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APPLICATION OF POWER REACTORS FOR TRANSMUTATION OF ACTINIDES

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ABSTRACT

Actinides from spent nuclear fuel are the most dangerous type of radioactive waste. The real way to reduce their amount and radiotoxicity is preliminary transmutation during 5-10 years before transportation of actinides into long-term storage. Purpose of this transmutation is to transform long-lived actinides, mainly ^{241}Am, into fission products and relatively short-lived ^{244}Cm. Equilibrium radiotoxicity in the storage after transmutation will be achieved much quicker and with much more low level, than without preliminary transmutation, because of respectively short half-life of ^{244}Cm.

Application of power reactors for transmutation will allow, if necessary, to realize in practice process of transmutation during comparatively short time, while building of specialized transmutation installations could be a long way in the future.

Possibility of realization of preliminary transmutation mode in power reactors of CANDU and VVER-1000 type is discussed in the paper. Passage to transmutation mode for the CANDU reactor does not require an essential changing of its technical parameters. Reactor CANDU can be capable to transmute Am and Cm from 2-3 reactors of VVER-1000 type. Transmutation mode for the VVER-1000 type reactor requires changing the design of fuel assemblies.

Keywords: actinide, transmutation, power reactors.

1. INTRODUCTION

Actinides from spent nuclear fuel are one of the most dangerous type of radioactive waste. Their incineration is the reliable way to decrease hazard of waste. The only known method to incinerate radioactive nuclei is transmutation.

The long-term transmutation in specialized high flux facilities with liquid fuel is most efficient mode of transmutation. It allows to incinerate the overwhelming fraction of actinides loaded in the transmutation facility. However construction of such facilities is a long way in the future.

In this paper, the approach of Bergelson (2003) was considered, in which actinides from spent fuel were transmuted in the reactor with solid fuel during a fixed time of 5-7 years, and then they were transferred into long-term storage. The purpose of such preliminary transmutation before ultimate storage was to incinerate a part of actinides and to transform another part into new actinides providing low level of radiotoxicity accumulated in the storage.

This approach is based on the fact that radiotoxicity of minor actinides from spent fuel of power reactor is determined by isotopes ^{241}Am and ^{244}Cm. At accumulation of minor actinides in long-term storage without transmutation, the main contribution to total radiotoxicity is given by ^{241}Am. So, if we consider actinides from PWR type reactor in 100 years of accumulation, the radiotoxicity of all isotopes, other than ^{241}Am, makes less than 10% with respect to total radiotoxicity. Consequently, it is expedient to perform preliminary transmutation of actinides before placement in long-term storage. The main purpose of this transmutation is to transmute ^{241}Am.

The concept of preliminary transmutation is illustrated by Fig.1. If actinides are periodically added to the long-term storage during a period of time without transmutation, their radiotoxicity increases continuously (curve *1*). However, if actinides are loaded into the storage after preliminary transmutation, the total radiotoxicity reaches a certain equilibrium level (curve *2* for 5-year transmutation and curve *3* for 7-year transmutation in CANDU reactor). This level can be much lower than radiotoxicity without transmutation. The stage of loading of storage facility by actinides (0-500 years in Fig.1) is followed by the stage of storage without addition of new actinides. Decay of actinides occurs in this stage (500-1000 years in Fig.1). So, preliminary transmutation allows to facilitate subsequent long-term storage.

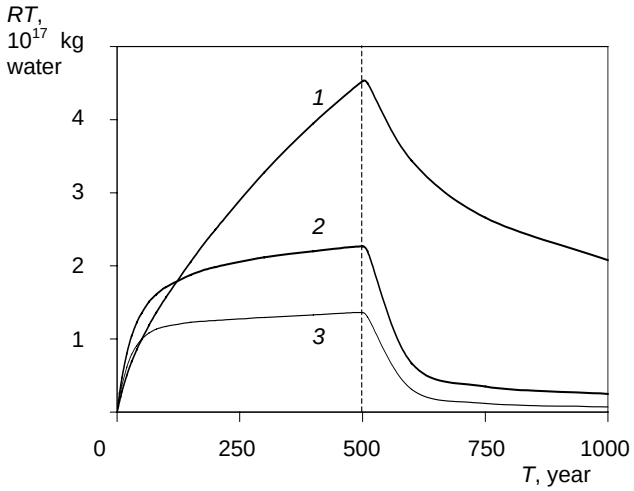

Fig.1. Radiotoxicity in storage facility during the stage of load of actinides an during the stage of the storage. 1 – load of actinides in the storage without transmutation, 2 – load of actinides after preliminary 5*-year transmutation, 3 – after* 7*-year transmutation*

Previous calculations of Bergelson (2003) demonstrated that power reactors with thermal neutron spectrum CANDU and VVER-1000 are most suitable for preliminary transmutation of actinides since transmutation in these reactors allows to limit long-term radiotoxicity in storage of spent fuel on the low level. Transmutation of actinides in fast reactors will result in essential increase of equilibrium radiotoxicity in the storage facility and time of its achievement.

In this paper, calculated results of transmutation mode in power reactors CANDU and VVER-1000 are presented. We supposed that transmutation was carried out in addition to the power production. We attempted to minimize the change of the reactor parameters in transmutation mode with respect to the standard mode of operation. Transmutation of americium and curium was considered. It was assumed that plutonium from spent fuel was not transmuted, as far as in conditions of closed fuel cycle the whole uranium and plutonium extracted from spent fuel should be recycled back in the reactor. However plutonium produced in the process of transmutation was taken into account for calculations. It was supposed that neptunium was not transmuted in the mode of preliminary transmutation since its radiotoxicity was negligible because of long half-life and did not influence upon equilibrium radiotoxicity in the storage facility.

The initial mixture of isotopes of Np, Am, Cm extracted from spent fuel (Table 1) does not multiply neutrons. However, nuclides with high cross sections of fission, including Pu isotopes, are formed in this mixture during the process of irradiation by thermal neutrons. An increase of neutron flux results in increase of the rate of accumulation of fissile nuclides and results in increase of multiplicating properties of the isotope mixture.

Table 1. Masses of Am and Cm isotopes in annual unloading of spent fuel after 3*-year cooling*

normalized by 1 *GW of electric power, kg/GW·year*

Nuclide	CANDU	VVER-1000
^{241}Am	6.8	5.9
^{242m}Am	-	0.02
^{243}Am	0.56	2.8
^{242}Cm	-	-
^{244}Cm	0.04	0.8
Total	7.4	9.5

2. TRANSMUTATION IN CANDU REACTOR

Heavy-water power reactor of CANDU type (Directory, 1994) operates with the fuel on the base of natural uranium with neutron flux density average over the fuel $\varphi_0 = 7 \cdot 10^{13}$ cm^{-2}s^{-1}. Reactor contains approximately 520 fuel channels with fuel assemblies. Fuel channels are located in nodes of square lattice. Standard fuel assembly consists of 37 pin fuel elements. Mean burnup of fuel is 4 MW·d/kg. For estimation of reactor parameters in transmutation mode, active core was separated into 20 square macrocells containing 5×5=25 channels with fuel assemblies in each macrocell (Fig.2).

It was supposed for calculations of transmutation mode that central fuel assembly in each macrocell was replaced by special transmutation target assembly with 37 pin target elements. Dimensions and design of target pins were identical to those for fuel pins. Target pins contained homogeneous mixture of americium and curium with the graphite instead of uranium fuel. Two variants of americium and curium loading in transmutation target assembly were considered: 9.5 kg and 4.75 kg. It will be recalled that annual unloading of americium and curium from one reactor VVER-1000 with 3-year cooling makes 9.5 kg (see Table 1).

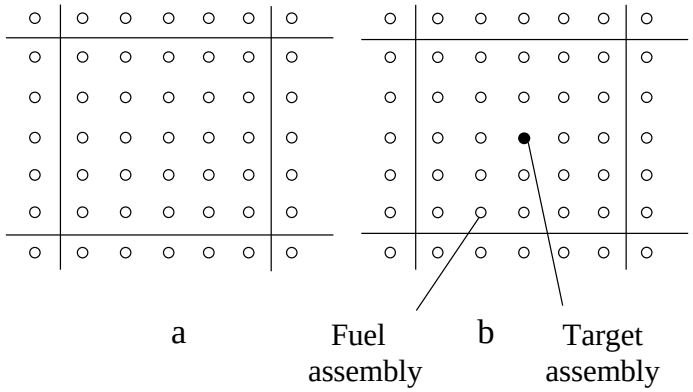

Fig. 2. Macrocell with all fuel assemblies (a) and macrocell with central target assembly for transmutation (b)

In order to value the possibility of realization of such transmutation mode in the CANDU reactor, comparative calculations of the square macrocell were made without and with loading of transmuted actinides in the central assembly of macrocell. Multiplication factor for square macrocell, neutron flux density, and heat power in central channel of macrocell were calculated using Monte-Carlo method and code MCNP-4b (Briesmeister, 1997). It was supposed for calculations that isotopic composition of fuel in fuel assemblies corresponded to average composition for the mode of continuous refuelling. This composition was calculated using module ORIGEN-S of the code SCALE (SCALE, 1993) for average burnup of 4 MW·d/t. Average thermal neutron flux density in the fuel was $\varphi_0 = 7 \cdot 10^{13}$ cm^{-2}s^{-1} (Directory, 1994).

Results of calculations – values of multiplication factors k for square macrocell, relative values of thermal neutron flux density φ/φ_0 and power p/p_0 for central channel of macrocell with transmutation target assembly are presented in Tables 2, 3, and 4 for different times of transmutation. Neutron flux density φ and power p corresponded to the central channel with transmutation target assembly, while φ_0 and p_0 corresponded to the fuel channel in the macrocell without target assembly. Multiplication factor in macrocell with fuel channels without target channel $k_0 = 1.056$.

Table 2. k, φ/φ_0, and p/p_0 in transmutation process for the load of 7.36 *kg of Am and Cm from*

CANDU reactor

	Time of transmutation, year			
	0	3	5	7
k	1.018	1.041	1.052	1.053
φ/φ_0	0.47	0.79	1.27	1.99
p/p_0	0.01	0.53	0.59	0.30

Table 3. k, φ/φ_0, and p/p_0 in transmutation process for the load of 4.75 *kg of Am and Cm from VVER-1000 reactor*

	Time of transmutation, year			
	0	3	5	7
k	1.025	1.057	1.056	1.055
φ/φ_0	0.75	1.47	1.95	2.68
p/p_0	0.01	0.44	0.27	0.16

Table 4. k, φ/φ_0, and p/p_0 in transmutation process for the load of 9.5 *kg of Am and Cm from VVER-1000 reactor*

	Time of transmutation, year			
	0	3	5	7
k	1.022	1.038	1.059	1.047
φ/φ_0	0.47	0.80	1.24	1.84
p/p_0	0.01	0.55	0.64	0.44

Change of isotopic composition of transmuted nuclides during transmutation was calculated using module ORIGEN-S of the code SCALE. Mean-square error of calculations of the *k* values was about 0.003. The load of americium and curium in transmutation target assembly slowly influences upon the multiplication factor, as it follows from the data of Tables 3 and 4. Neutron flux density in transmutation target assembly with load of americium and curium of 4.75 kg is higher than with load of 9.5 kg, and is higher than average flux density in the fuel. Heat power of transmutation target assembly makes an appreciable share of power of fuel assemblies.

The time of transmutation can be chosen according to the following considerations. If only fission products were loaded in the long-term storage, the time required to reach equilibrium radiotoxicity determined by ^{90}Sr and ^{137}Cs would be about 100 years. It could be supposed that in case of actinides, the time required to reach equilibrium also should be approximately 100 years.

Calculated ingestion radiotoxicity of actinides accumulated during time *T* in storage without transmutation as well as after 5- and 7-year preliminary transmutation of americium and curium in CANDU reactor is presented in Fig.3. The curve *1* describes the mode of storage without transmutation. The curves 2 and *3* describe the mode of storage after transmutation in CANDU reactor during 5 and 7 years. The rate of addition of actinides in storage corresponded to the average rate of feed of transmutation reactor by americium and curium from one VVER-1000 reactor.

These calculated results allow to formulate the following conclusions concerning to the transmutation mode of the power reactor.

1. Reactor CANDU can transmute americium and curium with the rate equal to the rate of their build-up in the reactor itself. In this mode of operation, fuel elements of seven fuel assemblies (that is 1.35% of total of 520 fuel assemblies) should be replaced by the same number of target elements with transmuted nuclides. The time of transmutation equals 7 years. Every year, one target assembly with irradiated target elements should be replaced by the "fresh" target assembly with americium and curium from spent fuel. Losses in the power production in this mode of operation do not exceed 1.35%.

2. Reactor CANDU can transmute americium and curium from 2-3 VVER-1000 reactors. In this mode, 20 fuel assemblies should be replaced by target assemblies. For the load of americium and curium in one target assembly 4.5 kg and time of irradiation 5 years, the rate of transmutation could be 19 kg per year. It should be reminded that annual unloading of americium and curium from one VVER-1000 reactor with 3-year cooling makes 9.5 kg. Thereby one CANDU reactor is capable to transmute americium and curium from two

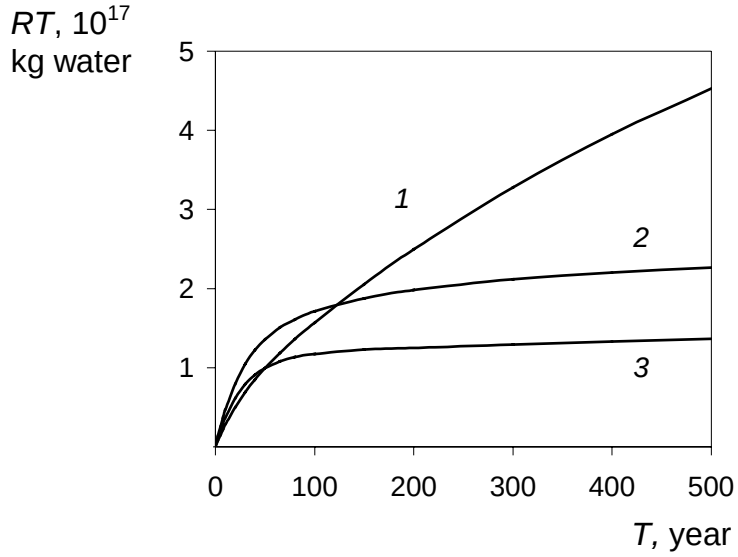

Fig.3. Radiotoxicity of actinides accumulated in the long-term storage facility: 1 – without transmutation, 2 – after 5*-year preliminary transmutation, 3 – after* 7*-year preliminary transmutation in CANDU reactor*

VVER-1000 reactors. For the load of americium and curium of 9.5 kg and irradiation time of 7 years, transmutation rate is 28.5 kg per year. That is to say, one CANDU reactor can support 3 supplier reactors of VVER-1000 type. In transmutation mode of CANDU reactor, active core contains transmutation target assemblies with actinides of different "ages" along with fuel assemblies in the ratio of 1 to 24. Therefore according to calculated data of the Tables 3 and 4, loss in the power production does not exceed ~1-2%.

3. In the mode of transmutation, the fuel consumption of the CANDU reactor decreases on account of burning of transmuted nuclides instead of the uranium. The saving of nuclear fuel makes about 1.5% in the mode of self-service and about 4% in the mode of support of 2-3 VVER-1000 reactors. The fuel saving could partially compensate the expenses of the target fabrication.

3. TRANSMUTATION IN VVER-1000 REACTOR

Transmutation mode of VVER-1000 power reactor should be quite different from that of CANDU reactor. Average flux density of thermal neutrons in the fuel of VVER-1000 reactor $\varphi_0 = 3 \cdot 10^{13}$ $cm^{-2}s^{-1}$ is approximately 2 times lower and number of fuel channels in the active core is 3 times less in comparison with heavy-water CANDU reactor. Besides, replacement of all fuel elements in one fuel assembly by target elements with transmuted nuclides will result in much higher depression of neutron flux and in much stronger influence on surrounding fuel assemblies. Therefore the mode of transmutation with replacement of certain fuel assemblies by the target assemblies and separate reloading of target and fuel assemblies can not be realized in light-water reactor.

For transmutation mode of VVER-1000, burnable absorbers with gadolinium should be used for replacement by target elements with transmuted actinides. Calculations made for the composition and geometry of fuel assemblies of VVER-1000 reactor (A VVER-1000, 2002) allowed us to formulate acceptable conditions for the realization of mode of transmutation of americium and curium in the VVER-1000 reactor.

In each fuel assembly 12 burnable absorbers should be replaced by target elements of the same size containing a mixture of graphite with transmuted isotopes. Both lifetime of fuel and time of target irradiation should be equal to 5 years. In the mode of partial reloading of fuel one fifth part of all assemblies (about 30 fuel assemblies with 360 target elements) should be reloaded every year. So, the mass of 9.5 kg of americium and curium could be loaded annually in the reactor. The mass of 26 g of nuclides to be transmuted will be contained in one target element in the beginning of irradiation. This mode of transmutation corresponds to the mode of self-service when the amount of transmuted nuclides equals to their build-up in the fuel of the reactor itself.

Calculated composition of nuclides contained in one target element before and after 5-year transmutation in VVER-1000 and CANDU reactors is presented in Table 5. The mass of actinides before transmutation is equal to the annual load of 9.5 kg divided by the number of 360 target elements in VVER reactor or divided by 37 target elements in one target assembly of CANDU reactor. After transmutation, the content of ^{241}Am with half-life of

432.2 years decreases approximately by 100 times. Radiotoxicity after transmutation is determined by ^{244}Cm with half-life of 18.1 years. Its content in target elements increases by 2 times during transmutation. These changes ensure reduction of equilibrium level of radiotoxicity in the long-term storage and reduction of time required to rich equilibrium. In the mode of self-service of VVER-1000 reactor, calculated saving of nuclear fuel makes about 3.6%, loss in the power production with account of power released in the targets does not exceed 1.5%.

Table 5. Masses of nuclides in one target element before and after 5*-year transmutation in CANDU and VVER-1000 reactors, g*

	CANDU		VVER-1000	
Nuclide	Before	After	Before	After
^{238}Pu	-	13.2	-	3.53
^{239}Pu	-	7.0	-	1.03
^{240}Pu	-	3.8	-	0.36
^{241}Pu	-	3.2	-	0.53
^{242}Pu	-	13.0	-	1.11
^{241}Am	159	1.6	16.4	0.19
^{242m}Am	0.5	0.049	0.06	0.01
^{243}Am	75.7	29.1	7.78	1.67
^{241}Am	-	2.2	-	0.17
^{242}Cm	-	0.22	-	0.02
^{244}Cm	21.6	52.4	2.2	4.12
^{245}Cm	-	2.4	-	0.83
^{246}Cm	-	3.2	-	0.56
Total	257	132	26.5	14.2

4. CONCLUSIONS

Heavy-water and light-water power reactors can be used for preliminary transmutation of actinides. Such transmutation allows to limit on relatively low level the radiotoxicity accumulated in long-term storage of spent fuel. CANDU reactor can operate both in transmutation and in self-service mode. One CANDU reactor in transmutation mode can support 3 supplier power reactors of VVER-1000 type. VVER-1000 reactor can operate in self-service mode.

Passage to transmutation mode in CANDU reactor does not require essential changes in parameters of the reactor. Similar mode for VVER-1000 assumes changing in the structure of fuel assemblies needed to increase the fuel lifetime.

Loss in the power production and saving of the fuel for both considered reactors-transmuters are of the same order of magnitude. In the mode of self-service, these values are 1 and 1.5% respectively for CANDU reactor and 1.5 and 3.5% for VVER-1000 reactor. It is necessary to note that these data of losses in the power production should be considered as approximate data. Exact data can be obtained either in experiments on reactors now in operation, or in result of precise steady-state and dynamic calculations of neutron distributions in the active core of considered reactors operating in transmutation modes.

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