

TRANSMUTATION OF AMERICIUM IN THERMAL REACTORS

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Abstract

If all plutonium is partitioned from spent fuel and transmuted, then americium and its daughter products are the main contributors to the radiotoxicity of spent fuel up to several thousands of years of storage. This paper describes the possible benefits when americium is irradiated in a thermal reactor.

After one million years of storage, Np-237 dominates the possible dose to the population due to leakage of actinides from underground disposal sites. Because Am-241 decays to Np-237, the radiotoxicity due to Np-237 is reduced by about 50% when Am-241 is transmuted to other nuclides upon irradiation in a thermal reactor. Secondly, the radiotoxicity of an americium sample can be reduced with a factor of 10 to 20 after irradiation for 3 to 6 years in a thermal neutron flux of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$. The strongly alpha and neutron emitting transmutation products can most probably not be recycled again, so a process is suggested in which americium is first irradiated and then put to final storage. It is shown that after a storage time of about one hundred years, the radiotoxicity of an irradiated americium sample is considerably reduced compared to the radiotoxicity of an unirradiated sample. The same holds for the alpha activity and the heat emission. This statement has been confirmed by a calculational case study on an americium containing sample, which was supposed to be irradiated for about three years in an experimental facility in the Petten High Flux Reactor.

Introduction

Partitioning and Transmutation (P&T) of actinides are generally said to have three main objectives:

- 1 reduction of the possible dose to the population due to leakage of actinides from underground disposal sites;
- 2 reduction of the total radiotoxic inventory of underground disposal sites;
- 3 using fissile actinides for energy production.

Each of these three objectives has its specific impact for the P&T strategy to be followed. The first objective implies that the initial inventory of Np-237 and its precursors shall be reduced, because Np-237 is a nuclide which is easily transported to the biosphere without much adsorption in the underground once it is released from the underground disposal site. Studies on underground disposal options in salt domes show that Np-237 dominates the possible dose to the population living some millions of years from now [1].

The second objective is met by heavily reducing the total amount of actinides in High Level Waste (HLW). It is believed that reducing the radiotoxic inventory per disposal site or reducing the number of disposal sites has some positive impact on the public opinion with regard to the nuclear waste problem. It also leads to lower probabilities of human intrusion in repositories and to reduced accompanied doses received by men after intrusion. For human intrusion scenarios the contribution of americium to the dose is dominant [1]. Also from the economic point of view it is advantageous to reduce the inventory or the number of underground disposal sites to be licensed. This can be accomplished by reduction of the heat emission of the waste.

The third objective is met by fissioning of actinides in nuclear energy parks with thermal and fast reactors. This was the main reason to apply reprocessing. Future incentives for partitioning of actinides, especially of plutonium, depend primarily on future prospects of nuclear energy, which are highly affected by political standpoints.

This paper attempts to give incentives for heterogenous transmutation of americium in thermal reactors based on the first two given objectives. It deals explicitly with transmutation of americium in thermal reactors. Although fast reactors may be more appropriate for transmutation of americium, it is expected that only few fast reactors will be available in the next decades of years. Unless otherwise stated, calculations have been done with the fuel depletion code ORIGEN-S [2] with accompanied data libraries based on JEF2.2 and EAF3 [3]. Radiotoxicities are based on Annual Limits on Intake (ALI) values for ingestion based on ICRP-61 [4]. References 5 and 6 give more details about the material presented in sections 2 to 5, and 6, respectively.

2 Production of americium

Table 2.1 gives the yearly produced masses of neptunium, plutonium, americium and curium in the Borssele Nuclear Power Plant (The Netherlands), which has an electric power of 450 MW. It is seen that americium is produced in only small quantities during reactor operation. However, it is produced in much larger quantities in spent fuel due to decay of Pu-241 with a half life of 14.7 years. The yearly produced quantities of americium as a function of the interim storage time after unloading are given in Table 2.2. It is seen that the yearly produced Am-241 mass increases from 0.7 kg at EOC to 8.2 kg after 10 years of interim storage due to decay of Pu-241. From these numbers it follows that plutonium once partitioned from spent fuel should not be stored for a long time, because a large fraction of Pu-241 will have decayed to Am-241, which decreases the fraction of fissile isotopes in the plutonium.

If plutonium is not used in MOX fuel, all Pu-241 will eventually decay to Am-241, leading to a yearly produced Am-241 mass of 20.6 kg. Because all Am-241 will eventually decay to Np-237, fissioning of Pu-241 in MOX fuel seems the most effective way to prevent the buildup of Np-237. Because partitioning of plutonium is done only some years after interim storage of spent fuel, the amount of Am-241 produced during interim storage equals about the yearly produced amount of Np-237 (4.9 kg). To meet the first objective (reduction of the possible dose to the population due to leakage of actinides from underground disposal sites) transmutation of Am-241 seems as important as transmutation of Np-237 itself. Unless otherwise stated, all results in the remainder of this paper refer to an initial amount of 4.9 kg Am-241 and 0.9 kg Am-243, corresponding with an interim storage time of five years. It must be kept in mind, however, that the yearly produced Am-241 mass changes when another interim storage time is used.

3 Radiotoxicity of americium

Figure 3.1 gives the radiotoxicity of the yearly produced spent fuel (12.5 t) in the Borssele NPP compared with the radiotoxicity of the total uranium ore needed for manufacturing this fuel (82.2 t). The radiotoxicity after several hundreds of years is completely determined by the actinides in the spent fuel. The contribution of each

actinide element and its daughter products to the radiotoxicity of actinides is shown in Figure 3.2. It is seen that plutonium is the main contributor (80 to 95%) up to one million years of storage. Americium contributes for about 20% up to 1,000 years, and the initial amounts of Am-241 and Np-237 equally contribute after $2 \cdot 10^5$ years of storage.

When partitioning of plutonium is already adopted, as is the case for many countries in Europe, and when the radiotoxicity of plutonium can be considerably reduced by transmutation, americium is the most important nuclide dominating the radiotoxicity of the HLW up to 50,000 years.

Table 2.1: *Yearly produced masses of neptunium, plutonium, americium and curium in the Borssele NPP.*

Nuclide	Half Life (a)	Mass (kg)
Np-237	$2.14 \cdot 10^6$	4.9
Pu-238	$8.78 \cdot 10^1$	1.6
Pu-239	$2.41 \cdot 10^4$	79.5
Pu-240	$6.54 \cdot 10^3$	28.2
Pu-241	$1.47 \cdot 10^1$	19.9
Pu-242	$3.76 \cdot 10^5$	5.5
Am-241	$4.32 \cdot 10^2$	0.7
Am-243	$7.39 \cdot 10^3$	0.9
Cm-242	$4.46 \cdot 10^{-1}$	0.2
Cm-244	$1.81 \cdot 10^1$	0.2

Table 2.2: *Yearly produced americium masses (kg) in the Borssele NPP as a function of the interim storage time after unloading.*

Nuclide	Storage Time (a)		
	2	5	10
Am-241	2.5	4.9	8.2
Am-243	0.9	0.9	0.9
Total	3.4	5.8	9.1

4 Transmutation of americium

Introduction

The most abundant americium isotope in spent fuel after a few years of interim storage is Am-241. Because it is not fissile, it is transmuted by neutron capture to Am-242m (10%) or to Am-242 (90%). The first-mentioned activation product has a relatively long half life of 141 years, is highly fissile, and can therefore easily be fissioned in a thermal neutron flux. The second activation product also has a high fission cross section, but decays with a half life of 16 hours to Cm-242 (83%) or Pu-242 (17%). For the Am-242 fission rate being equal to the decay rate, a thermal neutron flux as high as $7 \cdot 10^{15} \text{ cm}^{-2}\text{s}^{-1}$ is required.

The transmutation product Cm-242 decays to Pu-238 with a half life of 163 days, but can also be transmuted by neutron capture to the fissile isotope Cm-243. A thermal neutron flux as high as $5 \cdot 10^{15} \text{ cm}^{-2}\text{s}^{-1}$ would be needed

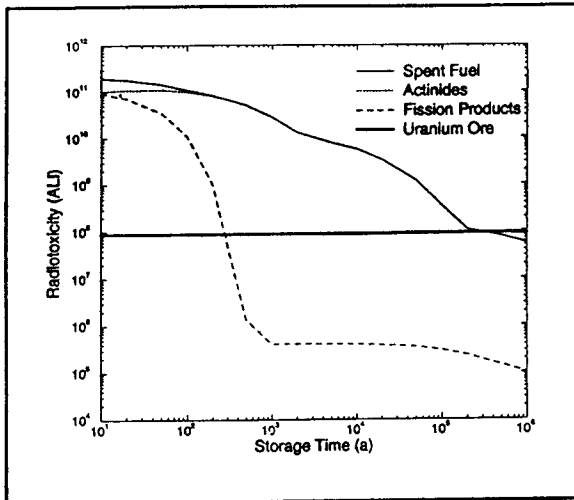


Fig 3.1: Yearly produced radiotoxicity of the Borssele NPP as function of storage time.

there will be successive transformation of the formed Pu-238 and Pu-242. The produced Pu-238 can be further transmuted to fissile Pu-239 and even Pu-241. However, as explained above, the formed Pu-242 is mainly transmuted to Am-243, and this amount of Am-243 (max 15% of the initially present Am-241) together with the initially present Am-243 (about 0.9 kg) is again transformed to Cm-244 upon neutron capture. Cm-244 is a short-lived neutron emitting isotope and another neutron capture would be necessary to transmute this isotope into fissile Cm-245. The whole process is illustrated in Figure 4.1, where the fission rate as a function of time is shown for 4.9 kg Am-241 and 0.9 kg Am-243 being irradiated in a thermal neutron flux of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$.

Transmutation of americium in thermal reactors leads to a mix of nuclides with a high α activity and a high spontaneous fission rate, which is practically impossible to reprocess, and which can not be used for manufacturing of new targets or fuel assemblies. Successive reprocessing is avoided by adapting a transmutation scheme in which the irradiated americium is disposed of.

First objective

To meet the first objective mentioned in section 1 (reduction of the possible dose to the population due to leakage of actinides from underground disposal sites) the amount of Np-237 and its precursors (Am-241, Pu-241 and Cm-245) should be reduced as much as possible. Table 4.1 gives the sum of radiotoxicities due to Np-237 and its precursors originating from americium after irradiation with a thermal neutron fluence of about $1.9 \cdot 10^{22} \text{ cm}^{-2}$ (corresponding with an irradiation of 6 years in a thermal neutron flux of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$), and after a storage time of one million years. Almost all initial Am-241 is transmuted, and the radiotoxicity due to Np-237 and its precursors in the irradiated americium after a storage time of one million years is reduced with a factor of 100. It must be noted however that Am-241 in spent fuel contributes for 50% to the radiotoxicity due to Np-237 after one million years, and that transmutation of Np-237 itself, also with yearly produced mass in the Borssele NPP of 4.9 kg (see Table 2.1), would be necessary to reduce the other contribution of 50%. This latter percentage

for the capture rate being equal to the decay rate. The formed amount of Pu-242 can be transmuted into the fissile Pu-243. However, this plutonium isotope decays with a half life of 5 hours to Am-243. In case of irradiation of Am-241 in low thermal neutron fluxes and moderate values of the neutron fluence, transmutation will result in production of Pu-238 (75%) and Pu-242 (15%), and fissioning of Am-242m (10%).

Transmutation of Am-241 in thermal reactors seems only attractive in a high thermal neutron flux of about $10^{16} \text{ cm}^{-2}\text{s}^{-1}$. The argument that transformation of Am-241 into short-lived Pu-238 is beneficial because it would reduce the production of 'mobile' Np-237 (the first objective mentioned in section 1) violates the second objective since the radiotoxicity or heat emission is not reduced. However, there could still be some incentive to transmute Am-241 in a thermal reactor when a very high neutron fluence (flux times irradiation time) is achieved. Then

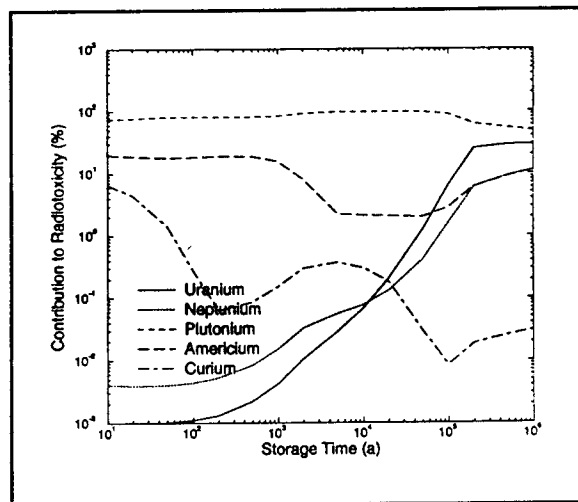


Fig 3.2: Relative contribution of each actinide element and its daughters products to the radiotoxicity of actinides in spent fuel.

decreases when longer interim storage times are applied for the spent fuel (see Table 2.2).

Second objective

The second objective (reduction of the total radiotoxic inventory of underground disposal sites) can be met by reducing the radiotoxicity and the α activity of the waste. In Figure 4.2, the radiotoxicity due to americium is shown before and after irradiation in a neutron flux with a thermal neutron fluence of $9.5 \cdot 10^{21} \text{ cm}^{-2}$ (corresponding with an irradiation of 3 years in a thermal neutron flux of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$). After about 70 years, the radiotoxicity due to the irradiated americium is lower than the radiotoxicity due to the unirradiated americium. It is also seen that the radiotoxicity due to the irradiated americium is lower for lower thermal neutron fluxes. This is mainly due to the fact that Cm-242 decays with half life of 163 days and that lower neutron fluxes lead to longer irradiation times when the neutron fluence is kept constant. More Cm-242 can be transmuted to Pu-239 (via α decay and subsequent neutron capture in Pu-238), and subsequently be fissioned when the irradiation time is longer.

Figure 4.3 gives the similar curves for a thermal neutron fluence which is twice as large; thus corresponding with an irradiation of 6 years in a thermal neutron flux of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$. The radiotoxicity due to the irradiated americium is already within 10 years lower than the radiotoxicity due to the unirradiated americium. For irradiation in thermal neutron fluxes of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$ and lower, the radiotoxicity due to the irradiated americium after 100 years of storage is about a factor of 20 lower than the radiotoxicity due to the unirradiated americium. It must be noted that Figures 4.2 and 4.3 give the radiotoxicity as function of storage time after irradiation. This means specifically that the irradiation time corresponding with each thermal flux is neglected.

Table 4.1: Total radiotoxicity and radiotoxicity due to Np-237 and its precursors after irradiation of 5.8 kg americium with a thermal neutron fluence of $1.9 \cdot 10^{22} \text{ cm}^{-2}$ and after a storage time of one million years.

Thermal Flux ($\text{cm}^{-2}\text{s}^{-1}$)	Radiotoxicity (ALI)	
	Total	Np-237+Precursors
0	$6.1 \cdot 10^6$	$6.1 \cdot 10^6$
$4 \cdot 10^{14}$	$7.0 \cdot 10^5$	$7.5 \cdot 10^4$
$2 \cdot 10^{14}$	$3.8 \cdot 10^5$	$7.0 \cdot 10^4$
$1 \cdot 10^{14}$	$2.7 \cdot 10^5$	$6.4 \cdot 10^4$
$4 \cdot 10^{13}$	$2.5 \cdot 10^5$	$6.3 \cdot 10^4$
$2 \cdot 10^{13}$	$2.6 \cdot 10^5$	$6.4 \cdot 10^4$

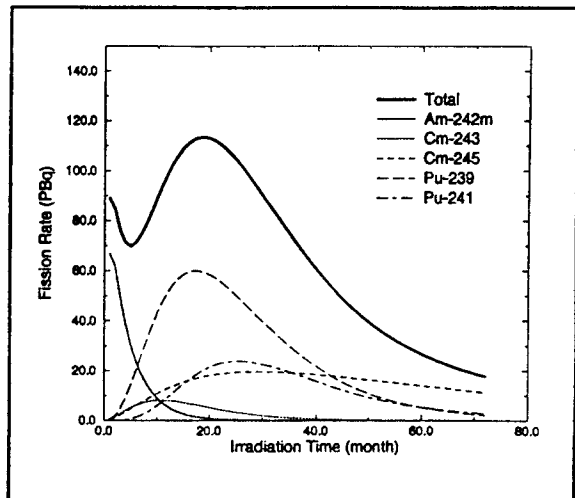


Fig 4.1: Fission rate as function of time of 5.8 kg americium irradiated in a thermal neutron flux of $10^{14} \text{ cm}^{-2}\text{s}^{-1}$.

Apart from other reasons, the second objective (reduction of the total radiotoxic inventory of underground disposal sites) may have some economic benefit when the number or the size of underground disposal sites can be reduced. Because the maximum amount of HLW to be stored at one disposal site is mainly determined by the total heat emission of the waste, and because the heat emission of actinides is mainly determined by α activity, it is assumed that the α activity at the time of disposal determines the maximum amount of actinide material

which can be stored at one specific disposal site. The α activity of the americium decay chain as a function of time is shown in Figure 4.4. It is seen that a considerable reduction of the α activity after about 100 years of storage can be achieved by irradiation with a thermal neutron fluence of $1.9 \cdot 10^{22} \text{ cm}^{-2}$.

Table 4.2: *The β^- activities of the yearly produced fission products of the Borssele NPP after a storage time of 50, 100 and 200 years.*

Time (a)	β^- activity (PBq)	Contribution (%)		
		Sr-90	Y-90	Cs-137
50	37	29.1	29.1	40.7
100	11	28.8	28.8	41.6
200	1.1	27.0	27.8	42.0

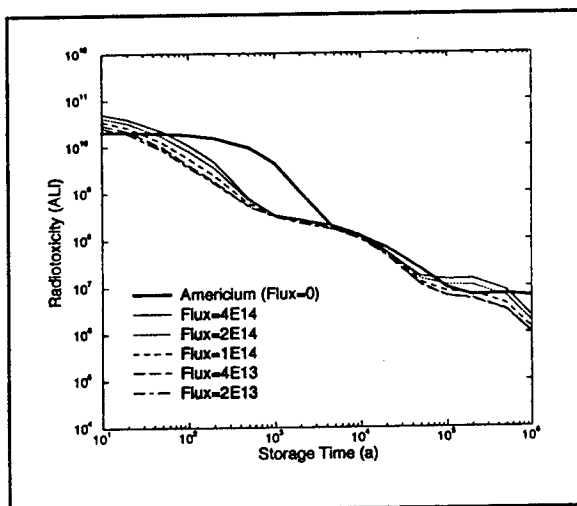


Fig 4.2: *Radiotoxicity due to americium as a function of storage time after irradiation with a thermal neutron fluence of $9.5 \cdot 10^{21} \text{ cm}^{-2}$.*

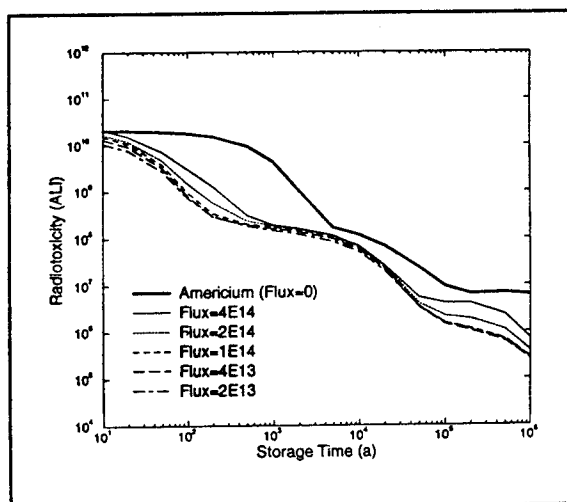


Fig 4.3: *Radiotoxicity due to americium as a function of storage time after irradiation with a thermal neutron fluence of $1.9 \cdot 10^{22} \text{ cm}^{-2}$.*

The heat emission of the actinides has to be compared with the total heat emission of the deposited waste, which is mainly determined by the β^- activity of the fission products. This activity is given in Table 4.2 for storage times of 50, 100 and 200 years. It is seen that the main contributors to the β^- activity are Sr-90, Y-90 and Cs-137. Because the proportional contributions of these isotopes to the β^- activity are almost independent of time, the average emitted energy per β^- decay for this mix of isotopes is constant and equals about 0.4 MeV. In Table 4.3, the average energy emission by β^- and γ decay of the yearly produced fission products of the Borssele NPP are given and compared with the energy emission by α decay of americium, for which it is assumed that the average emitted energy per α decay is 5.5 MeV. After a storage time of 100 years, americium contributes for about 30% to the heat emission. This percentage increases up to 80% after a storage time of 200 years. Therefore, transmutation of americium in order to reduce the heat emission of the waste at the time of final disposal seems only meaningful when an interim storage time of 100 years or longer is applied to fission products during which they are put to temporary storage (e.g. above ground). This interim storage time is about the same as the storage time required to get a considerable reduction of the α activity after transmutation of americium (see Figure 4.4). The α activity at the time of final disposal can be reduced with about 90% and the total heat emission with about 25% if an interim storage time of 100 years is applied, during which both fission products and transmutation

products of americium are put to temporary storage.

5 Inert Matrices

The radiotoxicity due to americium is only reduced considerably when a large percentage (about 75%) of the initial amount of americium has been transmuted into fissile isotopes and eventually fissioned. Such high burnups cannot be reached when americium is irradiated as pure Am_2O_3 with density of 10 g cm^{-3} , because damage to the Am_2O_3 matrix due to recoil and buildup of fission products will limit the burnup to much lower values. Therefore, americium should be irradiated at low concentrations, which can be achieved by diluting americium in some other matrix. Uranium oxide cannot be used as a matrix because the buildup of fissile plutonium isotopes and subsequent fissioning of these isotopes will considerably limit the burnup of americium. Multiple recycling of americium and transmutation products is necessary then. This is a strong incentive to consider transmutation of americium at low concentrations in some neutron inert matrix (e.g. Al_2O_3). Diluting americium in inert matrices can also be used to control the power density in the target containing the americium. Furthermore the relative transmutation efficiency increases considerably when americium is diluted. Of course, for the extended irradiation times assumed in this paper, there are also huge requirements on the cladding, which preferably is made of stainless steel.

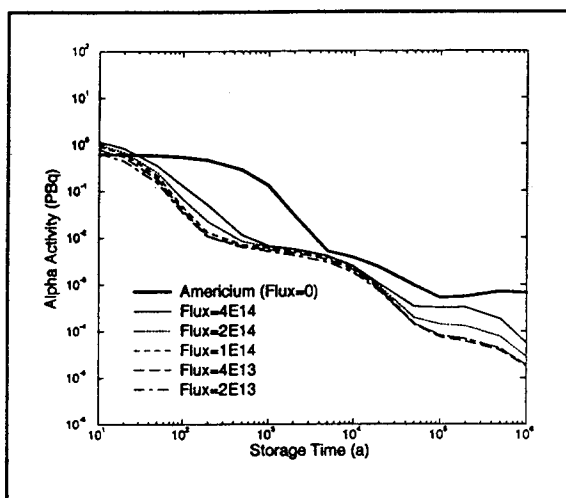


Fig 4.4: The α activity due to americium as function of storage time after irradiation with a thermal neutron fluence of $1.9 \cdot 10^{22} \text{ cm}^{-2}$.

Table 4.3: Energy emission of the yearly produced fission products (only $\beta^- + \gamma$ decay) and americium (only α decay) of the Borssele NPP after storage times of 50, 100 and 200 years.

Time (a)	Energy Emission (MeV s^{-1})				
	β^-	γ	$\beta^- + \gamma$	α	$\alpha + \beta^- + \gamma$
50	$1.5 \cdot 10^{16}$	$8.6 \cdot 10^{15}$	$2.4 \cdot 10^{16}$	$3.2 \cdot 10^{15}$	$2.7 \cdot 10^{16}$
100	$4.4 \cdot 10^{15}$	$2.7 \cdot 10^{15}$	$7.1 \cdot 10^{15}$	$3.0 \cdot 10^{15}$	$1.0 \cdot 10^{16}$
200	$4.4 \cdot 10^{14}$	$2.7 \cdot 10^{14}$	$7.1 \cdot 10^{14}$	$2.5 \cdot 10^{15}$	$3.2 \cdot 10^{15}$

6 Calculational Case Study for Irradiation in the Petten High Flux Reactor

Model

In the framework of the EFTTRA cooperation [11], calculations have been done for a specific experimental facility containing four americium samples [6]. The experimental facility consists of an aluminium ring with provisions for nine samples and with a gas hole in the centre. The facility is supposed to be irradiated in the Petten high flux reactor for about three years. The sample with radius of 2.5 mm is supposed to be made of Am_2O_3 diluted in an inert matrix of Al_2O_3 encapsulated by a titanium steel cladding. The americium composition as present after an interim storage time of five years is used (see Table 2.2). The initial density of Am_2O_3 in the sample is 0.4255 g cm^{-3} .

The results given in this paper have been obtained with the SCALE code system [7] in combination with ROLAIDS-CPM [8] to treat radial burnup effects [9]. The one-dimensional model of the experimental facility has been verified by one- and two-dimensional WIMS-6 [10] calculations. The results of the one-dimensional calculations with both the SCALE and the WIMS-6 code systems, agree very well with the two-dimensional WIMS-6 calculations [6]. In both models the americium sample is divided in five radial zones with equal volumes

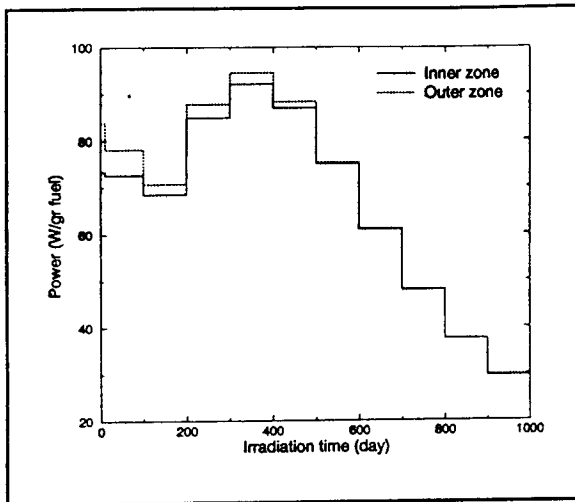


Fig 6.1: Power density in the inner and outer fuel zones as function of irradiation time.

to study radial burnup effects. All data used are based on JEF2.2 in 172 groups.

Results

The differences in burnup between the five zones of the samples are very small, because the differences in power history between the five zones are very small. This is seen in Figure 6.1, where the fission power in the americium sample for the inner and outer zones are given as a function of irradiation time. Because the fission power distribution over the sample is rather flat, it is expected that the radiation damage effects in the matrix due to fission product recoil and buildup will also be homogeneously distributed over the sample. The maximum power is reached after about one year of irradiation due to buildup of fissile Pu-239. The initial power is caused by neutron capture in Am-241 and fissioning of Am-242m. After irradiation of 1,000 days the actinide density in the sample has decreased from

0.3868 g cm⁻³ to 0.1096 g cm⁻³, of which 0.0615 g cm⁻³ is Cm-244.

The radiotoxicity of the samples is given in Figure 6.2. It is seen that the radiotoxicity is considerably reduced compared to the radiotoxicity of the unirradiated samples. The remaining radiotoxicity is due to Cm-244 mainly, which is formed by neutron capture in Am-243, and which decays with half life of 18 years to Pu-240. The transmutation efficiency is therefore expected to decrease when the initial Am-243 content in the americium sample increases.

7 Conclusions

The radiotoxicity due to spent fuel is mainly determined by plutonium. After a time period of 100,000 years, also uranium gives an important contribution to the radiotoxicity. However, if uranium and plutonium are partitioned from spent fuel in order to be recycled again, and when the radiotoxicity of plutonium is considerably reduced, then the radiotoxicity due to HLW up to 50,000 years is mainly determined by americium.

Partitioning and transmutation of americium has two possible benefits: reduction of the possible dose to the population due to leakage of Np-237 from underground disposal sites, and reduction of the total inventory (radiotoxicity) of underground disposal sites (this can be either reduction of the inventory per disposal site or reduction of the total number of disposal sites).

The initial amount of Am-241 in spent fuel contributes for about 50% to the total radiotoxicity of the daughter product Np-237 after 2 10⁵ years of storage. This percentage increases when the interim storage time of spent fuel increases. Because all Am-241 can be fissioned or transmuted to other nuclides, transmutation of americium leads to a 50% reduction of the total radiotoxicity due to Np-237, which leads to an equally large reduction of the possible dose to the population due to leakage of Np-237. It is clear however that also Np-237 should be transmuted if the radiotoxicity due to Np-237 after one million years of storage is to be reduced considerably.

The long-term radiotoxicity due to americium can be reduced with a factor of 20 after about one hundred years of storage, when it has been irradiated with a thermal neutron fluence of about 2 10²² cm⁻². Irradiation of americium in a thermal neutron flux of 10¹⁴ cm⁻²s⁻¹ (a characteristic value for the Petten High Flux Reactor), leads

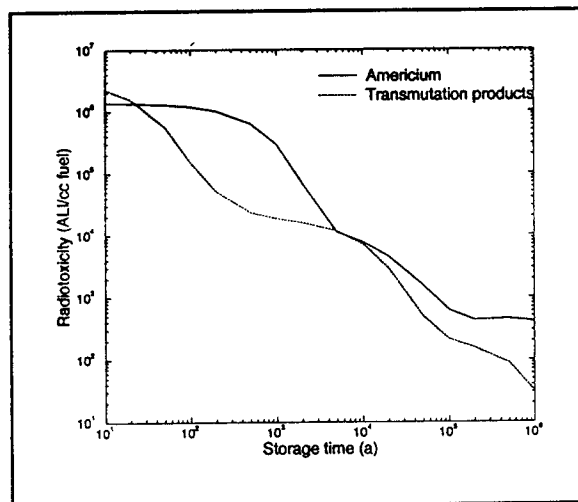


Fig 6.2: Radiotoxicity due to americium as function of storage time after irradiation in the Petten High Flux Reactor for 1000 days.

to the above-mentioned reduction factor for the radiotoxicity. This was confirmed by calculations on an experimental facility containing an americium sample, which was supposed to be irradiated in the Petten HFR for about three years. Generally it can be concluded that reduction of the radiotoxicity due to americium in thermal reactors is feasible when long irradiation times are applied. This would only be meaningful, however, when the radiotoxicity due to plutonium would also be reduced considerably.

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