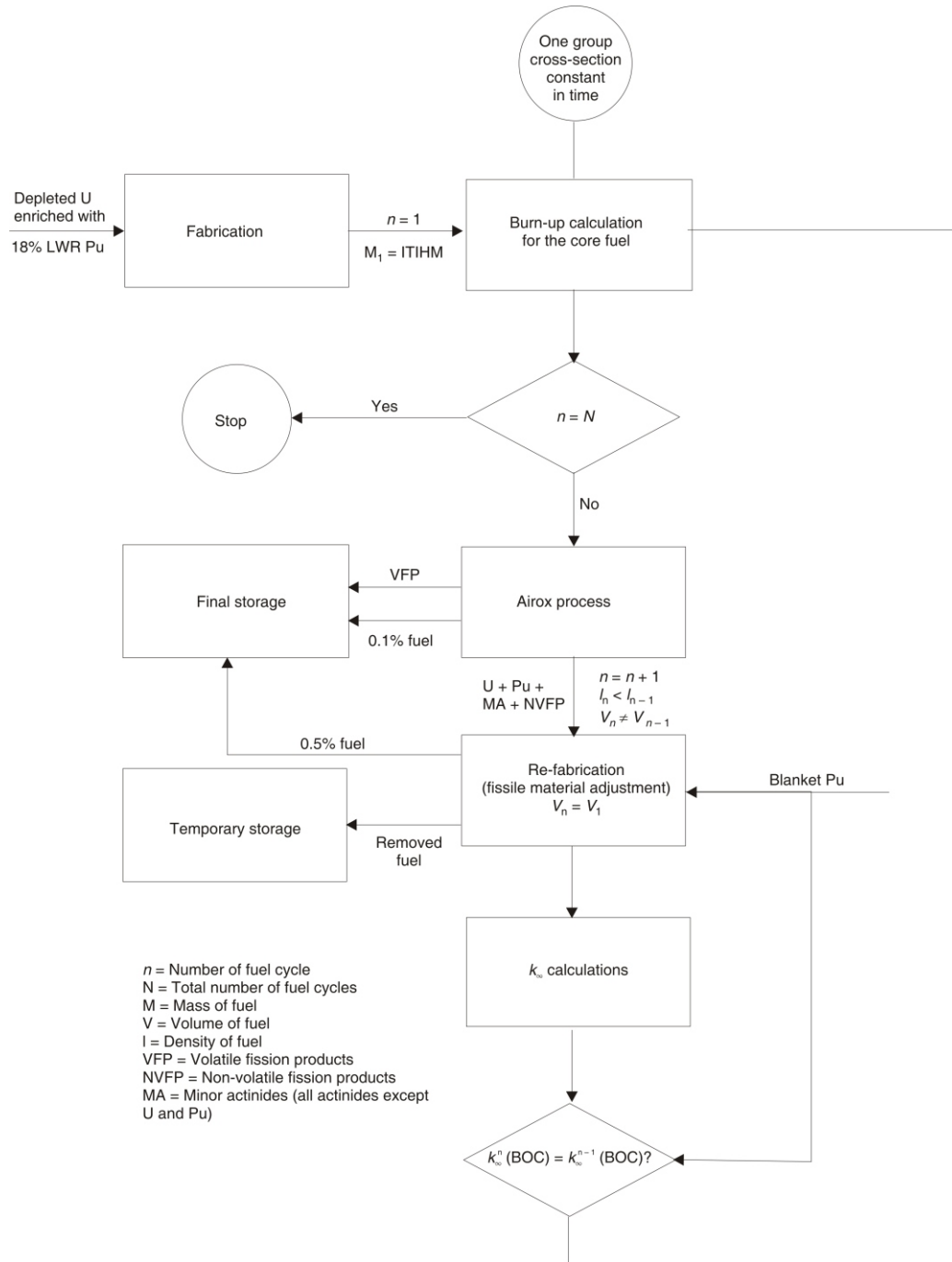


Figure 2. Flow diagram of depletion calculations and the adjustment of fissile material in AFR core fuel

and radial blankets is not sufficient for enrichment, additional Pu is assumed to be taken from the blankets of a similar LMFBR which may not be operating under the AFR scheme.

Determination of cumulative high active waste (HAW) per TIHM

In both recycling schemes, *i. e.* PuR and AFR, the cumulative waste per TIHM for the core fuel is calculated 7 years after the discharge of the fuel from the last cycle. In these calculations, the decay of nuclides

present in the waste from the first to the 10th cycle is taken into account. The total time from the start of the first cycle to the calculation of cumulative HAW is 46 years.

In PuR, the cumulative waste per TIHM for the core fuel is calculated by adding the waste from fabrication losses and the waste from the aqueous reprocessing of the spent fuel.

In AFR, the cumulative waste per TIHM for the core fuel is calculated by adding the fabrication losses, volatile fission products, losses in AIROX reprocessing and waste from the last fuel cycle according to the following two strategies:

- AFR (S1): U and Pu are separated by an aqueous process from the finally recycled fuel and the fuel removed during re-fabrication, and
- AFR (S2): U and Pu are not separated from the finally recycled or removed fuel.

The cumulative waste due to axial and radial blankets will be the same in both PuR and AFR, because they were started with the same material (*i. e.* depleted U) and are assumed to have the same specific power and burn-up characteristics in both recycling processes. The waste from the blankets being the same in PuR and AFR has not been considered.

RESULTS AND DISCUSSION

The results of calculations for in-pile (*e. g.* fuel inventory, fissile material requirements) and out-of-pile parameters (including activity, decay heat and radiation in the spent fuel and cumulative waste) for different recycling schemes in an LMFBR are presented. The results for each parameter are initially discussed for the respective recycling scheme and afterwards compared with those of the other recycling scheme.

Fuel inventory

The fuel inventory gramme (g) per TIHM at the charge and discharge of core fuel in each cycle in PuR and AFR is discussed in the following sub-sections:

PuR

The nuclide inventory per TIHM for the core fuel at charge and discharge in different fuel cycles in a PuR is given in tab. 1. The Pu vector has changed from the LWR Pu vector $^{238}\text{Pu}/^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}/^{242}\text{Pu}$ equal to 1.4/55.0/25.3/13.3/5.0, respectively, at the beginning of the first cycle, to 0.2/59.0/34.6/3.6/2.6, at the beginning of the 10th cycle. The amount of ^{239}Pu and ^{240}Pu has increased while that of ^{238}Pu , ^{241}Pu , and ^{242}Pu decreased. The behavior of different isotopes of Pu with the increase in the number of cycles is shown in fig. 3. Since the reactor is started with LWR Pu, it has relatively large isotopic concentrations of ^{238}Pu , ^{241}Pu , and ^{242}Pu at the beginning of the first cycle. In an irradiation cycle, the fuel changes mainly due to the burn-up of ^{239}Pu and ^{241}Pu and the buildup of ^{239}Pu via neutron captures in ^{238}U . Because of its large fission cross-section, ^{241}Pu burns more rapidly than ^{239}Pu . During re-fabrication, blanket Pu containing mainly ^{239}Pu is added. These isotopes come almost in equilibrium after cycle 3. However, a very small decrease in the value of ^{239}Pu and a very small increase in the value of ^{241}Pu occur after cycle 5, due to the increasing amount of ^{240}Pu .

However, the amount of ^{240}Pu increases continuously with the increasing number of cycles because its rate of production through neutron capture in ^{239}Pu is

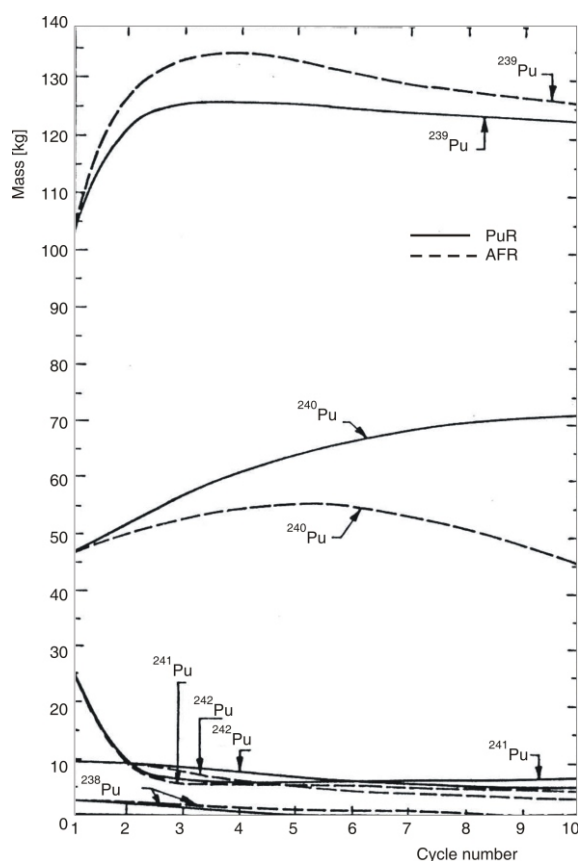


Figure 3. Amount of different Pu isotopes per TIHM present in the core fuel at the beginning of different fuel cycles in PuR and AFR

higher than its destruction through neutron absorption. The increase in the amount of ^{240}Pu contributes to the positive reactivity of the system because the specific reactivity worth of ^{240}Pu is positive. The specific reactivity worth is defined as the ratio of the change in core reactivity per incremental kilogram of the specific material loaded and is calculated by incrementing the initial load of each material by 10 kilograms [31]. The increase in the amount of ^{240}Pu through neutron capture contributes to the increase in the production of ^{241}Pu , adding to the positive reactivity of the system. Due to these two effects, the amount of ^{239}Pu required to maintain reactor criticality decreases slightly after the 5th cycle.

In the fuel discharged at each cycle, the amount of ^{241}Am produced mainly through the decay of ^{241}Pu behaves similarly as ^{241}Pu , *i. e.* it decreases for the first 4 cycles and then increases with the increasing number of cycles (tab. 1). ^{243}Am , produced via neutron capture in ^{242}Pu , similarly, decreases with the increasing number of cycles, as is the case with ^{242}Pu . Since the production of ^{244}Cm and higher isotopes of Cm are dependent on ^{243}Am , at the end of each cycle, they decrease in a similar manner with the increasing number of cycles as those pertaining to ^{243}Am . The amount of individual and total fission products produced in each cycle remains the same.

Table 1. Nuclide inventory (gramme per TIHM) at the charge and discharge of core fuel in different fuel cycles for PuR in a 1250 MWe LMFBFR

	Cycle 1		Cycle 3		Cycle 5		Cycle 7		Cycle 10	
	Chg. ^a	Disch. ^b	Chg.	Disch.	Chg.	Disch.	Chg.	Disch.	Chg.	Disch.
²³⁵ U	1.68+3*	6.75+2	1.60+3	6.58+2	1.59+3	6.56+2	1.59+3	6.53+2	1.58+3	6.52+2
²³⁶ U	0.0	2.05+2	0.0	1.98+2	0.0	1.98+2	0.0	1.98+2	0.0	1.99+2
²³⁸ U	8.14+5	7.19+5	7.99+5	7.07+5	7.94+5	7.03+5	7.92+5	7.01+5	7.90+5	7.00+5
²³⁷ Np	0.0	3.19+2	0.0	3.06+2	0.0	3.03+2	0.0	3.03+2	0.0	3.02+2
²³⁸ Pu	2.58+3	1.72+3	1.20+3	8.23+2	6.62+2	5.36+2	5.22+2	4.66+2	4.92+2	4.55+2
²³⁸ Pu	1.02+5	9.92+4	1.26+5	1.09+5	1.26+5	1.03+5	1.24+5	1.08+5	1.23+5	1.07+5
²⁴⁰ Pu	4.67+4	5.18+4	5.70+4	6.20+4	6.49+4	6.79+4	6.92+4	7.10+4	7.20+4	7.30+4
²⁴¹ Pu	2.46+4	1.08+4	6.87+3	6.76+3	6.54+3	7.20+3	7.01+3	7.61+3	7.38+3	7.90+3
²⁴² Pu	9.22+3	9.79+3	8.66+3	6.69+3	6.83+3	6.36+3	5.95+3	5.79+3	5.51+3	5.52+3
²⁴¹ Am	0.0	1.56+3	6.88+2	0.0	7.09+2	7.09+2	0.0	7.55+2	0.0	7.88+2
²⁴³ Am	0.0	1.51+3	0.0	1.23+3	0.0	9.93+2	0.0	8.86+2	0.0	8.34+2
²⁴² Cm	0.0	1.31+2	0.0	5.18+1	0.0	5.27+1	0.0	5.61+1	0.0	5.87+1
²⁴⁴ Cm	0.0	3.62+2	0.0	2.98+2	0.0	2.37+2	0.0	2.10+2	0.0	1.96+2
²⁴⁵ Cm	0.0	2.39+1	0.0	1.96+1	0.0	1.55+1	0.0	1.37+1	0.0	1.27+1
²⁴⁶ Cm	0.0	4.08-1**	0.0	5.75-1	0.0	4.53-1	0.0	3.98-1	0.0	3.70-1
²⁴⁸ Cm	0.0	3.24-4	0.0	2.56-4	0.0	2.00-4	0.0	1.75-4	0.0	1.63-4
²⁵² Cr	0.0	2.32-11	0.0	1.72-11	0.0	1.33-11	0.0	1.16-11	0.0	1.07-11
Total act.	4.00+6	8.97+5	1.00+6	8.97+5	1.00+6	8.97+5	1.00+6	8.97+5	1.00+6	8.97+5
⁹⁰ Sr	0.0	7.99+2	0.0	8.10+2	0.0	8.08+2	0.0	8.07+2	0.0	8.06+2
⁹⁵ Mo	0.0	1.75+3	0.0	1.77+3	0.0	1.77+3	0.0	1.77.3	0.0	1.77+3
⁹⁹ Tc	0.0	2.33+3	0.0	2.35+3	0.0	2.35+3	0.0	2.35+3	0.0	2.35+3
¹¹⁰ Pu	0.0	2.54+3	0.0	2.55+3	0.0	2.55+3	0.0	2.55+3	0.0	2.55+3
¹⁰⁶ Pu	0.0	8.43+2	0.0	8.22+2	0.0	8.24+2	0.0	8.26+2	0.0	8.27+2
¹⁰³ Rb	0.0	2.61+3	0.0	2.63+3	0.0	2.63+3	0.0	2.63+3	0.0	2.63+3
¹⁰⁵ Pd	0.0	2.21+3	0.0	2.19+3	0.0	2.19+3	0.0	2.19+3	0.0	2.19+3
¹⁰⁷ Pd	0.0	1.34+3	0.0	1.28+3	0.0	1.28+3	0.0	1.29+3	0.0	1.29+3
¹⁰⁰ Ag	0.0	7.04+2	0.0	7.39+2	0.0	7.39+2	0.0	7.41+2	0.0	7.42+2
¹³¹ Xe	0.0	1.99+3	0.0	2.03+3	0.0	2.03+3	0.0	2.03+3	0.0	2.03+3
¹³³ Cs	0.0	3.64+3	0.0	3.66+3	0.0	3.66+3	0.0	3.66+3	0.0	3.66+3
¹³⁴ Cs	0.0	2.19+2	0.0	2.14+2	0.0	2.13+2	0.0	2.12+2	0.0	2.12+2
¹³⁵ Cs	0.0	4.26+3	0.0	4.27+3	0.0	4.27+3	0.0	4.27+3	0.0	4.27+3
¹³⁷ Cs	0.0	3.76+3	0.0	3.75+3	0.0	3.75+3	0.0	3.75+3	0.0	3.75+3
¹⁴⁴ Ce	0.0	8.21+2	0.0	8.12+2	0.0	8.12+2	0.0	8.12+2	0.0	8.12+2
¹⁴¹ Pr	0.0	3.02+3	0.0	3.03+3	0.0	3.03+3	0.0	3.03+3	0.0	3.03+3
¹⁴³ Nd	0.0	2.55+3	0.0	2.54+3	0.0	2.54+3	0.0	2.54+3	0.0	2.54+3
¹⁴⁵ Nd	0.0	1.85+3	0.0	1.83+3	0.0	1.83+3	0.0	1.83+3	0.0	1.83+3
¹⁴⁷ Pm	0.0	7.88+3	0.0	7.84+2	0.0	7.84+2	0.0	7.84+2	0.0	7.85+2
¹⁴⁹ Sm	0.0	6.16+2	0.0	6.12+2	0.0	6.12+2	0.0	6.13+2	0.0	6.13+2
¹⁵¹ Sm	0.0	3.70+2	0.0	3.68+2	0.0	3.68+2	0.0	3.69+2	0.0	3.69+2
Total F. P.	0.0	1.03+5	0.0	1.03+5	0.0	1.03+5	0.0	1.03+5	0.0	1.03+5
Total F. P. + act.	1.00+6	1.00+6	1.00+6	1.00+6	1.00+6	1.00+6	1.00+6	1.00+6	1.00+6	1.00+6

* 1.68+3 = 1.68 10³** 4.08-1 = 4.08 10⁻¹^a Charge corresponds to the beginning of an irradiation cycle^b Discharge corresponds to the end of an irradiation cycle

AFR

The nuclide inventory per TIHM at the charge and discharge of the core fuel in each cycle of the AFR is given in tab. 2. The total amount of fuel per TIHM up to the 10th fuel cycle is decreased by about 31%. This is because of the reduction in the density of the fuel due to the accumulation of NVFP. The NVFP accumulated in the AFR have smaller densities than actinides. This effect is indicated by the swelling of the fuel pin. The density of the pseudo (average) non-volatile fission product oxide is taken to be 6.4 g/cc [32]. Since the reactor configuration is fixed, the volume of the fuel is constant. By refabrication of fuel elements, the mass of the fuel containing NVFP going into a constant volume will be less than that of the fuel which does not contain fission products. All of the fuel obtained through AIROX reprocessing cannot be used in the refabrication of fuel elements to be reintroduced into the reactor. The surplus fuel is removed and placed in a temporary storage.

The Pu vector has changed from LWR Pu vector $^{238}\text{Pu}/^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}/^{242}\text{Pu}$ equal to 1.4/55.0/25.3/13.3/5.0, respectively, at the beginning of the first cycle, to 0.4/69.8/25.1/2.7/2.0 at the beginning of the 10th cycle. The behavior of different isotopes of Pu with an increase in the number of cycles in an AFR is given in fig. 3. The curves for Pu isotopes in AFR have almost the same structure as those in PuR. The deviations of the curves in AFR relative to PuR are explained below:

The amounts of ^{239}Pu and ^{238}Pu are larger in AFR than in PuR, while those of ^{240}Pu , ^{241}Pu , and ^{242}Pu are smaller. In principle, all Pu isotopes should have smaller amounts in AFR than in PuR because, during re-fabrication, a fraction of Pu isotopes is taken out along with the fuel removed from the recycled fuel. However, ^{238}Pu and ^{239}Pu are exceptions to this rule. The fuel is removed because of the reduction in the density of the recycled fuel due to the accumulation of NVFP. Fuel is also removed so as to make room for the addition of fissile material in order to maintain the criticality of the reactor. Figure 4 shows the amount of different materials taken out along with the removed recycled fuel during the re-fabrication of fuel elements in different cycles.

The amount of ^{238}Pu is larger in AFR than that in PuR because of the recycling of ^{237}Np and ^{241}Am in AFR. ^{238}Pu is produced through neutron captures in ^{237}Np and by the decay of ^{242}Cm produced through captures in ^{241}Am . In PuR, ^{237}Np and ^{241}Am are not recycled.

The amount of ^{239}Pu is larger in AFR than in PuR. This is mainly because of the accumulation of fission products and minor actinides which produce negative reactivity in the reactor. This negative reactivity is compensated by adding the fissile material, mainly ^{239}Pu .

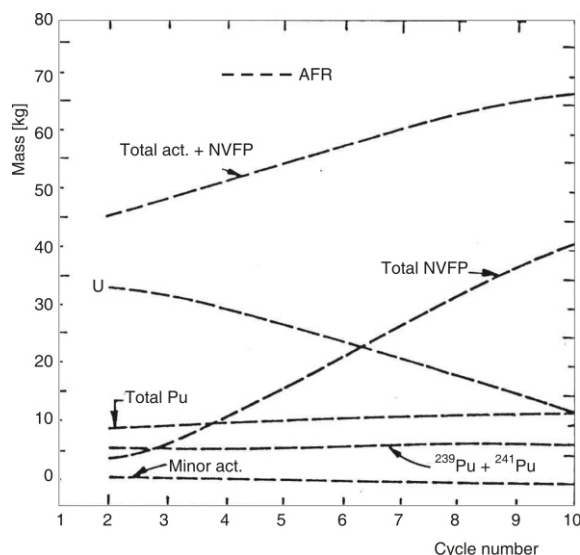


Figure 4. Amount of different materials removed from the recycled core fuel during re-fabrication in AFR

In AFR, the amount of ^{239}Pu decreases slowly with the increasing number of cycles after the 3rd cycle. This is mainly due to the following reasons.

- The amount of fission products taken out in each cycle along with the removed recycled fuel increases with the increasing number of cycles (fig. 4) because the concentration of fission products in the fuel increases. As a consequence, with the increase in the number of cycles, less fission products are added to the fuel (fig. 5). Therefore, in order to compensate their effect, the increase in the amount of ^{239}Pu is less required in later than in the earlier cycles.

- In an LMFBR, fission products important for absorption (e.g. ^{105}Pd , ^{101}Ru , ^{103}Rh , ^{99}Tc , ^{133}Cs , ^{107}Pd , ^{149}Sm , ^{147}Pm , ^{151}Sm , ^{145}Nd , ^{143}Nd , and ^{95}Mo) which

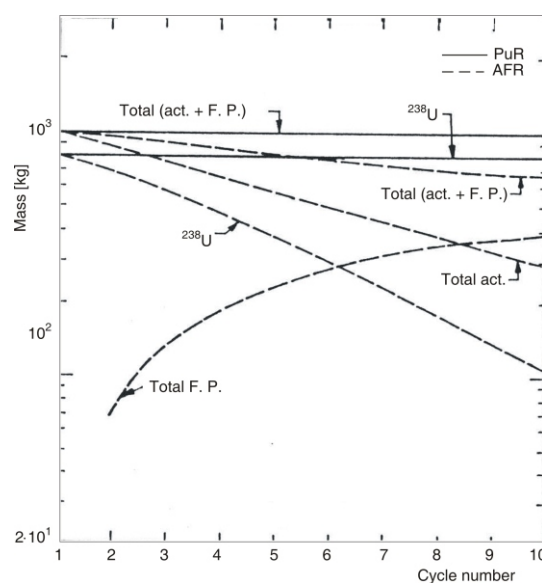


Figure 5. Amount of actinides and fission products per TIHM present in the core fuel at the beginning of different fuel cycles in PuR and AFR

Table 2. Nuclide inventory (gramme per TIHM) at the charge and discharge of core fuel in different fuel cycles for PuR in a 1250 MWe LMFBR

	Cycle 1		Cycle 3		Cycle 5		Cycle 7		Cycle 10	
	Chg. ^a	Disch. ^b	Chg.	Disch.	Chg.	Disch.	Chg.	Disch.	Chg.	Disch.
²³⁵ U	1.68+3*	6.75+2	2.54+2	1.15+2	5.72+1	3.87+1	2.97+1	2.58+1	1.82+1	1.60+1
²³⁶ U	0.0	2.05+2	2.30+2	2.18+2	1.78+2	1.53+2	1.21+2	1.04+2	7.01+1	6.14+1
²³⁸ U	8.14+5	7.19+5	5.64+5	4.95+5	3.78+3	3.27+5	2.38+5	2.02+5	1.06+5	8.71+4
²³⁷ Np	0.0	3.19+2	4.18+2	4.48+2	3.84+2	3.47+2	2.64+2	2.26+2	1.26+2	1.05+2
²³⁸ Pu	2.50+3	1.72+3	1.58+3	1.49+3	1.37+3	1.24+3	1.11+3	9.82+2	7.62+2	6.54+2
^{238m} Pu	1.02+5	9.92+4	1.34+5	9.48+4	1.32+2	7.75+4	1.29+5	6.14+4	1.26+5	4.30+4
²⁴⁰ Pu	4.67+4	5.18+4	5.29+4	5.83+4	5.50+4	5.90+4	5.34+4	5.50+4	4.53+4	4.65+4
²⁴¹ Pu	2.46+4	1.08+4	6.28+3	6.37+3	5.89+3	6.46+3	5.61+3	6.25+3	4.95+3	3.90+3
²⁴² Pu	9.22+3	9.79+3	7.91+3	7.12+3	5.72+3	5.42+3	4.51+3	4.53+3	3.56+3	3.72+3
²⁴¹ Am	0.0	1.56+3	1.92+3	1.50+3	1.52+3	1.22+3	1.31+3	1.03+3	9.97+2	7.48+2
²⁴³ Am	0.0	1.51+3	2.04+3	2.29+3	2.02+3	1.97+3	1.62+3	1.58+3	1.20+3	1.23+3
²⁴² Cm	0.0	1.31+2	3.33+1	1.58+2	2.88+1	1.40+32	2.73+1	1.37+24	2.47+2	1.22+2
²⁴⁴ Cm	0.0	3.62+2	8.82+2	1.46+3	1.56+3	1.80+3	1.54+3	1.63+3	1.20+3	1.29+3
²⁴⁵ Cm	0.0	2.39+1	1.04+2	2.18+2	2.83+2	3.50+2	3.31+2	3.48+2	2.09+2	2.76+2
²⁴⁶ Cm	0.0	7.08-1**	6.56+0	2.23+1	4.18+1	6.95+1	8.37+1	1.07+2	1.06+2	1.17+2
²⁴⁸ Cm	0.0	3.24-4	1.18-2	9.05-2	3.04-1	7.95-1	1.38+0	2.39+0	3.75+0	4.82+0
²⁵² Cr	0.0	2.32-11	1.05-8	5.11-7	3.96-6	3.10-5	8.05-5	3.11-4	8.50-4	1.93-3
Total act.	1.00+6	8.97+5	7.73+5	6.70+5	5.96+5	4.83+5	4.38+5	3.36+5	2.90+5	1.91+5
⁹⁰ Sr	0.0	7.99+2	1.36+3	2.05+3	2.29+3	2.89+3	2.85+3	3.39+3	3.18+3	3.57+3
⁹⁵ Mo	0.0	1.75+3	3.51+3	4.96+3	5.97+3	7.12+3	7.39+3	8.30+3	8.01+3	8.66+3
⁹⁹ Tc	0.0	2.33+3	4.00+3	5.74+3	6.44+3	7.74+3	7.65+3	8.67+3	8.01+3	8.84+3
¹¹⁰ Pu	0.0	2.54+3	2.13+3	4.37+3	2.62+3	4.84+3	2.71+3	4.97+3	2.03+3	4.96+3
¹⁰⁶ Pu	0.0	8.43+2	2.44+2	8.57+2	2.41+2	8.58+2	2.37+2	8.52+2	2.28+2	8.31+2
¹⁰³ Rb	0.0	2.61+3	4.66+3	6.43+3	7.23+3	8.34+3	8.16+3	8.83+3	7.80+3	8.21+3
¹⁰⁵ Pd	0.0	2.21+3	3.66+3	5.14+3	5.67+3	6.77+3	6.66+3	7.57+3	7.03+3	7.83+3
¹⁰⁷ Pd	0.0	1.34+3	2.28+3	3.37+3	3.92+3	4.95+3	5.13+3	6.13+3	5.26+3	7.23+3
¹⁰⁰ Ag	0.0	7.64+2	1.32+3	1.96+3	2.29+3	2.90+3	3.03+3	3.67+3	3.89+3	4.59+3
¹³¹ Xe	0.0	1.99+3	1.01+2	2.13+3	1.00+2	2.12+3	9.80+1	2.10+3	9.44+1	2.06+3
¹³³ Cs	0.0	3.64+3	3.50+1	3.66+3	3.42+1	3.61+32	3.30+1	3.53+3	3.12+1	3.39+3
¹³⁴ Cs	0.0	2.19+2	1.54+0	2.38+3	1.68+0	2.68+2	1.87+0	3.05+2	2.22+0	3.64+2
¹³⁵ Cs	0.0	4.26+3	4.07+1	4.31+2	4.01+1	4.28+3	3.92+1	4.25+3	3.78+1	4.17+3
¹³⁷ Cs	0.0	3.76+3	3.49+1	3.78+3	3.43+1	3.76+3	3.36+1	3.73+3	3.24+1	3.86+3
¹⁴⁴ Ce	0.0	8.21+2	3.21+2	8.21+2	3.11+2	8.06+2	3.01+2	7.88+23	2.83+2	7.54+2
¹⁴¹ Pr	0.0	3.02+3	5.69+3	8.49+3	1.02+4	1.27+4	1.33+4	1.56+4	1.56+4	1.77+4
¹⁴³ Nd	0.0	2.55+3	4.55+3	6.67+3	7.72+3	9.47+3	8.59+3	1.10+4	1.05+4	1.17+4
¹⁴⁵ Nd	0.0	1.85+3	3.21+3	4.70+3	5.38+3	6.62+3	6.67+3	7.72+3	7.40+3	8.30+3
¹⁴⁷ Pm	0.0	7.88+3	7.31+2	1.06+3	8.02+2	1.13+3	8.40+2	1.20+3	8.92+2	1.29+3
¹⁴⁹ Sm	0.0	6.16+2	8.86+2	1.14+3	1.20+3	1.39+3	1.36+3	1.56+3	1.49+3	1.67+3
¹⁵¹ Sm	0.0	3.70+2	5.32+2	7.04+2	7.56+2	9.16+2	9.18+3	1.09+3	1.09+3	1.27+3
Total F. P.	0.0	1.03+5	1.32+5	2.34+5	2.39+5	3.41+5	3.20+5	4.22+5	3.93+5	4.98+5
Total F. P. + act	1.00+6	1.00+6	9.05+5	9.04+5	8.24+5	8.24+5	7.58+5	7.58+5	6.83+5	6.89+5

* 1.68+3 = 1.68 10³** 7.08-1 = 7.08 10⁻¹^a Charge corresponds to the beginning of an irradiation cycle^b Discharge corresponds to the end of an irradiation cycle

produce the negative reactivity in a reactor, increase less rapidly with the increasing number of cycles and reach almost an equilibrium after the fifth cycle (fig. 6). This is because, as the concentration of fission products increases with the increasing number of cycles, larger amounts of important fission products are converted to less important fission products, due to neutron capture (for example, ^{105}Pd , which has a relatively higher capture cross-section ($\sigma_c = 0.8 \text{ b}^*$), is converted to ^{106}Pd ($\sigma_c = 0.23 \text{ b}$). As a consequence, the amount of ^{239}Pu required to compensate for the negative reactivity of NVFP increases less rapidly with the increasing number of cycles.

– In an AFR, the amount of ^{238}U decreases almost constantly, along with the increasing number of cycles. This decrease in the amount of ^{238}U , as mentioned previously, is due to the reduction in the density of the fuel, as well as to its removal along with the removed recycled fuel, so as to make room for the fissile material which needs to be added to guarantee the criticality of the reactor. Since ^{238}U has a very small negative specific reactivity worth, the decrease in the amount of ^{238}U gives rise to positive reactivity. This means that somewhat less fissile material, *i. e.* ^{239}Pu , is required to maintain the same criticality at the beginning of each cycle. In earlier cycles, this positive effect in reactivity is compensated by the large negative effect in reactivity due to the large increase in important NVFP, since NVFP have a comparatively large negative specific reactivity as compared to ^{238}U . This is why the amount of ^{239}Pu required for the criticality of the reactor increases in earlier cycles.

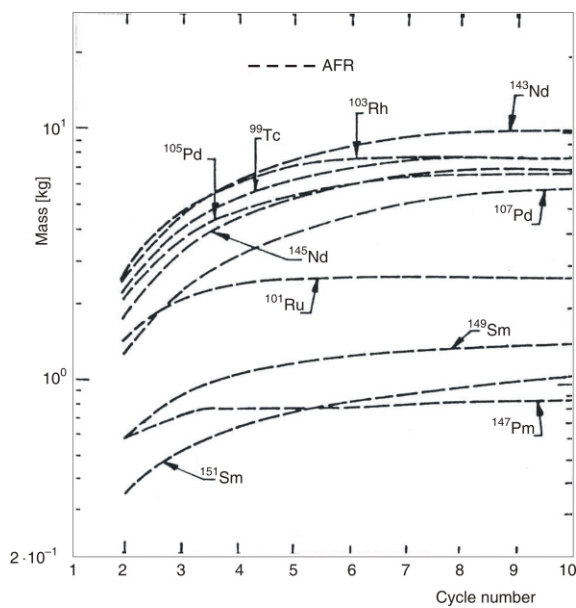


Figure 6. Amount of different fission products per TIHM present in the core fuel at the beginning of different fuel cycles in AFR

* $\text{b} = 10^{-28} \text{ m}^2$

In later cycles, the important NVFP increase less rapidly or almost reach equilibrium, so that the effect of positive reactivity due to the decrease in the amount of ^{238}U becomes dominant over the negative effect due to NVFP. Therefore, in later cycles, the amount of the ^{239}Pu required to maintain the criticality of the reactor decreases.

– The amount of minor actinides increases in the first few cycles and then decreases with the increase in the number of cycles, as depicted in fig. 7, this being particularly true of ^{241}Am which has a high negative specific reactivity worth, on the decline with the rise in the number of cycles. ^{241}Am decreases because ^{241}Pu decreases and, due to the large absorption cross-section of the former, it does not get accumulated. ^{243}Am which also has a negative specific reactivity worth, increases for the first four cycles and then decreases. In the first few cycles, it gets accumulated because its production rate through captures in ^{242}Pu is higher than its destruction through absorption. In later cycles, as its concentration is on the increase, its destruction rate through absorption also rises. On the other hand, its production rate through captures in ^{242}Pu decreases because ^{242}Pu decreases with the increasing number of cycles. Therefore, the amount of ^{243}Am will decrease in later cycles. ^{244}Cm , having a small positive reactivity, increases rapidly in earlier cycles and then slowly

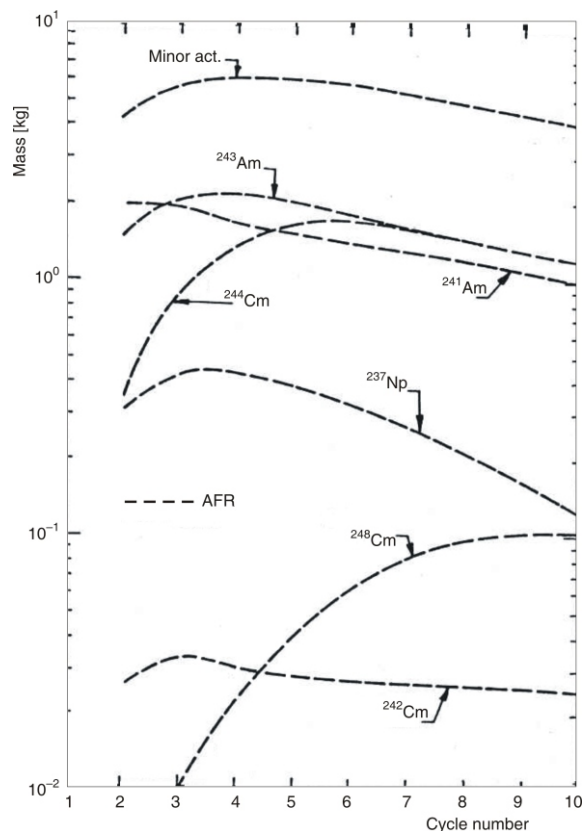


Figure 7. Amount of minor actinides per TIHM present in the core fuel at the beginning of different fuel cycles in AFR

decreases over later cycles. The reason for this behavior is similar to that of ^{243}Am . ^{244}Cm is produced through captures in ^{243}Am . The behavior of ^{241}Am , ^{243}Am , and ^{244}Cm gives rise to positive reactivity in the reactor in later cycles. Therefore, instead of an increase as in earlier cycles, the amount of ^{239}Pu will decrease, due to minor actinides in the later cycles.

In an AFR, in contrast to PuR, the amount of ^{239}Pu decreases in later cycles. This is due mainly to the following reasons:

- the production of ^{240}Pu through neutron captures in ^{239}Pu decreases because the amount of ^{239}Pu decreases with the increasing number of cycles, and
- along with the recycled fuel, a certain amount of ^{239}Pu is also removed during the re-fabrication of fuel elements. This fuel is removed because of the reduction in the density of the fuel, as well as so to make room for the fissile material which has to be added to guarantee the criticality of the reactor.

Amount of fissile material added to guarantee the criticality of the reactor

To guarantee the criticality of the reactor, a certain amount of fissile material has to be added during the fabrication/re-fabrication of core fuel elements. The amount of fissile material ($^{239}\text{Pu} + ^{241}\text{Pu}$) added per TIHM with respect to fuel cycle numbers of the individual recycling schemes in an LMFBR is given in fig. 8. The first cycle is the same, both in PuR and AFR. The core fuel in the first cycle is enriched with LWR Pu. In the second and later cycles, only core Pu is recycled in PuR, while all actinides and NVFP are recycled in AFR. The fissile material added in these cycles to achieve the criticality of the reactor during the re-fabrication of core fuel elements is blanket Pu, mainly ^{239}Pu (~95%). The behavior of the curves is shown in fig. 8, upon which the second cycle can be explained as follows.

PuR

In PuR, the amount of fissile material added during the re-fabrication of core fuel elements decreases slowly with the increasing number of cycles and then almost reaches equilibrium. This slight decrease is due to an increase in the amount of ^{240}Pu in the recycled core fuel which adds positive reactivity to the reactor, as its specific reactivity worth is slightly positive. The cumulative amount of fissile material required to be added per TIHM of the PuR core fuel, in the 10 fuel cycles, including the first one, is about 292 kg.

AFR

In an AFR, the amount of fissile material added to guarantee the criticality of the system during the re-fabrication of core fuel elements increases with the increasing number of cycles and is higher than in PuR.

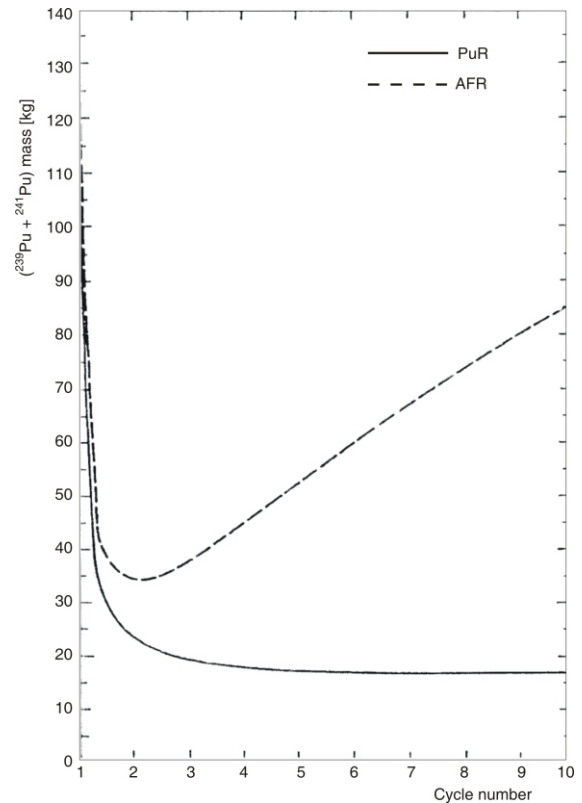


Figure 8. Amount of fissile material ($^{239}\text{Pu} + ^{241}\text{Pu}$) added per TIHM during fabrication/re-fabrication of the core fuel to maintain the criticality of the reactor for different recycling schemes in an LMFBR

This higher amount of required fissile material is mainly due to following reasons:

- because of the accumulation of fission products in the recycled fuel, a negative reactivity is produced which has to be compensated by the addition of fissile material,

– in an AFR, the amount of ^{238}U decreases with the increasing number of cycles, as demonstrated in fig. 5. This decrease in ^{238}U causes a reduction in the buildup of ^{239}Pu through neutron captures during the irradiation cycle. Therefore, in AFR, the burn-up swing, *i. e.* the difference between the amount of fissile material ($^{239}\text{Pu} + ^{241}\text{Pu}$) per TIHM at the beginning and at the end of an irradiation cycle, increases with the increasing number of cycles, as shown in fig. 9. This difference is higher in AFR than in PuR because of the decrease in ^{238}U , as mentioned above. To guarantee the criticality of the reactor in the next cycle, this disparity in the fissile material has to be compensated in the re-fabrication of fuel elements, requiring higher amounts of fissile material in an AFR than in a PuR scheme. Furthermore, due to the decrease in ^{238}U , its contribution to fast fissions in the system will also decrease, leading to a further increase in the amount of fissile material to be added, and

- as already mentioned, a certain amount of fissile material is taken out along with the fuel removed

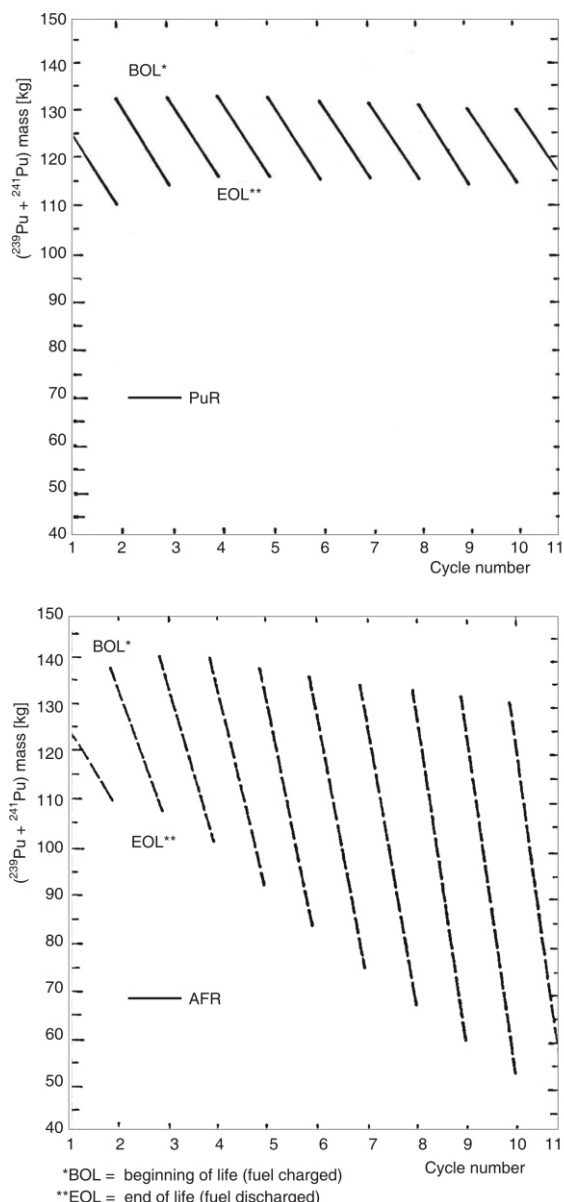


Figure 9. Amount of fissile material ($^{239}\text{Pu} + ^{241}\text{Pu}$) per TIHM present in the core at the charge and discharge of the fuel in each fuel cycle in PuR and AFR

from the recycled material. The amount of fissile material taken out needs to be compensated for.

In an LMFBR, the necessary amount of fissile material per cycle to be added to the reactor for its criticality increases almost constantly with the increasing number of cycles. This is because, with the increasing number of cycles, the decline in the amount of ^{238}U remains almost constant and, therefore, the production of ^{239}Pu during an irradiation cycle decreases constantly, as discussed above. Although important NVFP almost reach equilibrium after cycle 5 (fig. 6), the amount of fissile material to be added due to this effect is small compared to that added due to the decrease in ^{238}U . Because of the small capture cross-sections of the NVFP in a fast reactor spectrum, the fissile mate-

rial penalty due to NVFP is not as severe in fast reactors as in thermal reactors. The fissile material penalty in fast reactors comes mainly from the reduction in the amount of ^{238}U . However, the decrease in ^{238}U is a consequence of the accumulation of NVFP, as discussed in this sub-section.

The cumulative amount of fissile material ($^{239}\text{Pu} + ^{241}\text{Pu}$) required to be added per TIHM of core fuel in an AFR over the 10 cycles, including the first cycle, is about 664 kg. This is approx. 128% more than required for PuR.

Cumulative high active waste per TIHM

The cumulative waste per TIHM from the core fuel in PuR and AFR is calculated after 10 cycles, according to the assumption described in the sub-section *Determination of cumulative high active waste (HAW) per TIHM*. The amount of different nuclides in this cumulative waste, seven years after the discharge of the fuel from the 10th cycle, is presented in tab. 3. As mentioned above, the activity, decay heat, ingestion hazards and neutron radiation in the cumulative waste from core fuel in different recycling schemes and from the axial and radial blanket fuel with respect to waste decay times are given in fig. 10. The ingestion hazard (annual ingestion hazard) is defined as the volume of water required to dilute it to its maximum permissible concentration for continuous ingestion by an occupationally exposed individual per year. In this paper, the ingestion hazard is calculated according to German limits for the annual intake [33], an unit commonly used in literature.

Before discussing the results in detail, it is helpful to keep in mind the following general conclusions obtained from fig. 10:

- for the first three centuries, the level of activity, nuclear waste decay heat and ingestion hazards were dominated by fission products, namely, ^{90}Sr and ^{137}Cs . Other major contributors to this activity are ^{241}Am , ^{241}Pu , ^{244}Cm , and, as to the decay heat, those of ^{238}Pu , ^{241}Am , and ^{244}Cm , respectively. ^{241}Am is either present in the cumulative waste or built up through the decay of ^{241}Pu after the discharge,
- after approx. 3000 to 6 10^4 years, the main contributors to these parameters are ^{243}Am , ^{240}Pu , and ^{239}Pu ,
- after approx. 6 10^4 years, the long-lived fission products, namely, ^{99}Tc , ^{129}I , and ^{135}Cs , contribute to the level of activity and decay heat, in addition to actinides like ^{239}Pu , ^{237}Np , and its daughter-products (e. g. ^{229}Th , ^{217}Th , ^{213}Po , and ^{213}Bi). The ingestion hazard after about 6 10^4 years is dominated by ^{226}Ra , a daughter-product of ^{234}U , and
- over the first century of decay, major contributors to neutron radiation in the waste come from ^{244}Cm

Table 3. Cumulative waste (gramme per TIHM) of the core fuel for different recycling schemes in a 1250 MWe LMFBR

	PuR	AFR	
		AFR (S1)	AFR (S2)
²³² U	2.31-4	3.56-4	7.12-3
²³⁵ U	7.32+1	1.23+1	1.46+2
²³⁸ U	7.05+4	2.27+4	3.20+5
²³⁷ Np	3.58+3	4.53+2	4.53+2
²³⁸ Pu	1.96+2	7.37+1	1.49+3
²³⁹ Pu	1.07+4	5.12+3	1.00+5
²⁴⁰ Pu	8.04+3	3.85+3	8.61+4
²⁴¹ Pu	2.68+2	1.60+2	5.43+3
²⁴² Pu	6.86+2	4.01+2	7.75+3
²⁴¹ Am	1.19+4	6.20+3	6.20+3
²⁴³ Am	1.06+4	2.47+3	2.47+3
²⁴⁴ Cm	9.78+2	1.42+3	1.42+3
²⁴⁵ Cm	1.65+2	4.68+2	4.68+2
²⁴⁶ Cm	4.83+0	1.67+2	1.67+2
²⁴⁸ Cm	2.16-3	5.98+0	5.98+0
U	7.07+4	2.27+4	3.21+5
Np	3.58+3	4.53+2	4.53+2
Pu	1.99+4	9.61+3	2.01+5
Am	2.28+4	8.79+3	8.79+3
Cm	1.17+3	2.10+3	2.10+3
Minor act.	2.78+4	1.13+4	1.13+4
Total act.	1.18+5	4.38+4	5.33+5
⁹⁰ Sr	4.62+3	4.28+3	4.28+3
⁹⁹ Tc	2.36+4	1.39+4	1.39+4
¹³⁷ Cs	2.18+4	2.15+4	2.15+4
¹⁵¹ Sm	3.06+3	1.75+3	1.75+3
Total F. P.	1.03+6	1.03+6	1.03+6
Total waste	1.15+6	1.07+6	1.56+6

and ²⁴⁶Cm. Then, up to about to 5 10⁴ years, they are replaced by ²⁴⁶Cm and, later on, ²⁴⁸Cm and ²⁴²Pu take over their role.

These general conclusions, along with the amount of cumulative waste given in tab. 3, are meant to aid the understanding of the results plotted in fig. 10.

PuR

The amount of activity, decay heat, ingestion hazard, and neutron radiation with respect to waste decay times in the cumulative waste per TIHM of the PuR core fuel is given in fig. 10. The main contributors to these parameters regarding the waste decay times under consideration are those mentioned above.

AFR

The results for the cumulative waste per TIHM of the core fuel calculated by the two strategies are compared first and, then, to PuR.

AFR(S1) vs. AFR(S2)

The amount of U and Pu in the cumulative waste is higher by about a factor of 14 to 20, respectively, in an AFR(S2) in comparison to AFR(S1), because in

AFR(S2) these elements are not separated through the aqueous reprocessing of its last cycle. Due to these elements (especially because of the presence of ²⁴¹Pu and its decay into ²⁴¹Am), the amount of activity, decay heat and ingestion hazard after about 250 years to 10⁶ years of waste decay time is by a factor of about 2 to 11 higher in AFR(S2) than in AFR(S1). For the first two to three centuries, the amount of decay heat is higher by about 10% to 200% in AFR(S2) than in an AFR(S1), mainly because of the higher amounts of ²³⁸Pu and ²⁴¹Am (produced through decay of ²⁴¹Pu) in an AFR(S2), as opposed to AFR(S1).

AFR vs. PuR

The cumulative mass of total actinides in the waste is about 62% smaller in AFR(S1) compared to that of PuR, mainly because of the very small losses assumed in the AIROX process, as shown in tab. 3. The cumulative mass of Am and Np decreases by about 60% and 90%, respectively, while that of Cm, which is a strong neutron emitter through spontaneous fission, increases by about 80%. From the point of view of short and long-term waste hazard, among fission products, ⁹⁰Sr, ¹³⁷Cs, ⁹⁹Tc, ¹²⁹I, and ¹³⁵Ca are important contributors. Cs and I, being volatile, are not recycled. The transmutation of ⁹⁰Sr is very small (only about 7%) because it has a very small capture cross-section ($\sigma_c = 0.013$ b) in a fast reactor spectrum. Fission product ⁹⁹Tc ($\sigma_c = 0.55$ b) is reduced by about 40%.

At first, for nearly two centuries, the amount of activity, decay heat and ingestion hazard is almost the same in both AFR(S1) and PuR. This is because the main contributors (*i. e.* ⁹⁹Sr and ¹³⁷Cs) to these parameters in the decay times under consideration remain about the same both in AFR(S1) and PuR. After about 200 years, the amount of activity, decay heat and ingestion hazard is smaller in an AFR(S1) compared to that of PuR over the entire waste decay period considered in this paper (*i. e.* up to 10⁶ years). This is because of the smaller amounts of minor actinides and Pu isotopes in AFR(S1) than in PuR. At about 1000 years, the amount of activity, decay heat and ingestion hazard is smaller by about 50%, respectively, in AFR(S1) than in PuR.

As for AFR strategy 2, the cumulative mass of total actinides in the waste is for a factor of about 4.5 higher in AFR(S2) than the one pertaining to PuR, because in AFR(S2) the U and Pu are not separated through an aqueous reprocessing of the last cycle. Over the entire waste decay period considered in this paper, the amount of activity, decay heat and ingestion hazard is larger in AFR(S2), as compared to that in PuR, mainly because of the higher amount of Pu in AFR(S2) than in PuR. At about 1000 years, the amount of activity, decay heat and ingestion hazard is greater by a factor of about 2.3, respectively, in AFR(S2), as compared to that of PuR.

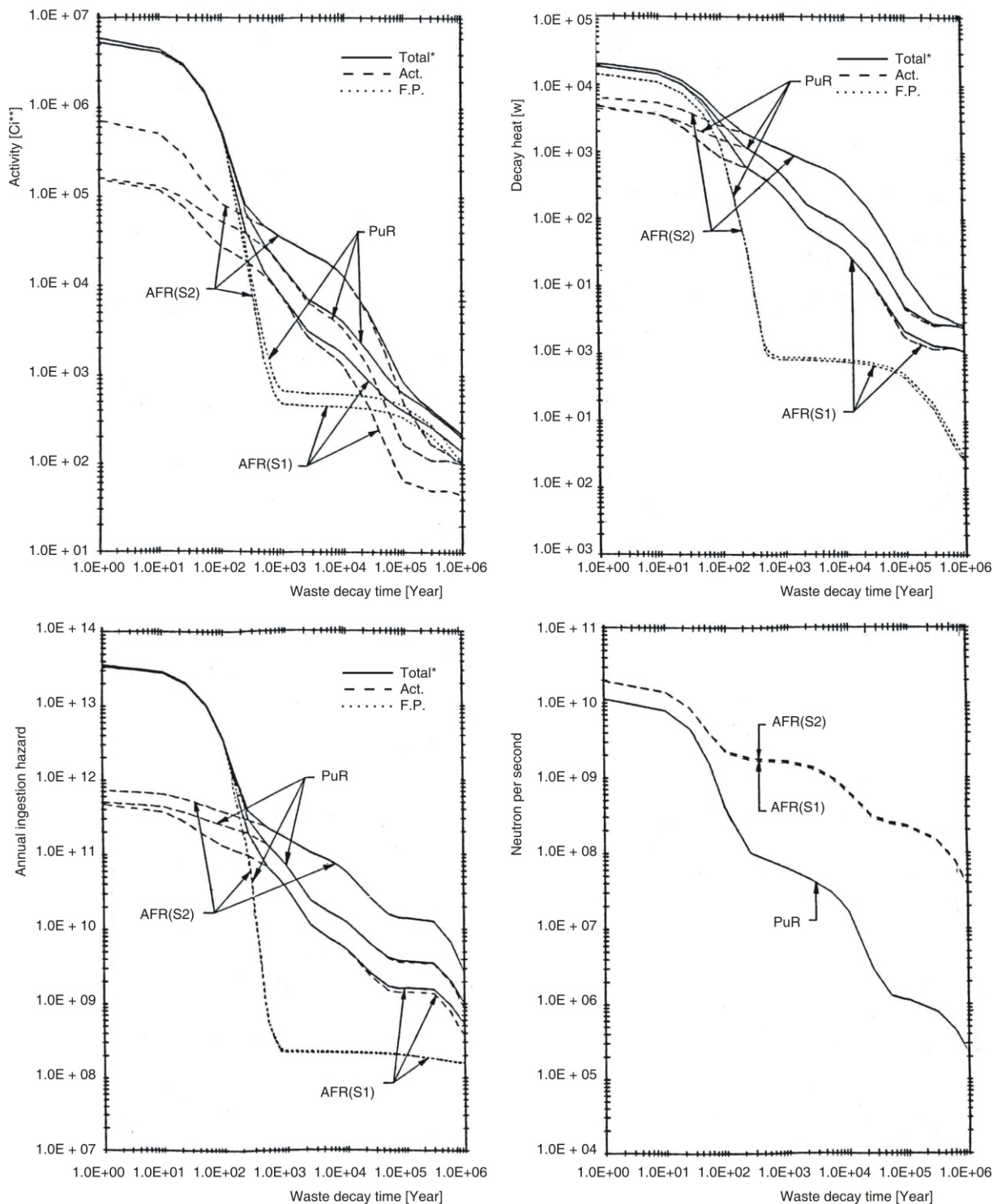


Figure 10. Amount of activity, decay heat, ingestion hazard and neutron radiation with respect to decay time in cumulative waste per TIHM of the core fuel produced through different recycling schemes, i.e. PuR, AFR(S1), and AFR(S2)

*Total actinides + fission products

**Ci = $3.7 \cdot 10^{10}$ Bq

The amount of neutron radiation is a factor of about 2 to 160 larger in AFR than in PuR over the waste decay time between one to 10^6 years. This is because of the high amounts of Cm produced in an AFR.

As far as waste hazards are concerned, these results show that, in an LMFBR, for waste decay times

up to about 200 years, there is no advantage of AFR(S1) over PuR. After this decay time, AFR(S1) is advantageous to PuR with respect to activity, decay heat and ingestion hazard of the cumulative waste, but no such advantage of the former over the latter exists with respect to neutron radiation. However, AFR(S2)

is disadvantageous to PuR in all respects, *i. e.* activity, decay heat, ingestion hazard and neutron radiation of the cumulative waste, over the entire waste decay period considered in this paper (*i. e.* up to 10^6 years). It has to be mentioned here that if the multiple recycling through the AIROX process is to be considered, then the AFR(S1) strategy should be the preferred one.

CONCLUSIONS

Multiple recycling of actinides and NVFP in fast reactors through the dry re-fabrication/reprocessing AIROX process has been the subject of this study. The results discussed in the previous section show that the recycling of actinide and fission products through the AIROX process produces a reduction in potential waste hazards and that the AFR scheme is beneficial in fast reactors. Multiple recycling seems to be one of the possible methods of reducing potential hazards of radioactive waste in the future.

ACKNOWLEDGEMENT

The help and guidance received from late Dr. H. Kuesters of Forschungszentrum, Karlsruhe, Germany, is hereby gratefully acknowledged.

AUTHOR CONTRIBUTIONS

A. Salahuddin performed the theoretical calculations and analysis. Both the authors contributed in manuscript writing and results discussion. M. Iqbal prepared figures also.

REFERENCES

- [1] Haug, H. O., Amount, Disposal and Relative Toxicity of Long-Lived Fission Products and Actinides in the Radioactive Wastes of Nuclear Fuel Cycles, KfK-2022, 1975
- [2] McKay, A., Destroying Actinides in Nuclear Reactors, *Nuclear Engineering International*, 23 (1978), 266, pp. 40-43
- [3] Kessler, G., Nuclear Fission Reactors – Potential Role and Risks of Converters and Breeders, Springer-Verlag Wien, 1982
- [4] Tedder, D. W., Finney, B. C., Blomeke, J. O., Chemical Processing Facilities for Partitioning Actinides from Nuclear Fuel Cycle Waste Mixtures, EUR-6929, 1980
- [5] Tedder, D. W., Waste Management Recycle Systems for Treating Mixed Oxide Fuel Fabrication Waste, *Proceedings*, International Symposium on Actinide Recovery from Waste and Low-Grade Sources, August 24-25, New York City, USA, 1981
- [6] Choi, H. B., Ko, W. I., Yang, M. S., Economic Analysis on Direct Use of Spent Pressurized Water Reactor Fuel in CANDU Reactors-I: DUPIC Fuel Fabrication Cost, *Nuclear Technology*, 134 (2001), 2, pp. 110-129
- [7] Choi, H. B., et al., Economic Analysis on Direct Use of Spent Pressurized Water Reactor Fuel in CANDU Reactors-II: DUPIC Fuel-Handling Cost, *Nuclear Technology*, 134 (2001), 2, pp. 130-148
- [8] Ko, W. I., et al., Economic Analysis on Direct Use of Spent Pressurized Water Reactor Fuel in CANDU Reactors-III: Spent DUPIC Fuel Disposal Cost, *Nuclear Technology*, 134 (2001), 2, pp. 149-166
- [9] Ko, W. I., Choi, H. B., Yang, M. S., Economic Analysis on Direct use of Spent Pressurized Water Reactor Fuel in CANDU Reactors-IV: DUPIC Fuel Cycle Cost, *Nuclear Technology*, 134 (2001), 2, pp. 167-186
- [10] Colby, L. J., et al., Comparative Fuel Cycle Evaluations – Low Decontamination Pyro-Processing vs. Aqueous Reprocessing,” Part 1 – Atomics International, Canoga Park, Cal., USA, NAA-SR-8036, 1963
- [11] Bodine, J. E., et al., Second-Cycle AIROX Reprocessing and Pellet Refabrication of Highly Irradiated Uranium Dioxide, Atomics International, Canoga Park, Cal., NAA-SR-11375, 1965
- [12] Warren, K. S., Ferris, E. M., Oxidation and Chlorination of UO_2 - PuO_2 , ORNL-3977, USA, 1966
- [13] Barleon, L., Dorner, S., Fission Products Recycling – A Method to Reduce Waste Disposal Problems, KfK-2540, 1978
- [14] Takeda, H., Hoshino, T., Segawa, T., Research and Development of the Voloxidation Process, Conference of Fast Reactor Fuel Reprocessing, Dounreay, 15-18 May, 1979, pp. 123-132
- [15] Ali, S. A., et al., Voloxidation Experiment with a Continuously Working Prototype Furnace, KfK-2940 1980
- [16] Guon, J., et al., Low Decontamination Reprocessing Studies on Irradiated Uranium Dioxide Reactor Fuel, Atomics International, Canoga Park, California, NAA-SR-7136, 1962
- [17] Fischer, U., Wiese, H. W., Improved and Consistent Determination of the Nuclear Inventory of Spent PWR Fuel on the Basis of Cell-Burnup Methods Using KORIGEN, KfK-3014 1983
- [18] Iqbal, M., et al., A Comparative Study to Investigate burn-up in Research Reactor Fuel Using Two Independent Experimental Methods, *Ann. of Nucl. Energy*, 28 (2001), 17, pp. 1733-1744
- [19] Bakhtyar, S., Salahuddin, A., Nuclide Inventory and Decay Characteristics of 5 MW PARR-1 HEU Spent Fuel Element, Technical Report, PINSTECH-126, PINSTECH, Islamabad, Pakistan, 1992
- [20] Bakhtyar, S., Mahmood, A., Salahuddin, A., burn-up Dependent Isotopic Concentrations and Decay Parameters of PARR-1 LEU Silicide Fuel, Technical Report, PINSTECH-154, PINSTECH, Islamabad, Pakistan, 1996
- [21] Rider, F., Compilation of Fission Product Yields, NEDO-12154-2(B), Vallecitos Nuclear Centre, USA, 1980
- [22] Mougnot, J. C., et al., The Super Phenix Fuel Cycle-Technical and Economical Outlooks, Fast Reactor Fuel Cycles, BNES, London, 1982
- [23] Croff, A. G., Mcadoo, J. W., Bjerke, M. A., LMFBR Models for the ORIGEN-2 Computer Code, ORNL/TM-7176, 1981 and ORNL/TM-7176/R1, 1983
- [24] Hoebel, W., Numerical Methods Used in the Two-Dimensional Neutron Diffusion Program DIXY, *Proceedings*, GAMM-Workshop on Fast Solution Methods for Discretized Poisson Equations, Karlsruhe, Germany, 1977, pp. 199-216
- [25] Kieffhaber, E., et al., The KFKINR-Set of Group Constants: Nuclear Data Basis and First Results of its Ap-

- plication to the Recalculation of Fast Zero-Power Reactors, KfK-1572, 1972
- [26] Abagjan, L. P., *et al.*, Multigroup Constants for Reactor Calculations, KfK-tr-144, 1964
- [27] Goel, B., Krieg, B., Status of the Nuclear Data Library KEDAK-4, KfK-3838, 1984
- [28] Broeders, I., *et al.*, MIGROS-3, A Code for the Generation of Group Constants for Reactor Calculations from Neutron Nuclear Data in KEDAK-Format, KfK-2388, 1977
- [29] Kiefhaber, E., KFKINR-2, An Improved Version of the KFKINR-Set of Group Constants, Private Communications, 1983
- [30] Gruppelaar, H., 26-Group Cross-Sections for Fission Products, ECN Petten, The Netherlands, Private Communications, Atomics International, 1982
- [31] Murphy Jr., D. J., Farr, W. M., Ganapol, B. D., Power Production and Actinide Elimination by Fast Reactor Recycle, *Nuclear Technology*, 45 (1979), pp. 299-306
- [32] Strausberg, S., Multiple Reprocessing and Refabrication Experiments on Simulated UO₂-FissiaPellets, NAA-SR-7138, Atomics International, Canoga Park, Cal., 1962
- [33] ***, Regulation on the Protection Against Damage Caused by Ionising Radiation (Radiation Protection Ordinance – Strisch V, from 13. 10. 1976) Federal Law Gazette (1976), 2905-95, Apendix 4, Addendum of the 26. 01. 1977, Federal Law Gazette 185 (1977), *Bundesgesetzblatt*, 185 (1977)

Received on May 5, 2012

Accepted on December 3, 2012

Асиф САЛАХУДИН, Масуд ИГБАЛ

**ПОРЕЂЕЊЕ ШЕМЕ ЗА РЕЦИКЛИРАЊЕ АКТИНИДА И ФИСИОНИХ
ПРОДУКАТА СА ШЕМОМ ЗА РЕЦИКЛИРАЊЕ ПЛУТОНИЈУМА
У БРЗИМ РЕАКТОРИМА**

Проучавано је рециклирање актинида и неиспарљивих продуката у брзим реакторима помоћу AIROX сувог репроцесирања, као могући начин да се смањи дугорочна потенцијална опасност од нуклеарног отпада у односу на ону која произилази из прераде у PUREX влажном процесу. Обављена су израчунавања ради поређења шеме за рециклирање актинида и продуката фисије са шемом за рециклирање плутонијума у брзом реактору. У ову сврху измењена је верзија KORIGEN кода развијена у Карлсруеу, у Немачкој, за прорачун генерисања и слабења изотопа. Начињена је потпуно нова библиотека приноса фисионих продуката за брзе реакторе којим је замењена стара KORIGEN библиотека. За потребе ове студије, стандардни 26-то групни сет нуклеарних података KFKINR проширен је додавањем пресека 13 значајних актинида и 68 најзначајнијих фисионих продуката. Потврђено је да ових 68 фисионих продуката сачињава 95% укупног приноса фисионих продуката и око 99.5% укупне апсорпције услед фисионих продуката у брзим реакторима. Такође је испитано која је количина фисибилног материјала потребна да се осигура критичност реактора током процеса рециклирања и прорачунат је кумулативни високоактивни отпад по тони иницијалног тешког метала. Резултати показују да рециклирање актинида и фисионих продуката у брзим реакторима AIROX поступком доводи до смањења потенцијалне опасности од радиоактивног отпада.

Кључне речи: брзи реактор, рециклирање, актинид, фисиони производи, плутонијум, PUREX, AIROX