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On the transmutation of Am in a fast lead-cooled system

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Abstract. Characteristics of the equilibrium fuel cycle for the core or a blanket of ADS having the structure of the core of a fast lead-cooled reactor of type BREST (Russian abbreviation for 'Bystryy Reaktor so Svintsovym Teplonositelem') in a mode of americium transmutation are calculated. Americium loading was taken 5% of heavy atoms. Keeping the average multiplication factor the same as in a standard equilibrium cycle, reactivity swing over 1 year's microcycle is about 1%, that demands partial fuel reloading with a periodicity of about one month. For one year of operation, 61 kg of americium is destroyed, and due to increased ^{238}Pu content, americium is mainly converted to fission products. Thus in a system of 1 GWt (thermal), 87 kg of americium can be transmuted yearly. The estimate of the reactivity void effect has shown that it increases to 0.6% almost linearly with the void fraction increasing up to 25% and reaches its maximum of 0.7% at a void fraction of about 50%. Application of similar strategy for ADS with a sub-criticality level ≈0.96–0.98 can essentially relax safety problems related to positive void effects.

Keywords. Americium; transmutation; lead-cooled; fuel cycle; reactivity.

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1. Introduction

Nuclear waste transmutation is an option to reduce the hazards in the spent nuclear fuel, that are posed by about 1–2% of the content of the spent fuel, leaving the other 98% relatively harmless over the long term. The problematic isotopes can be converted to either stable or short-lived hazards. The main elements found in spent fuel are long-lived radio-nuclides: major actinides (U, Pu), minor actinides (Np, Am, Cm) and fission products. ^{241}Am ($T_{1/2} = 432$ years) is one of the main contributors among the minor actinides (see figure 1) and its amount in spent fuel increases with storage time due to the decay of ^{241}Pu ($T_{1/2} = 14.2$ years).

A typical yearly americium discharge from 1 GWe LWR (WWR-1000) is about 20 kg (at 10 yr cooling time). The purpose of the work was to study the transmutation of americium in a fast spectrum lead-cooled system. The model of BREST-OD-300 core [1] was used in the study. Similar composition can be used in the blanket of accelerator driven systems.

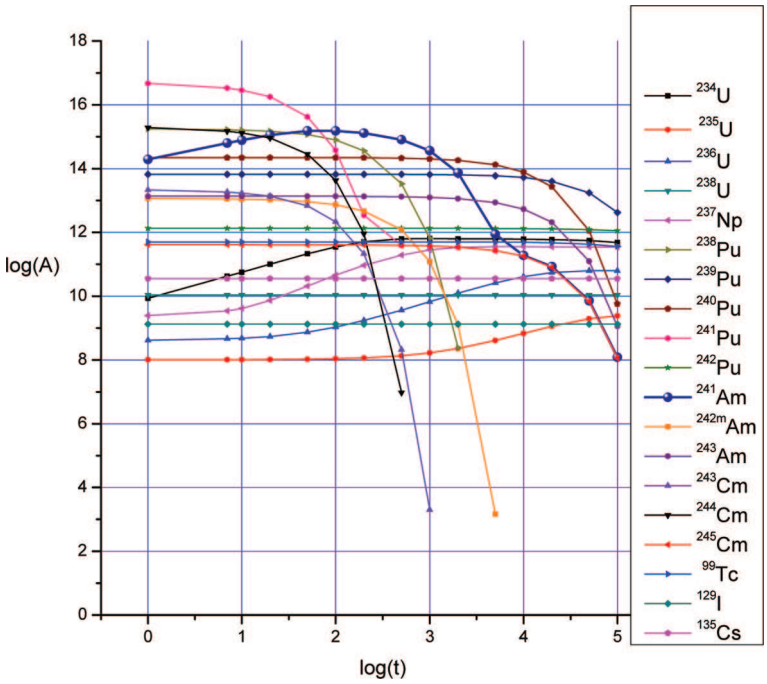

Figure 1. Activities A (Bq/tHM) as a function of decay time t (yr), of PWR spent fuel (MOX type) having burnup of 33 GWd/tHM.

2. The model

The study of americium transmutation in a model of fast reactor of BREST-OD-300 (700 MW thermal power) was performed under the following main assumptions: (1) mean value of the multiplication factor over fuel cycle was the same as that for the standard equilibrium cycle, (2) the initial inventory of americium differs from its equilibrium value and is about 5% [1], (3) the history of the irradiation is repeated from cycle to cycle, that is the reactor is operated in an equilibrium mode. All chains for heavy nuclides and fission products were taken into account (about 110 isotopes). Fuel cycle length was taken as 5 years as in the standard cycle (at power load factor 0.75) with the burnup of 76, 63 and 46 MWd/kg for three zones of reactor core with total loading of 16.2 t of heavy atoms. The simulation of fuel cycles was performed by TRIFON code [2] until almost an equilibrium mode of operation was reached. It was supposed that a part of Pu was used in the next cycle, the burnt americium was added (from the spent fuel of LWR) and Cm was sent to a storage to decay into ^{239}Pu and ^{240}Pu.

3. The results

To understand the influence of different isotopes on the reactivity of the system, the same sample amount of each nuclide (numerically equal to 1% of total Pu)

Table 1. The change of reactivity (%) on dependence on time t, years, due to the insertion of the sample amount of each nuclide.

t	0	1	2	3	4	5
^{243}Cm	1.66	1.29	1.00	0.79	0.62	0.49
^{245}Cm	1.51	1.21	0.97	0.79	0.65	0.53
^{241}Pu	1.09	0.81	0.61	0.46	0.35	0.27
^{239}Pu	0.73	0.61	0.51	0.43	0.37	0.31
^{238}Pu	0.40	0.36	0.33	0.30	0.28	0.25
^{242}Cm	0.30	0.37	0.36	0.33	0.30	0.28
^{244}Cm	0.10	0.14	0.16	0.18	0.19	0.20
^{240}Pu	0.05	0.08	0.10	0.11	0.12	0.12
^{242}Pu	0.02	0.01	0.01	0.00	0.00	0.00
^{238}U	−0.05	−0.04	−0.03	−0.02	−0,01	−0.01
^{237}Np	−0.16	−0.11	−0.07	−0.04	−0.01	0.01
^{241}Am	−0.16	−0.09	−0.04	0.00	0.03	0.05
^{243}Am	−0.18	−0.16	−0.13	−0.10	−0.08	−0.06

was inserted into the fuel in the standard equilibrium cycle (see table 1). The initial negative influence of ^{241}Am changes to positive one due to its conversion into ^{238}Pu and then into ^{239}Pu. ^{238}Pu is not a bad fissile and fertile isotope, since its influence on the reactivity is rather high and stable (the latter due to its conversion into good fissile isotope ^{239}Pu). It should also be noted that ^{237}Np, in the conditions typical for a reactor with lead coolant, has poor fissile properties and can hardly be used in the multiplier region of one-directionally coupled system, as was expected in a number of papers (it has good properties as a fissile material with the threshold fission cross-section in hard neutron spectrum without intensive moderation of neutrons as in the case for gaseous coolant).

The difference in the initial nuclear content in standard and Am modes of operation is shown in table 2.

The results of the calculation have shown that the reactivity changes over one year's micro-cycle increased to 1% as compared to 0.1% in the standard cycle. This means that the time interval between partial re-loadings in the standard cycle should be decreased to one month instead of one year to maintain the efficiency of the control rod system no higher than in the standard cycle. Time dependence of isotope's masses in the reactor core is shown in figure 2.

The ratio of plutonium isotope's masses at the EOC and BOC are close to one another and are close to the value 1.1 (table 3). Therefore, relative to plutonium, the cycle is close to equilibrium with a conversion ratio of 1.1. It is important to note that for ^{238}Pu the ratio of its masses at the EOC and BOC is close to that for the rest of the plutonium isotopes. However, in the first cycle of americium transmutation, the ^{238}Pu contents increases due to capture of neutrons by ^{241}Am with its conversion into ^{242}Cm and then into ^{238}Pu with ^{242}Cm having a half-life of 162 days.

For the determination of americium contribution into the process of chain transmutations, additional calculations were performed, in which the total fuel volume

Table 2. The initial nuclear content (%) for standard (Std) cycle and the cycle with americium transmutation (Am).

	Cycle	
Nuclide	Std	Am
^{234}U	0.000	0.435
^{235}U	0.067	0.107
^{236}U	0.041	0.098
^{238}U	85.404	78.840
^{237}Np	0.063	0.109
^{238}Pu	0.158	1.847
^{239}Pu	8.981	7.435
^{240}Pu	4.123	3.891
^{241}Pu	0.485	0.476
^{242}Pu	0.304	0.878
^{241}Am	0.288	4.806
^{242m}Am	0.011	0.211
^{243}Am	0.074	0.867

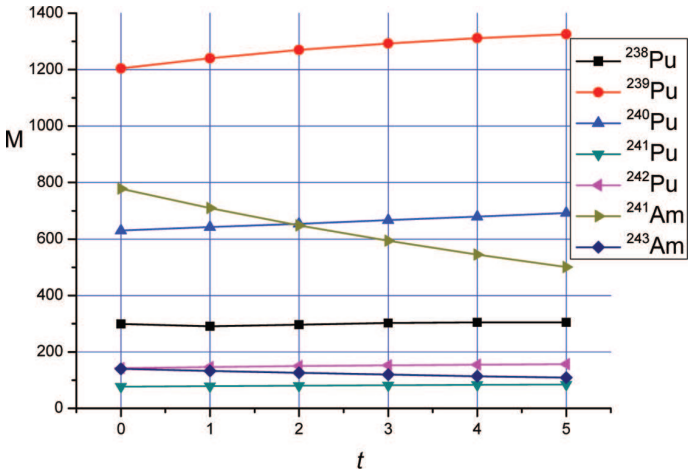

Figure 2. The dependence of isotope's masses (M) on time t (yr).

was divided into two parts – with americium and with the rest of the isotopes (uranium, plutonium, neptunium) in accordance with their mass content (table 2). It should be noted (see table 4) that of the total americium mass (953 kg) in all the chain terms, only 122 kg or 13% convert to fission products, and the rest mass of americium (183 kg) converts into plutonium and curium isotopes. However, the accumulation of a large amount of ^{238}Pu from americium (120 kg) in a high degree is compensated by burning-out of 115 kg of ^{238}Pu due to its increased content in the initial fuel. As a result, americium is mainly converted into fission products and the whole process does not lead to the essential accumulation of ^{238}Pu. Rather, high

Table 3. The isotope's masses for the BOC and EOC. Dm – the change of masses after 5 years of irradiation; m(EOC)/m(BOC) – the ratio of the final mass to the initial mass.

Time	m(BOC)	m(EOC)	Dm	$\frac{m(\text{EOC})}{m(\text{BOC})}$
^{234}U	70.5	66.1	−4.4	
^{235}U	17.3	16.6	−0.74	
^{236}U	15.9	16.0	0.11	
^{238}U	12772.0	11822.0	−950.0	
^{237}Np	17.79	19.8	2.06	
^{238}Pu	299.3	304.5	5.18	1.074
^{239}Pu	1204.5	1325.5	121.0	1.10
^{240}Pu	630.4	692.4	62.1	1.10
^{241}Pu	77.0	84.6	7.6	1.10
^{242}Pu	142.2	156.3	14.0	1.10
^{241}Am	778.5	500.5	−278.0	
^{242}Am	0	0.08	0.08	
^{242m}Am	34.2	37.8	3.5	
^{243}Am	140.4	109.2	−31.2	
^{242}Cm	0	17.9	17.9	
^{243}Cm	0	1.5	1.5	
^{244}Cm	0	35.5	35.5	
^{245}Cm	0	2.58	2.58	
Total	16200.0	15210.2	−989.2	

content of ^{238}Pu at the BOC (about 2% of heavy atoms) has a positive influence on the reactivity, slightly decreasing in time, i.e. this isotope turns out to be a good fissionable material with a positive contribution to the balance of neutrons (see table 1).

For five years of irradiation, 955 kg of U are burnt; 211 kg of plutonium, 12 kg of ^{237}Np and 40 kg of curium (^{243}Cm, ^{244}Cm, ^{245}Cm) are accumulated; 990 kg of heavy atoms are converted to fission products; 305 kg of americium (of them 91% ^{241}Am) are eliminated, a part of which is converted into plutonium and curium isotopes and another part into fission products. Curium was not supposed to be returned into the reactor, but moved into a storage, where ^{243}Cm and ^{244}Cm decay into ^{239}Pu and ^{240}Pu with half-lives of 30 and 18.1 years respectively. Thus 61 kg of americium are eliminated yearly.

A more thorough analysis has shown that only 122 kg of the total amount of americium (953 kg) are converted to fission products (13%) out of the total mass of americium, while the rest of the mass is converted into plutonium and curium. Nevertheless, a large amount of ^{238}Pu (136 kg) generated from americium in a high degree is compensated by the burning-out of 113 kg of this isotope contained in the fuel component. Together this leads to equilibrium mode for this important isotope.

The estimate of the reactivity void effect has shown that it increases to 0.6% almost linearly with the void fraction increasing up to 25% and reaches its maximum

Table 4. Partial contribution to mass changes for 5 year's fuel cycle (kg).

Nuclide	Fuel	Americium	Total
^{234}U	−6.67	2.26	−4.41
^{235}U	−0.84	0.10	−0.74
^{236}U	0.11	0.00	0.11
^{237}U	0.02	0.00	0.02
^{238}U	−950.00	0.00	−950.00
^{237}Np	−1.85	3.91	2.06
^{239}Np	1.46	0.00	1.46
^{238}Pu	−115.11	120.28	5.18
^{239}Pu	112.36	8.68	121.04
^{240}Pu	58.07	4.03	62.10
^{241}Pu	7.43	0.16	7.60
^{242}Pu	−16.18	30.21	14.03
^{241}Am	28.18	−306.22	−278.05
^{242}Am	0.00	0.08	0.08
^{242m}Am	0.74	2.79	3.53
^{243}Am	14.03	−45.23	−31.20
^{242}Cm	−0.09	17.95	17.86
^{243}Cm	−0.11	1.61	1.50
^{244}Cm	0.80	34.65	35.46
^{245}Cm	−0.13	2.71	2.58

of 0.7% at the void fraction of about 50%. This can bring about some safety problems in reactor operation.

Application of similar strategy for accelerator driven systems with a sub-criticality level of ≈0.96–0.98 can essentially relax this problem. Accelerator current must be 10–20 mA (for lead target and 1 GeV proton energy). Besides, in this case the time period between partial fuel reloadings can be increased up to 1 year. However, power flattening factor in the system with external neutron source will be higher (1.5–2 times) than in the reactor.

4. Conclusions

The main purpose of this paper was to demonstrate the possibility of a fast lead-cooled reactor to convert the most hazardous minor actinide Am to less hazardous fission products, that transmutation of which needs special considerations. It is shown that for a model of fast lead-cooled reactor of thermal power 700 MW in one year of operation, 61 kg of americium can be destroyed. At the increased content of ^{238}Pu, the americium is mainly converted to fission products. The estimate of the reactivity void effect has shown that it increases to 0.6% with the void fraction increasing up to 25% and reaching its maximum of 0.7% at the void fraction ≈50%. Application of similar strategy for ADS with a sub-criticality level of ≈0.96–0.98 can essentially relax safety problems related to positive void effects.

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