Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Nuclear Materials 320 (2003) 156–162
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Transmutation characteristics in thermal and fast neutron spectra: application to americium

V. Berthou [a,*], C. Degueldre [b], J. Magill [a]

[a] European Commission, Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Germany
[b] PSI, Laboratory for Material Behaviour, 5232 Villigen, Switzerland

Abstract

In this paper, a method is introduced which allows a quick and accurate evaluation of the overall transmutation rate of a nuclide in fast and thermal neutron spectra. The method is applied to ^{241}Am, a main contributor to the waste toxicity in the nuclear fuel cycle. Results show good agreement with the detailed calculations using ORIGEN code.

1. Introduction

^{241}Am is one of the main contributors to the waste toxicity of the spent fuel. Its transmutation into less radiotoxic isotopes is considered in nuclear reactors. In this study both thermal and fast neutron spectra have been compared for the transmutation of ^{241}Am. Two methods have been considered. The overall decrease in actinides mass during the irradiation has been examined using direct ORIGEN calculations. In a second method, the overall burnout time, necessary time to fission half of the initial quantity of ^{241}Am, has been calculated, and compared to the first method.

2. Reaction path of ^{241}Am under neutron irradiation

The neutron path of ^{241}Am under thermal neutron irradiation is shown in Fig. 1 [1].

In both thermal and fast neutron spectra the main reaction is (n,γ) radiative capture (see cross-sections in Table 1). In a fast neutron spectrum, there is more competition between capture and fission since the capture to fission ratio is 78 in thermal spectrum but only 3 in fast spectrum (for ^{241}Am).

During neutron irradiation, there are three basic reactions which lead to the disappearance of the actinides: capture, fission and decay.

From Fig. 1 it can be seen that ^{241}Am creates ^{242m}Am (10% branching ratio) and ^{242}Am (90% branching ratio) by thermal neutron capture. Under fast neutron capture, the branching ratios are 15% for ^{242m}Am and 85% for ^{242}Am. In both spectra, ^{242m}Am transmutes mainly by fission. ^{242}Am, with a half-life of 16 h, has two decay modes: β^- emission that leads to ^{242}Cm (83% branching ratio) and electron capture that leads to ^{242}Pu (17% branching ratio). ^{242}Cm decays, with a half-life of 160 days to ^{238}Pu by α emission. Through radiative capture, ^{238}Pu gives rise to ^{239}Pu. ^{239}Pu is removed predominantly by fission. Through radiative capture, the second daughter product of ^{242}Am decay, ^{242}Pu, gives rise to ^{243}Pu which then decays, with a half-life of 5 h, to ^{243}Am. Through decay and capture, ^{243}Am gives rise to ^{244}Am, ^{244}Cm and ^{245}Cm. ^{245}Cm is removed predominantly by fission.

Table 1 gives a summary of the main actinides in spent fuel together with their half-lives, their fission, capture and absorption cross-sections in both thermal and fast reactors. These cross-sections are taken from the ORIGEN [2] libraries: PWRUS50.lib for the thermal spectrum and AMOPUUUC.lib for the fast spectrum.

* Corresponding author.
E-mail address: berthou@itu.fzk.de (V. Berthou).

0022-3115/03/$ - see front matter © 2003 Elsevier Science B.V. All rights reserved.
doi:10.1016/S0022-3115(03)00183-1

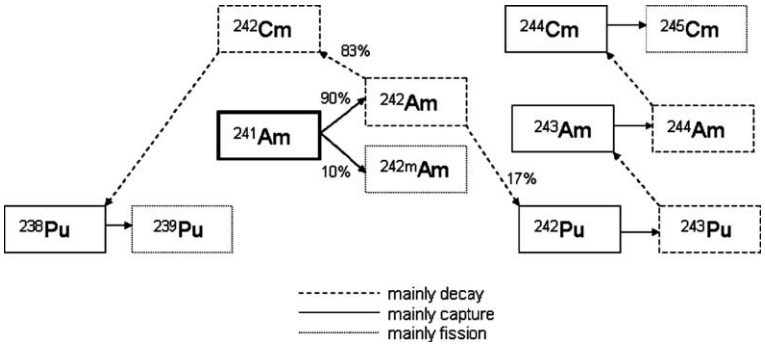

Fig. 1. Reaction path of ^{241}Am under thermal neutron irradiation.

Table 1
Actinides half-lives and fission, capture, and absorption cross sections for thermal and fast neutron reactors (ORIGEN libraries)

	Half-life (years)	Thermal			Fast		
		σ_c (b)	σ_f (b)	σ_a (b)	σ_c (b)	σ_f (b)	σ_a (b)
^{235}U	Stable	9.51	41.00	50.51	0.53	1.87	2.40
^{238}U	Stable	0.92	0.10	1.02	0.28	0.05	0.33
^{237}Np	$2 \times 14\ 10^6$	30.97	0.52	31.49	1.43	0.39	1.82
^{238}Pu	87.7	29.78	2.20	31.98	0.69	1.19	1.88
^{239}Pu	2.4×10^4	57.75	101.50	159.25	0.47	1.82	2.29
^{240}Pu	6.6×10^3	143.30	0.59	143.89	0.48	0.42	0.91
^{241}Pu	14.3	35.37	107.00	142.37	0.44	2.44	2.88
^{242}Pu	3.7×10^5	31.55	0.43	31.98	0.40	0.31	0.71
^{241}Am	432	83.25	1.06	84.31	1.32	0.35	1.67
^{242m}Am	141	83.24	397.70	480.94	0.36	3.90	4.26
^{243}Am	7.37×10^3	49.68	0.41	50.09	1.01	0.27	1.28
^{242}Cm	162.8 days	5.67	0.53	6.20	0.31	0.20	0.51
^{243}Cm	29.1	8.08	68.16	76.24	0.23	2.60	2.83
^{244}Cm	18.1	13.75	0.89	14.64	0.79	0.49	1.28
^{245}Cm	8.5×10^3	24.89	147.00	171.89	0.30	2.59	2.89
^{246}Cm	4.73×10^3	2.92	0.59	3.51	0.22	0.32	0.54

3. Irradiation of ^{241}Am ORIGEN calculations

3.1. Total actinide mass evolution

In order to compare the efficiency of the thermal and fast spectrum to transmute ^{241}Am, ORIGEN calculations have been made. Irradiation of ^{241}Am have been simulated under constant neutron flux of 3×10^{14} or 10^{15} n/cm^2/s, for the two different spectra (libraries PWRUS50.lib for the thermal spectrum and AMO-PUUUC.lib for the fast spectrum), and during a long period of time.

Fig. 2 shows the evolution of the actinides masses versus the time, for the four different cases (two different spectra and two different levels of flux).

In Fig. 2 we can see that the total amount of actinides is decreasing very slowly in fast spectrum. With a standard flux for fast spectrum ($\phi = 10^{15}$ n/cm^2/s), only 17% of the actinides are fissioned after 4000 days (11 years). Half of the actinide mass would be fissioned after 31 years. From Fig. 2 clearly the thermal spectrum is more efficient to transmute ^{241}Am. In the case of a standard thermal flux of 3×10^{14} n/cm^2/s, the disappearance of the actinides up to 95% can be reached after around 9000 days (less than 25 years). With a higher flux of 10^{15} n/cm^2/s better results can be obtained and the fission of the actinides up to 95% is reached after only 4000 days (less than 11 years).

Clearly transmutation rates are higher in thermal spectrum than in fast spectrum for the same level of flux [3]. The transmutation rate is also higher in a typical PWR flux than in a typical fast reactor flux, although the level of flux is higher in the latter. An incineration rate of 95% is achieved in 25 years in a standard PWR thermal flux ($\phi = 3 \times 10^{14}$ n/cm^2/s), while in a typical fast reactor flux ($\phi = 10^{15}$ n/cm^2/s), 150 years is required.

The transmutation rate can be further increased by using a higher thermal flux. In a thermal flux of 10^{15} n/

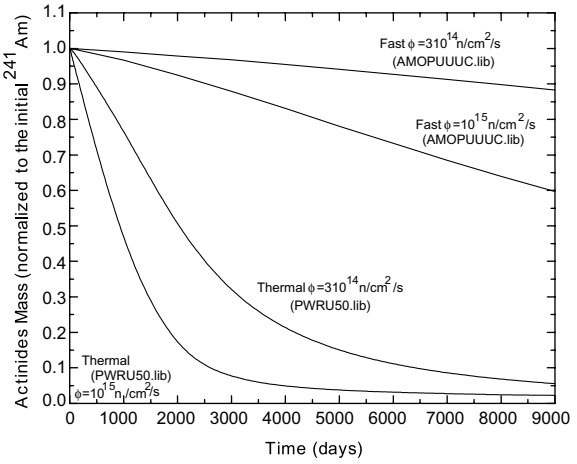

Fig. 2. Total actinide mass versus time in different neutron spectra and fluxes.

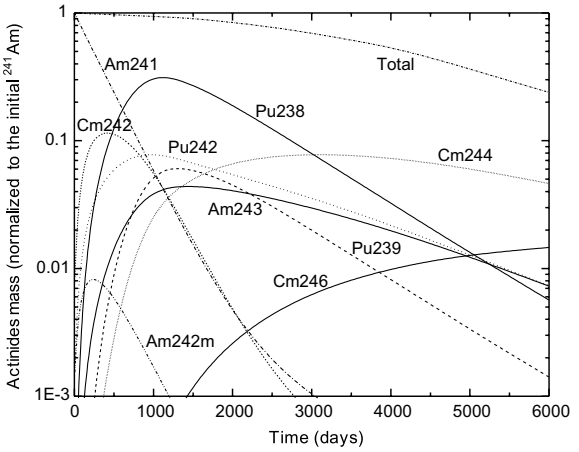

Fig. 3. Isotopic evolution in a thermal spectrum with a constant flux of 3×10^{14} n/cm^2/s. Actinide mass is normalized to the initial isotope mass.

cm^2/s, an incineration rate of 95% is achieved in 11 years. This high flux, however, is not reachable in conventional PWR due to technological limits (heat removal, etc). Such high thermal flux can, however, be reached in a moderated zone of a fast reactor. This idea of using thermal zones of a fast reactor has been proposed by EDF [6]. In this P&T strategy, Pu is multi-recycled in fast reactor, while special targets of Am are placed in moderated assemblies of the fast reactor for a long period of time (10 years). This strategy allows reducing the waste isolation times from 130 000 years (no P&T foreseen) up to 1500 years [4–6].

3.2. Evolution of the concentration of the isotopes in each studied case

3.2.1. Thermal spectrum, $\phi = 3 \times 10^{14}$ n/cm^2/s

Fig. 3 shows the evolution of the ^{241}Am and its main reaction products. We notice the quick build up of ^{242}Cm, coming directly from the decay of ^{242}Am. ^{242m}Am and ^{242}Pu are also build up quite quickly as they are the two secondary reactions (10% branching ratio of ^{242m}Am by direct capture of the ^{241}Am, and 15% of production of ^{242}Pu by successive captures of the ^{241}Am and ^{242}Am). In the mean time the mass of ^{241}Am decreases very fast. Following the build up of ^{242}Cm (maximum mass reached at 500 days), we see the build up of ^{238}Pu (maximum mass reached at 1200 days), decay product of ^{242}Cm. Then we notice the build up of ^{239}Pu (produced by capture of ^{238}Pu) which fissions. ^{242}Pu is quite slow to disappear as its burnout time is quite long (4 years).

This is the isotope (and its reaction products) that takes the longest time to be burnt. ^{244}Cm is built up quite slowly and will be the last one to disappear (captures to ^{245}Cm and then fissions) as it is created by the successive decay and capture of ^{242}Pu, and as its burnout time is also quite long (4.6 years).

3.2.2. Thermal spectrum, $\phi = 10^{15}$ n/cm^2/s

Fig. 4 shows the evolution of the ^{241}Am and its main reaction products. Here again is studied the irradiation in thermal spectrum of ^{241}Am, but under a higher flux. Then the same reactions occur and the same isotopes are created. Quickly ^{242m}Am, ^{242}Cm and ^{242}Pu are build up, but in this case, faster than before (few hundred days), due to the higher flux that accelerates the capture. Just afterwards, around 500 days, we notice the build up of ^{243}Am, reaction product of ^{242}Pu (its mass peak arises just after the one of ^{242}Pu). Following the mass peak of

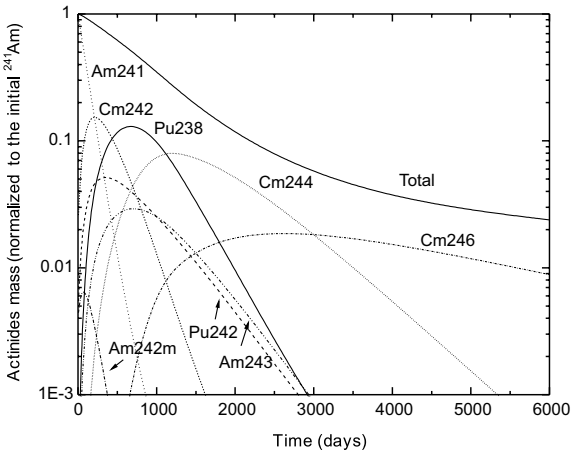

Fig. 4. Isotopic evolution in a thermal spectrum with a constant flux of 10^{15} n/cm^2/s. Actinide masse is normalized to the initial isotope mass.

^{242}Cm, around 700 days, the one of ^{238}Pu, decay product of ^{242}Cm, arises. In this case we observe the decrease of the quantity of ^{244}Cm (peak around 1100 days), and some creation (although in lower quantity than ^{244}Cm) of ^{246}Cm. The ^{245}Cm (created by capture of ^{244}Cm) mainly fissions, but its capture cross-section is also important and can lead to the creation of ^{246}Cm.

3.2.3. Fast spectrum, $\phi = 10^{15}$ n/cm^2/s

Fig. 5 shows the evolution of ^{241}Am and its main reaction products. We notice the build up of some isotopes coming from the capture of ^{241}Am: ^{242m}Am, ^{242}Cm, and then, coming from ^{242}Cm, the ^{238}Pu and ^{239}Pu. We also notice the presence of a small quantity of ^{237}Np, α decay product of ^{241}Am.

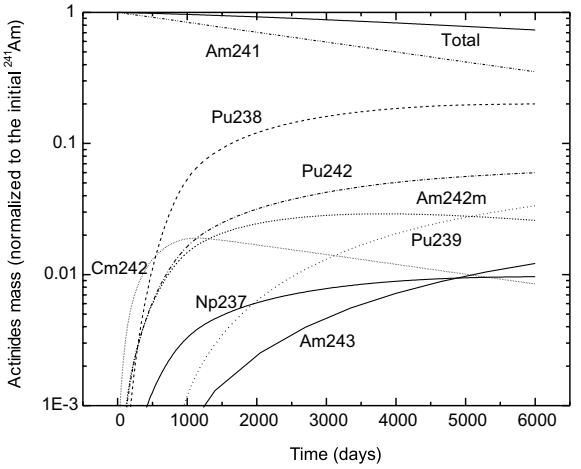

Fig. 5. Isotopic evolution in a fast spectrum with a constant flux of 10^{15} n/cm^2/s. Actinide mass is normalized to the initial isotope mass.

4. The burnout time

4.1. Definition of the burnout time and individual values for the main actinides

The time evolution of a quantity N of an isotope under neutron irradiation is [1]

$$\frac{dN}{dt} = -(\lambda + \sigma\phi)N,$$

where ϕ is the flux, σ the absorption cross-section, and λ the decay constant. If decay is the predominant reaction, then the time evolution of the quantity N is defined by

$$\frac{dN}{dt} = -\lambda N.$$

This equation leads by integration to $N(t) = N_0 e^{-\lambda t}$ where N_0 is the initial quantity. The necessary time to reduce by half the initial quantity is then $\tau_\lambda = \ln 2/\lambda$, where τ_λ is the half-life of the isotope.

By extension, if neutron absorption is the predominant reaction, then the time evolution of the quantity N is defined by

$$\frac{dN}{dt} = -\sigma\phi N.$$

Table 2
Actinide burnout times for thermal and fast neutron spectra

	Half-life (years)	Burnout time (days)			
		Thermal 3×10^{14} (n/cm^2/s)	Thermal 10^{15} (n/cm^2/s)	Fast 3×10^{14} (n/cm^2/s)	Fast 10^{15} (n/cm^2/s)
^{235}U	Stable	529	159	11 162	3349
^{238}U	Stable	26 181	7854	80 637	24 191
^{237}Np	2.14×10^6	849	255	14 691	4407
^{238}Pu	87.7	836	251	14 212	4264
^{239}Pu	2.4×10^4	168	50	11 673	3502
^{240}Pu	6.6×10^3	186	56	29 426	8828
^{241}Pu	14.3	188	56	9291	2787
^{242}Pu	3.7×10^5	836	251	37 582	11 275
^{241}Am	432	317	95	16 058	4817
^{242m}Am	141	56	17	6275	1883
^{243}Am	7.37×10^3	534	1601	20 933	6280
^{242}Cm	162.8 days	4313	1294	52 807	15 842
^{243}Cm	29.1	351	105	9450	2835
^{244}Cm	18.1	1826	548	20 871	6261
^{245}Cm	8.5×10^3	156	47	9245	2773
^{246}Cm	4.73×10^3	7619	2286	49 411	14 823

This equation leads by integration to $N(t) = N_0 e^{-\sigma\phi t}$ where N_0 is the initial quantity. The necessary time to reduce by half the initial quantity is then $\tau_{BU} = \ln 2/\sigma\phi$ where τ_{BU} is the burnout time of the isotope.

The burnout time then is the time necessary to transmute by neutron absorption half of the quantity of a nuclide under neutron irradiation. Ten burnout times are required to transmute a significant amount of the considered actinide (up to $1/2^{10} = 1/1024 = 0.001$).

Table 2 shows the burnout times in thermal and fast neutron spectrum and for two different levels of flux. We notice that burnout times have very different values depending on the actinide, the spectrum and the flux level. For any actinide, the burnout time is shorter in thermal spectrum, due to the higher cross-sections. Higher flux accelerates the transmutation.

This burnout time refers to the disappearance of an isotope. If the capture is the main reaction, then its 'transmutation' leads to the creation of another actinide.

4.2. Evaluation of an 'overall' burnout time for ^{241}Am

We introduce in this section the concept of the 'overall' burnout time. The calculation method is based on the following algorithm (Fig. 6):

1. For the selected nuclide, the individual burnout time is evaluated, i.e. $\ln 2/(\lambda + \sigma_a\phi)$, where λ is the decay constant, σ_a the absorption cross-section ($\sigma_a = \sigma_f + \sigma_c$), and ϕ the neutron flux.
2. From a comparison of the branching ratios of the various processes (decay, fission, capture), we select the reaction with the highest branching ratio (BR_λ, BR_c or BR_f).

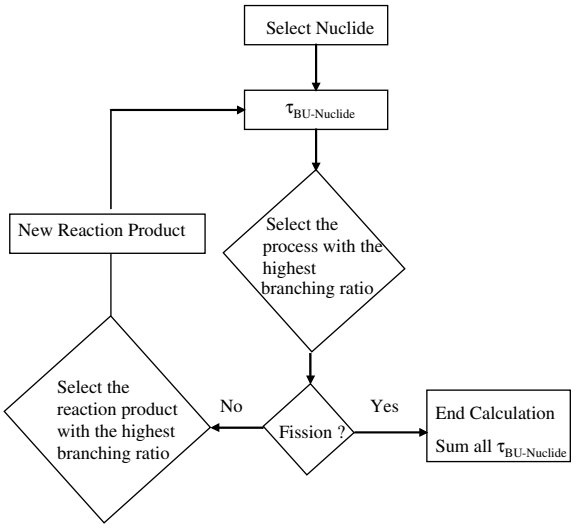

Fig. 6. Overall burnout time algorithm.

3. If multiple reaction products, then select the reaction product with the highest branching ratio ($BR_{\lambda 1}$, $BR_{\lambda 2}$, etc, BR_{c1}, BR_{c2}, etc).
4. We repeat steps 1, 2 and 3 for each new reaction product to obtain the burnout time and the main reaction product until a new reaction product is selected for which fission is the main process.
5. We add the individual burnout time of all the reaction products selected to obtain the 'overall' burnout time.

Consider the case of ^{241}Am. In both thermal and fast spectrum, several reactions occur before reaching the fission of one of ^{241}Am reaction products. The branching ratios are calculated for each nuclide with the following formulas [1]:

$$BR_{decay} = \lambda/[\lambda + (\sigma_f + \sigma_c)\cdot\phi],$$

$$BR_{capture} = \sigma_c\cdot\phi/[\lambda + (\sigma_f + \sigma_c)\cdot\phi],$$

$$BR_{fission} = \sigma_f\cdot\phi/[\lambda + (\sigma_f + \sigma_c)\cdot\phi].$$

The 'overall' burnout time of ^{241}Am in thermal spectrum has been calculated with the algorithm:

(i) ^{241}Am burnout time is calculated.
(ii) The three branching ratios are: $BR_{decay} = 0.2\%$, $BR_{capture} = 98.5\%$, $BR_{fission} = 1.25\%$. Capture of ^{241}Am has the highest branching ratio.
(iii) The capture leads to ^{242}Am ground state ($BR_{c1} = 90\%$) and to ^{242m}Am metastable state ($BR_{c2} = 10\%$). Then ^{242}Am is selected as main reaction product.
(iv) ^{242}Am burnout time is calculated.
(v) $BR_{decay} = 100\%$ for ^{242}Am. The decay is the main process.
(vi) ^{242}Am mainly decays to ^{242}Cm ($BR_{\lambda 1} = 83\%$) and to ^{242}Pu ($BR_{\lambda 2} = 17\%$). Therefore the main reaction product is ^{242}Cm.
(vii) ^{242}Cm burnout time is calculated.
(viii) $BR_{decay} = 99.9\%$. ^{242}Cm mainly decays to ^{238}Pu.
(ix) ^{238}Pu burnout time is calculated.
(x) $BR_{decay} = 2.54\%$, $BR_{capture} = 90.77\%$, $BR_{fission} = 6.71\%$. ^{238}Pu mainly captures to ^{239}Pu.
(xi) ^{239}Pu burnout time is calculated.
(xii) $BR_{decay} = 0\%$, $BR_{capture} = 36.26\%$, $BR_{fission} = 63.73\%$. ^{239}Pu mainly fissions.
(xiii) Sum of the burnout time of all the involved isotopes.

In fast spectrum the calculation process is the same, except that it ends up at the ^{238}Pu which mainly fissions.

Eq. (1) shows the calculation of the 'overall' burnout time for ^{241}Am (the term in bracket is only included for the thermal spectrum calculations):

$$\tau_{BU} = \tau_{BU(Am241)} + \tau_{BU(Am242)} + \tau_{BU(Cm242)} + \tau_{BU(Pu238)} + (\tau_{BU(Pu239)}). \quad (1)$$

The specific values of the burnout times taken into account are summarized in Table 3.

For a standard neutron flux in thermal spectrum (3×10^{14} n/cm^2/s), we have a total burnout time of $T = 1481$ days (4 years). Then to transmute the total amount of actinides we need 1481 days $\times$ 10 years, i.e. 40 years. For a higher flux ($\phi = 10^{15}$ n/cm^2/s) we obtain $T = 556$ days (1.5 years), then it requires 15 years to fission the ^{241}Am and all its reaction products. For a standard fast spectrum ($\phi = 10^{15}$ n/cm^2/s), 25 years is obtained as the overall burnout time, that means that 250 years are required to transmute the actinides. We can already notice that the thermal spectrum is better to transmute ^{241}Am as it is much faster to fission it and all its reaction products.

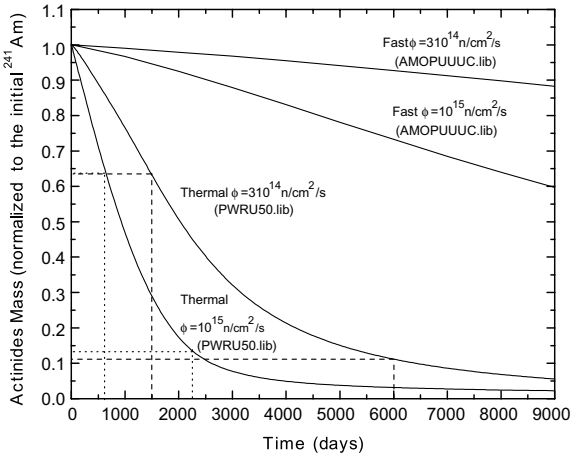

Fig. 7. Actinides mass evolution in different spectrum and fluxes versus the time.

5. Comparison of ORIGEN calculations and the overall burnout time method

Fig. 7 shows the evolution of the actinides mass versus the time, for the four different cases (two different spectra and two different levels of flux). We observe in Fig. 7 that, for thermal spectrum, and with high flux level, after the theoretical burnout time of 556 days, the remaining quantity of actinides is 64%. It is more than the theoretical value of 50% (Eq. (1)). We also notice that after 2200 days (four burnout times) the remaining actinide quantity is around 11%, where it should be 6% in theory (after four burnout times, one 16th of the quantity should remain, i.e., 6%). This is due to the fact that only the main reactions are taken into account in the burnout time calculation. The parallel reaction that occurs (decay of ^{242}Am to ^{242}Pu) leads to the creation of isotopes such as ^{243}Am or ^{244}Cm. Some of them have very long burnout time and remain present longer in the fuel. Then the actinide mass is increasing, as we notice in ORIGEN calculations.

The same comparison has been made for thermal spectrum and with a typical PWR flux level. After one burnout time (1480 days), 64% of the actinides mass has not been fissioned, and after four burnout times (6000 days), only 10% (in theory it should be one 16th, i.e. 6%). Here again the values obtained by ORIGEN calculations are above the theoretical ones, but this is due to the parallel reactions that are not taken into account in the overall burnout time method, and which create more actinides.

Table 3
Burnout times of the isotopes in the ^{241}Am reaction chain

	Main reaction	Thermal flux 3×10^{14}	Thermal flux 10^{15}	Fast flux 3×10^{14}	Fast flux 10^{15}
$\tau_{BU(Am241)}$	Capture (days)	317	95	16058	4817
$\tau_{BU(Am242)}$	Decay (days)	0.667	0.667	0.667	0.667
$\tau_{BU(Cm242)}$	Decay (days)	160	160	160	160
$\tau_{BU(Pu238)}$	Capture-fission (days)	836	251	14212	4264
$\tau_{BU(Pu239)}$	Fission (days)	168	50	–	–
Total		1481 (4 years)	556 (1.5 years)	30430 (83 years)	9241 (25 years)

Table 4
Comparison between the remaining mass with ORIGEN and burnout time calculations after one and four burnout times

	Eq. (1)	Thermal flux 3×10^{14} ORIGEN	Thermal flux 10^{15} ORIGEN	Fast flux 10^{15} ORIGEN
One burnout time (%)	50	64	64	60
Four burnout times (%)	6.25	12	10	10

For the typical fast flux, the overall burnout time is 25 years, and at this time 60% of the actinide mass is remaining in ORIGEN calculations.

Table 4 shows the remaining mass in ORIGEN calculations compared to the theoretical ones after the calculated burnout time:

This overall burnout time method gives rapidly a good order of magnitude for the time necessary to fission an isotope and all its reaction products.

6. Conclusion

We have studied the transmutation of ^{241}Am in both thermal and fast neutron spectra with two methods. We have proposed simple method to evaluate the overall time required to transmute an isotope. It has been based on an algorithm. The main reaction chain of a selected nuclide is considered. The chain stops when a reaction product mainly fissions. The sum of the burnout times of the reaction products involved in the chain represents the overall burnout time of the initial nuclide. Comparison of the results with a more exact ORIGEN calculation shows good agreement. This method gives quickly a good order of magnitude of the overall transmutation rate of an actinide in fast and thermal neutron spectra. This method can be further applied to other actinides.

References

[1] J. Magill, Nuclides.net. An Integrated Environment for Computations on Radionuclides and their Radiation, Springer Verlag, ISBN 3-540-43448-8, 2002. For further information, see the Nuclides.net website at http://www.nuclides.net.

[2] http://www.ornl.gov/origen-arp/origen-arp.html.

[3] J.M. Li, J.L. Kloosterman, Netherlands Energy Research Foundation ECN, Report ECN-C-95-055, 1995.

[4] D. Haas, R.J.M. Konings, J. Magill, V. Berthou, in: Proceedings of ENC 2002 Scientific Seminar, Lille, France, 7–9 October 2002.

[5] J. Magill, V. Berthou, J. Galy, R. Schenkel, H.-W. Wiese, G. Heusener, J. Tommasi, G. Youinou, European Commission, JRC, Institute for Transuranium Elements, Technical Report, JRC-ITU-TN-2002/24.

[6] J.-P. Grouiller, S. Pillon, C. de Saint Jean, F. Varaine, L. Leyval, G. Vambenepe, B. Carlier, these Proceedings doi:10.1016/S0022-3115(03)00184-3.