TRANSMUTATION OF AMERICIUM IN FISSION REACTORS

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ABSTRACT

To get a considerable reduction of the radiotoxicity due to americium, a thermal neutron fluence of $2 \cdot 10^{22}$ cm^{-2} or a fast neutron fluence of $2 \cdot 10^{24}$ cm^{-2} is required. Irradiation in a thermal neutron flux leads to lower masses of ^{234}U and precursors and of ^{237}Np and precursors, but to higher curium masses and much higher neutron emission rates than irradiation in a fast neutron flux. Therefore, irradiation in a fast neutron flux has preference when multiple recycling is adopted. When once-through burning is applied, irradiation in a thermal neutron flux can be applied. Then irradiation in a HWR has preference above irradiation in a PWR or in a HTGR.

INTRODUCTION

Americium is produced in nuclear reactors in rather small quantities: 1.6 kg of ^{241}Am and 2.0 kg of ^{243}Am per GW$_e$a produced. However, ^{241}Am is produced in much larger quantities in the nuclear fuel cycle due to decay of ^{241}Pu with a half life of 15 years. When spent fuel is stored for 5 years prior to reprocessing, it contains already 10.9 kg of ^{241}Am per GW$_e$a produced. In the remainder of this paper, the isotopic ^{241}Am:^{243}Am composition is assumed to be 10.9:2, corresponding with an interim storage time of 5 years.

This americium contributes to the radiotoxicity of actinides in spent fuel for about 20% between 200 and 1000 years of storage. This is shown in figure 1. When the plutonium is partitioned from the spent fuel, the contribution of americium even exceeds 90%. Because the ^{241}Am mass in spent fuel equals that of ^{237}Np ($\approx$ 11 kg per GW$_e$a produced) and all ^{241}Am eventually decays to ^{237}Np, the ^{241}Am and the ^{237}Np equally contribute to the radiotoxicity after 200,000 years of storage. From these data, two incentives to transmute americium can be derived [1,2]:

1. All ^{241}Am eventually decays to ^{237}Np, which is easily transported to the biosphere without much absorption in the underground once released from the repository.

2. The ^{241}Am contributes to the radiotoxicity of the spent fuel for about 20% between 200 and 1000 years of storage. This contribution increases even up to 90% when plutonium is partitioned from the spent fuel.

Of course, the question remains whether these reasons are sufficient to support transmutation of americium. The first incentive implies a reduction of the risk in normal evolution scenarios, the second one a reduction of the risk in human intrusion scenarios. In both cases, however, a certain risk to future generations is reduced at the expense of an increase of the risk to the present generation. This means that at the extreme, the present generation which benefits from nuclear energy, will also have the burden of it. This enables one to decide clearly whether the benefits of nuclear energy outweighs the drawbacks.

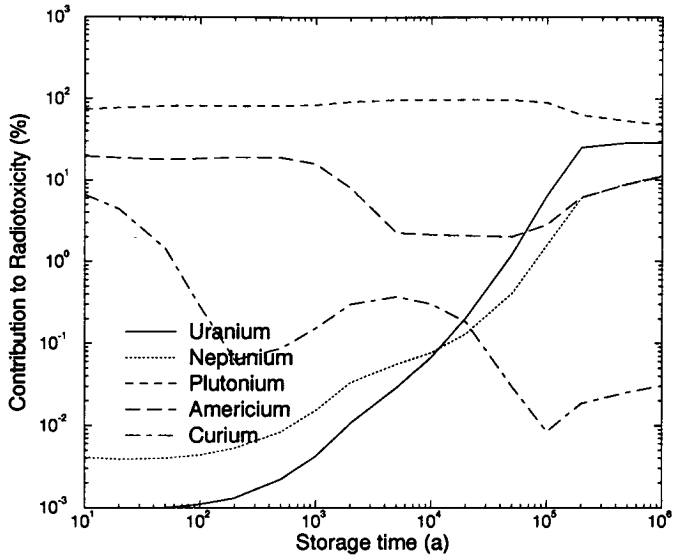

Figure 1: The contribution of each actinide element and its daughter products to the radiotoxicity of actinides in spent fuel as a function of the storage time. The contribution of fission products is neglected.

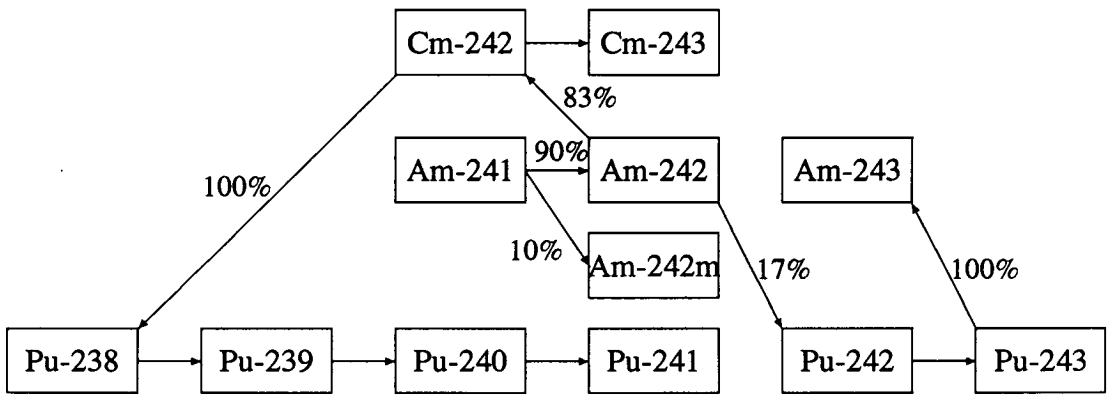

Figure 2: Main transmutation route of ^{241}Am in a thermal neutron flux.

IRRADIATION IN A THERMAL OR FAST NEUTRON FLUX?

As mentioned above, the most abundant americium isotope in spent fuel is ^{241}Am, and its abundancy increases when the storage time between unloading the spent fuel and reprocessing increases.

The main transmutation route of ^{241}Am in a thermal neutron flux is shown in figure 2. Because ^{241}Am is not fissile, it is transmuted by neutron capture to ^{242m}Am (10%) and ^{242}Am (90%). The first-mentioned transmutation product is highly fissile, and can therefore easily be fissioned. The second product is also fissile, but decays with a half life of 16 hours to ^{242}Cm (83%) and

^{242}Pu (17%). The curium isotope decays to ^{238}Pu with a half life of 163 days, and this can be transmuted to the fissile ^{239}Pu. The produced ^{242}Pu is further transmuted to ^{243}Pu, which decays quickly to ^{243}Am. This produced ^{243}Am together with the initially present ^{243}Am is transmuted to ^{244}Cm upon a neutron capture and subsequent β^- decay. The ^{244}Cm can be further transmuted to the fissile ^{245}Cm. The transmutation route of ^{241}Am in a fast neutron flux is mainly the same as in a thermal flux, but due to the smaller ratio of capture-to-fission cross sections, a larger fraction of the actinides is fissioned instead of transmuted to other nuclides.

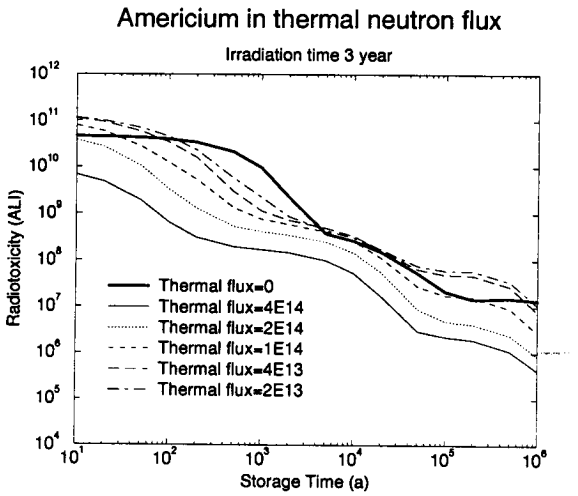

Figure 3: Radiotoxicity of americium irradiated for 3 years in a thermal neutron flux.

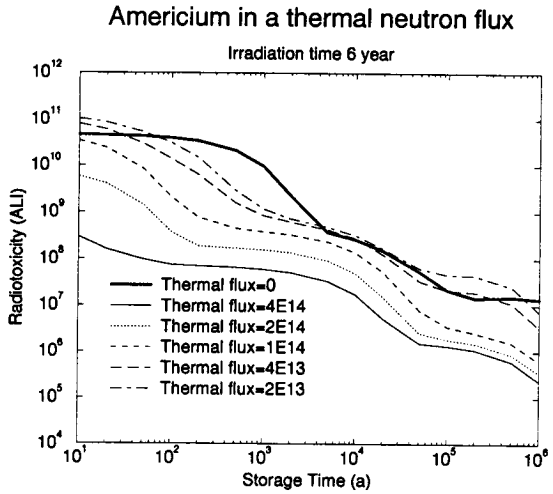

Figure 4: Radiotoxicity of americium irradiated for 6 years in a thermal neutron flux.

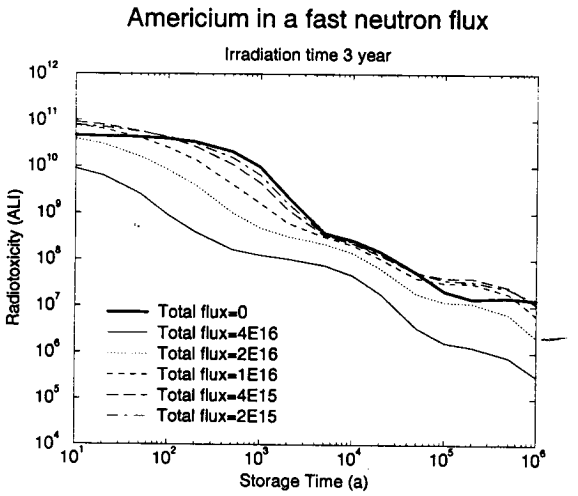

Figure 5: Radiotoxicity of americium irradiated for 6 years in a fast neutron flux.

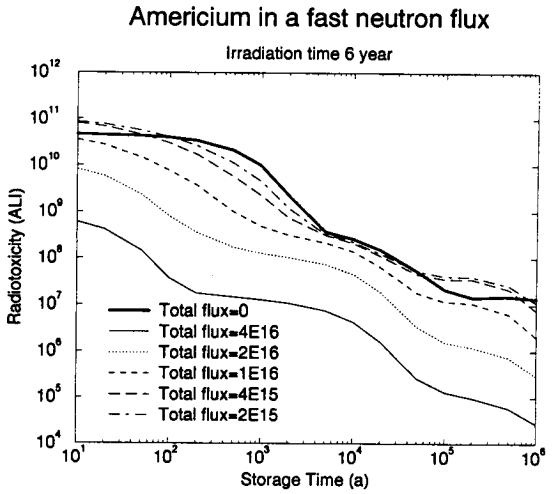

Figure 6: Radiotoxicity of americium irradiated for 6 years in a fast neutron flux.

To decide which neutron spectrum has preference for transmutation of americium, a parametric study with the fuel depletion code ORIGEN-S [3] and accompanied data libraries [4] has been done, and results are shown in figures 3 to 6. Figures 3 and 4 show the radiotoxicity of 12.9 kg of americium (10.9 kg of ^{241}Am and 2 kg of ^{243}Am) after an irradiation time of 3 or 6 years in a thermal neutron flux with a spectrum characteristic for an UO_2 fueled PWR. The flux values in the figures are the *thermal* neutron fluxes ($E < 0.5$ eV), and it is assumed that the total neutron

flux is 6.7 times higher than the thermal neutron flux. In figures 5 and 6, the americium is assumed to be irradiated in a fast neutron flux with a spectrum characteristic for Superphénix . Also the flux values have been increased, because the total neutron flux in a fast reactor is about 100 times larger than the thermal neutron flux in a thermal reactor. For example, the thermal neutron flux in a PWR has a characteristic value of $4 \cdot 10^{13}$ cm^{-2} s^{-1} , while the total neutron flux in Superphénix has a characteristic value of $4 \cdot 10^{15}$ cm^{-2} s^{-1} .
The figures show that the radiotoxicity due to americium can be considerably reduced upon irradiation in a thermal or fast neutron flux. The most important parameters are the flux value and the irradiation time. To achieve a considerable reduction of the radiotoxicity, the values of these parameters should be such that the thermal neutron fluence has a value of at least $2 \cdot 10^{22}$ cm^{-2} (corresponding with an irradiation time of 6 years in a thermal neutron flux of $1 \cdot 10^{14}$ cm^{-2} s^{-1}), and that the fast neutron fluence has a value of at least $2 \cdot 10^{24}$ cm^{-2} (corresponding with an irradiation time of 6 years a fast neutron flux of $1 \cdot 10^{16}$ cm^{-2} s^{-1}). The irradiation in a thermal neutron flux with a fluence of $2 \cdot 10^{22}$ cm^{-2} gives about the same reduction of the radiotoxicity as the irradiation in a fast neutron flux with a fluence of $2 \cdot 10^{24}$ cm^{-2}. From the figures, it is difficult to decide which neutron spectrum has preference for transmutation of americium.
However, there are many other parameters which determine the effectiveness of transmutation of americium. Examples are the total actinide mass, the mass of the mobile ^{237}Np and its precursors, the mass of ^{234}U and its precursors (because the radon produced by decay of these nuclides can give a large dose rate to the population living in the far future), the mass of the higher curium isotopes, the α activity and the neutron emission rate of the irradiated americium. These parameters are given in table I.
When the results of calculations 2 and 8 on the one side, and calculations 12 and 18 on the other are compared with each other (recall that these calculations give about the same reduction of the radiotoxicity), it is seen that the total actinide mass in calculations 2 and 8 is less than in calculations 12 and 18, which means that a higher portion of the initially present americium has been fissioned in calculations 2 and 8. Also the masses of ^{234}U and precursors and of ^{237}Np and precursors are less in calculations 2 and 8 than in calculations 12 and 18. However, irradiation of americium in a fast reactor leads to less curium which is an advantage because this nuclide can hardly be recycled. The α activity of the irradiated americium show no preference for irradiation in a thermal neutron flux or a fast neutron flux, but the neutron emission rate is about 100 higher in calculations 2 and 8 than in calculations 12 and 18. This is probably a strong incentive to transmute americium in a fast neutron flux when recycling of the transmutation products in the irradiated americium is adopted.
Another parameter, which favours transmutation of americium in a fast neutron flux, is the neutron economy. Americium irradiated in a thermal neutron flux absorbs more neutrons than are produced, and extra enrichment is needed to compensate this. When americium is irradiated in a fast neutron flux, the fission power is mainly due to ^{241}Am, ^{242m}Am, ^{238}Pu and ^{239}Pu and this implies that less neutrons are required to fission a nuclide and that the overall neutron balance is positive. In other words, americium irradiated in a fast neutron flux behaves more like a fuel.
In conclusion: to transmute americium such that a considerable reduction of the radiotoxicity is achieved, a high neutron fluence is required. This can be obtained in two ways:

1. Once-through burning, in which americium is irradiated for a long time in a high thermal or fast neutron flux. The irradiated americium is not reprocessed, but either repacked for re-irradiation (if possible), or sent to final storage.

2. Multiple recycling in which americium is irradiated for a limited time followed by re-processing and recycling of the transmutation products. In this case, irradiation in a fast

neutron flux has preference to reduce the curium production and the neutron emission rate of the irradiated americium.

The first option seems a good starting point to buildup experience with irradiation of americium in reactors and to achieve a reduction of the radiotoxicity without the use of fast reactors. This seems a feasible option as long as fast reactors are not yet available. However, when the reprocessing losses are small, the second option is expected to yield a larger reduction of the radiotoxicity due to the reprocessing of the irradiated americium and the recycling of the transmutation products. This option is favourable when new fast reactors will be build. To speed up the transmutation process, one can also consider irradiation of americium in a moderated subassembly in the blanket of a fast reactor.

Table I: The total actinide mass, the mass of ^{234}U and its precursors (^{234}U+P), the mass of ^{237}Np and its precursors (^{237}Np+P), and the mass of ^{243}Cm till ^{249}Cm (^{243}Cm+up) after irradiation of 10.9 kg of ^{241}Am and 2 kg of ^{243}Am. Also the α activity and the neutron emission rate of the irradiated americium after 2 years of decay are given.

Calc number	Flux (cm^{-2} s^{-1})	Actinide mass (g)	^{234}U+P mass (g)	^{237}Np+P mass (g)	^{243}Cm+up mass (g)	α activity (PBq)	n source (TBq)
3 years in a thermal neutron flux							
1	$4{\cdot}10^{14}$	708	34	20	632	0.68	5.9
2	$2{\cdot}10^{14}$	2144	239	127	1483	3.2	0.78
3	$1{\cdot}10^{14}$	5054	1305	328	2064	6.2	0.06
4	$4{\cdot}10^{13}$	9515	4191	633	1644	9.3	0.02
5	$2{\cdot}10^{13}$	11370	5113	1975	1036	10.3	0.01
6 years in a thermal neutron flux							
6	$4{\cdot}10^{14}$	243	0.1	0.4	233	0.09	10.6
7	$2{\cdot}10^{14}$	624	2.5	18	592	0.51	4.8
8	$1{\cdot}10^{14}$	1906	95	116	1410	2.8	0.63
9	$4{\cdot}10^{13}$	5739	1557	398	1815	5.6	0.03
10	$2{\cdot}10^{13}$	9207	3834	685	1502	6.9	0.02
3 years in a fast neutron flux							
11	$4{\cdot}10^{16}$	713	68	72	357	0.74	$9.2{\cdot}10^{-3}$
12	$2{\cdot}10^{16}$	3187	924	343	953	3.4	$9.4{\cdot}10^{-3}$
13	$1{\cdot}10^{16}$	7055	2648	1511	1223	7.1	$1.2{\cdot}10^{-2}$
14	$4{\cdot}10^{15}$	10740	3168	4782	874	7.7	$9.5{\cdot}10^{-3}$
15	$2{\cdot}10^{15}$	11970	2308	7191	524	5.8	$6.2{\cdot}10^{-3}$
6 years in a fast neutron flux							
16	$4{\cdot}10^{16}$	59	0.3	5	41	0.05	$1.9{\cdot}10^{-2}$
17	$2{\cdot}10^{16}$	714	56	74	336	0.65	$7.8{\cdot}10^{-3}$
18	$1{\cdot}10^{16}$	3140	850	346	866	2.7	$8.6{\cdot}10^{-3}$
19	$4{\cdot}10^{15}$	8130	2950	2186	1069	5.8	$1.1{\cdot}10^{-2}$
20	$2{\cdot}10^{15}$	10700	3122	4783	794	5.9	$8.7{\cdot}10^{-3}$

TRANSMUTATION OF AMERICIUM IN FISSION REACTORS

In the remainder of this paper, attention will be focussed to once-through burning of americium in a Heavy Water Reactor (HWR), in a Pressurized Water Reactor (PWR), and in a High Temperature Gas-cooled Reactor (HTGR), and to multiple recycling in a Liquid Metal-cooled Reactor (LMR). For all reactor types, it has been assumed that the americium is diluted in a neutron inert matrix. This is a necessary condition, because the radiotoxicity of the sample is only considerably reduced when a large percentage of the initially present americium is eventually fissioned. This cannot be achieved 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 buildup to much lower values. Therefore, americium should be irradiated at low concentrations, which can be achieved by diluting the americium in some other matrix. UO_2 cannot be used, because the buildup and fissioning of plutonium isotopes will limit the burnup of the americium in the sample. This is a strong incentive to use a neutron and chemical inert matrix like Al_2O_3 . Diluting the americium in a neutron inert matrix can also be used to control the power density in the target and to increase the relative transmutation rate of the americium.

The power density of an americium sample irradiated in a HWR, PWR, HTGR or LMR are shown in figures 7 till 10. Here the americium densities are selected according to two criteria:

1. The maximum power density in the target may not exceed the average power density in the fuel of the reactor considered.

2. The burnup in the target may not exceed the burnup of standard fuel of the reactor considered.

The first constraint was limiting for the HWR and the LMR, which lead to americium densities of $4^w\%$ and $70^w\%$, respectively. (These percentages are the weight fractions of the Am_2O_3 in the mix of Am_2O_3 and Al_2O_3 , where the Al_2O_3 density is 4 g cm^{-3} and the Am_2O_3 density 10 g cm^{-3}). The americium density in the LMR is much higher than in the HWR, because it is assumed that the target irradiated in the LMR will be reprocessed and that the transmutation products will be recycled, while once-through burning is assumed for the HWR. The second constraint was limiting for the PWR and the HTGR. This means that the burnup of the targets when all initial americium will have been fissioned, is comparable to the burnup of ordinary fuel of the reactor type considered. This also implies once-through burning for these two reactor types.

The power density in the americium target reaches a maximum after 600 days for the HWR, after 1300 days for the PWR, after 1000 days for the HTGR and after 1200 days for the LMR. Clearly, the americium is most effectively transmuted in the HWR, because of the low actinide density in the target and the high thermal neutron flux. This flux ranges from $7.3 \cdot 10^{13}$ cm^{-2} s^{-1} at BOL to $6.5 \cdot 10^{14}$ cm^{-2} s^{-1} after 1800 days of irradiation in the HWR, while these values are $3.3 \cdot 10^{13}$ cm^{-2} s^{-1} at BOL and $4.5 \cdot 10^{13}$ cm^{-2} s^{-1} after 1260 days of irradiation in a PWR. Reducing the americium density in the PWR to $4^w\%$ (like in the HWR) gives almost no improvement of the transmutation process. Then the maximum power density is reached after 1200 days of irradiation instead of 1300 days. The thermal neutron flux in the americium sample irradiated in the HTGR is not known, but the *total* neutron flux has a constant value of $1.4 \cdot 10^{14}$ cm^{-2} s^{-1} during the irradiation. This value is rather low, which implies that a very long irradiation time is required to achieve a high neutron fluence.

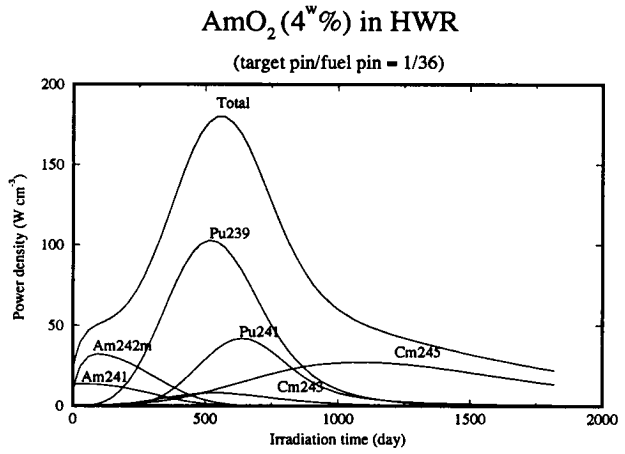

Figure 7: Power density in an americium target irradiated in a HWR.

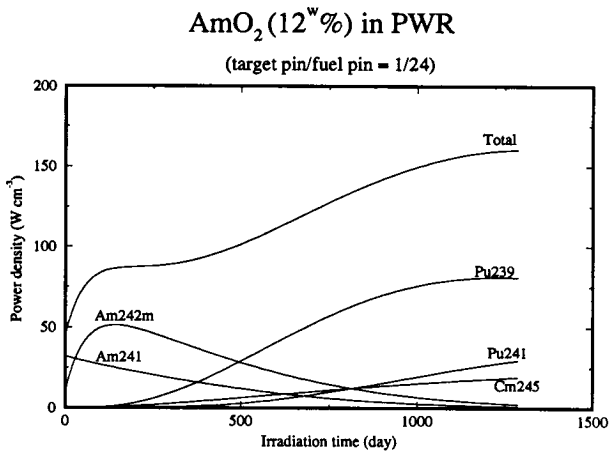

Figure 8: Power density in an americium target irradiated in a PWR.

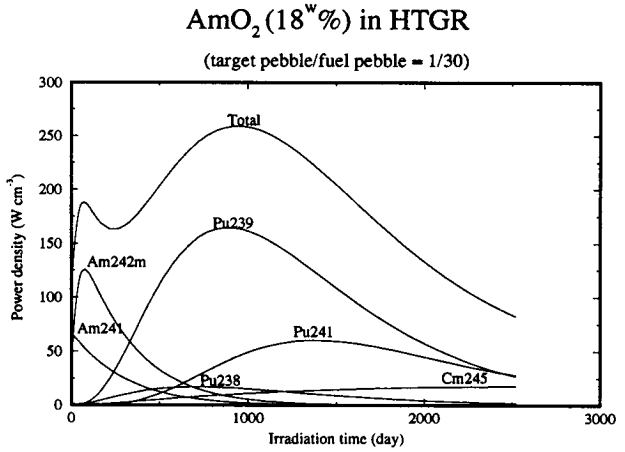

Figure 9: Power density in an americium target irradiated in a HTGR.

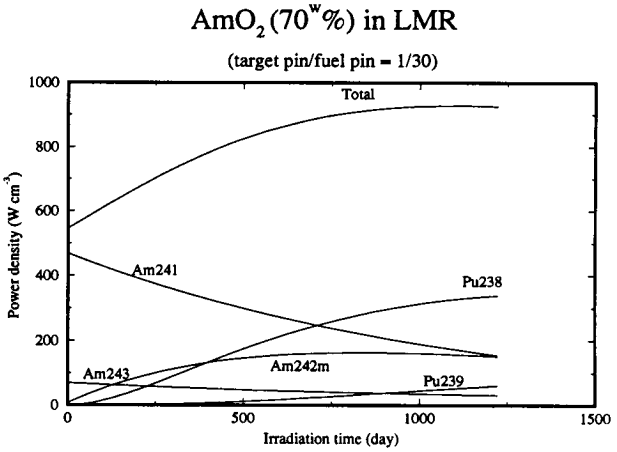

Figure 10: Power density in an americium target irradiated in Superphénix .

Recall that the most important period with respect to the radiotoxicity of americium is between 200 and 1000 years of storage, because the contribution of americium to the radiotoxicity of actinides in spent fuel is largest then (see figure 1). The radiotoxicity of the samples irradiated in the four different reactor types is shown in figure 11. Clearly, the reduction of the radiotoxicity is largest for the HWR (a factor of 10 to 20 between 200 and 1000 years of storage).
After about $1 \cdot 10^5$ years, the product of radiotoxicity and mobility of the nuclides determines the risk, which implies that the mass of ^{237}Np and precursors is most relevant. This mass is reduced with a factor of 150 after an irradiation time of 1000 days in the HWR. This practically means that the dose risk to future generations due to leakage of ^{237}Np is reduced with 50% (recall that the initial ^{237}Np mass in the spent fuel equals that of the ^{241}Am, which implies that transmutation of ^{237}Np itself is equally important to reduce the dose risk due to leakage of ^{237}Np).

CONCLUSIONS

To reduce the radiotoxicity due to americium, a high neutron fluence in a thermal or fast neutron flux is required. Irradiation in a thermal neutron flux with a thermal neutron fluence of $2 \cdot 10^{22}$ cm^{-2} or in a fast neutron flux with a total neutron fluence of $2 \cdot 10^{24}$ cm^{-2} gives about the same reduction of the radiotoxicity. For these values of the fluence, an irradiation time of 6 years

in a thermal neutron flux of $1 \cdot 10^{14}$ cm^{-2} s^{-1} or in a fast neutron flux of $1 \cdot 10^{16}$ cm^{-2} s^{-1} is required. Irradiation in a thermal neutron flux leads to less ^{234}U and precursors and to less ^{237}Np and precursors, but to a higher curium content and to a much higher neutron emission rate than irradiation in a fast neutron flux. This implies that irradiation in a fast neutron flux has preference when multiple recycling is adopted. In case of once-through burning (irradiation of americium without recycling), irradiation of americium in a thermal neutron flux might be a good alternative. This can be achieved by irradiation of americium in a thermal reactor, or in a moderated subassembly in a fast reactor. When the first option is adopted, a HWR seem to perform better than a PWR or a HTGR due to the relative high thermal neutron flux.

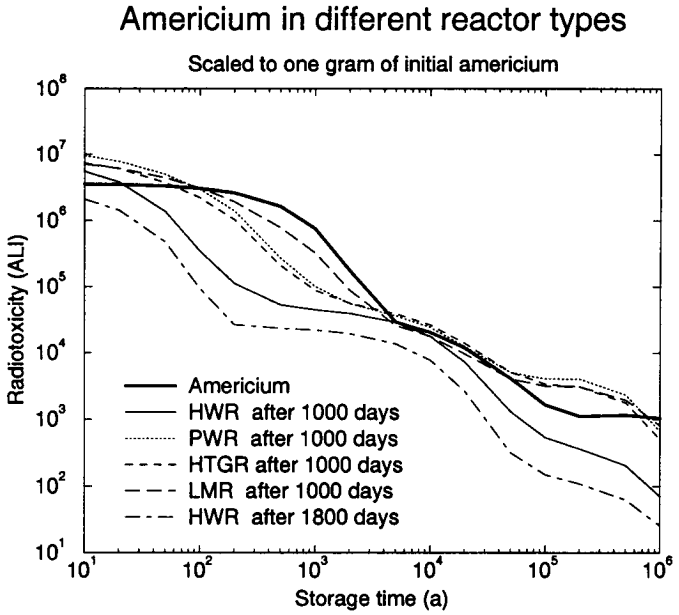

Figure 11: The radiotoxicity of one gram of americium irradiated in four different reactor types as a function of the storage time. The radiotoxicity due to fission products is neglected.

ACKNOWLEDGEMENTS

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REFERENCES

(1) J.L. Kloosterman, *Incentives for Transmutation of Americium in Thermal Reactors*, Netherlands Energy Research Foundation (ECN), Petten, The Netherlands, Report ECN-R–94-022, Oct 1994.

(2) J.L. Kloosterman and W.J.M. de Kruijf, *Transmutation of Americium in Thermal Reactors*, Proc. of 3rd Int. Inf. Exch. Mtng. on Actinide and Fiss. Prod. P&T, Cadarache, France, Dec 1994. Also report ECN-RX–94-116.

(3) O.W. Hermann and R.M. Westfall, *ORIGEN-S, SCALE Module to Calculate Fuel Depletion, Actinide Transmutation, Fission Product Buildup and Decay, and Associated Radiation Source Terms*, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, Feb 1989.

(4) J.L. Kloosterman and J.E. Hoogenboom, *New Data Libraries for Transmutation Studies*, These Proceedings, 1995.