IS IT EXPEDIENT TO USE THE FAST REACTORS FOR TRANSMUTATION OF THE LONG-LIVED RADIONUCLIDES?

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ABSTRACT

Main calculated characteristics of transmutation of the long-lived actinides in different reactors are compared in the paper. The radiotoxicity is used as the measure of radiation danger. Power thermal-neutron VVER-1000, fast neutron BN-600, SUPER-PHENIX-1200, and being developed BREST-1000 type reactors, and also high flux subcritical facility with liquid fuel ADS-800 are considered as the reactors-transmuters. Highest level of the equilibrium radotoxicity is reached in the reactor of BREST-1000 type. Reactors of other types gave some lower values. The much lower equilibrium radiotoxicity is reached in the facility ADS-800. It is by 4 - 8 times less than in other facilities. Because of use of liquid fuel, high flux of thermal neutrons, and continuous removal of the fission products, ADS-800 may operate in the mode of equilibrium with relatively low content of long-lived radiotoxicity in the blanket. The common result is that transmutation of long-lived radiotoxicity in the fast reactors is not effective in comparison with the analogous processes in other facilities

1. INTRODUCTION

Nowadays a significant part of the experts and specialists busy with the problems of transmutation holds to the idea that use of the fast reactors allows to realize the process of transmutation of actinides in the most effective way. This position is clearly seen from scientific publications in periodicals and the reports at the conferences. Such conception rests upon the well and widely known data in accordance with which practically all even isotopes of transuranium elements (except ^{238}Np and ^{242}Am) and odd isotopes ^{237}Np, ^{241}Am, and ^{243}Am are fissioned in fast spectrum better than in the thermal one. The paper [1] may be indicated as an example of such approach. And perhaps the fact is ignored that the even isotopes poorly fissioned in thermal spectrum possess rather high radiation capture cross-section while the odd isotopes possess high fission cross-sections for thermal neutrons.

In this paper, we compared the efficiency of transmutation of the long-lived nuclides in different reactors. The conception of the radiotoxicity R was used as the measure of radiation danger. The values of R for separate isotopes were defined on the base of maximum permissible activity in water according to the radiation safety standards [2]. The long-lived radiotoxicity of the spent nuclear fuel is defined mainly by plutonium and minor actinides - neptunium, americium and curium [3]. The contribution of the long-lived fission products is rather low. That is why we analyzed only

transmutation of the minor actinides. For this purpose we calculated the processes of transformation of them together with their daughter's nuclei including fission of them in reactor's facilities. We did not consider plutonium accumulated in the nuclear fuel in the process of operation of the power reactors. But we took into account formation of plutonium in the process of transmutation of the isotopes of neptunium, americium and curium. The plutonium is formed mainly as ^{238}Pu in the process of irradiation of ^{237}Np and of decay of ^{242}Cm.

The software package of the codes SCALE-4.3 [4] was used for the multigroup calculations. The existing power reactors on thermal neutrons of the VVER-1000 type, on fast neutrons of the types BN-600 and SUPER-PHENIX(SPX)-1200 [5], as well as the being developed BREST-1000 with lead coolant [6], and also high flux subcritical ADS-800 with liquid fuel described in [7] were considered as the reactors-transmuters. For description of the process of transmutation, we used the model where continuous irradiation of the being transmuted nuclides under constant neutron fluxes and spectra was supposed. We took neutron fluxes and spectra for this model the same as average over the core fluxes and spectra in the fuels of the considered reactors. The fact that power reactors are operating with capacity factor of approximately 0.8 was taken into account by reduction of neutron fluxes for actinide irradiation by 20 %. Such simplified model allows to compare the results of transmutation in different facilities by a simple and demonstrative method.

2. CALCULATION RESULTS

It was thought that the process of transmutation is being realized under continuous feed by new portions of minor actinides. We accepted the rate of feed to be equal to the average rate of accumulation of the minor actinides in one power reactor of the VVER-1000 or SUPER-PHENIX(SPX)-1200 types. The values and compositions of the feed were calculated for burn-up of the fuel in the reactor of the type VVER-1000 up to 40 GWday/ton and in the reactor of the type SUPER-PHENIX-1200 - to 44 GWday/ton correspondingly. The calculated amount of the minor actinides extracted from the spent nuclear fuel after 10 years cooling is presented in the table I. It is normalized by 1 GW of electric power. This means that the data for the SPX-1200 are recalculated for power 1000 MW.

Table I. The amount of the minor actinides extracted from the spent nuclear fuel, kg/year

Reactor	^{237}Np	^{241}Am	^{242}Am^{m}	^{243}Am	^{244}Cm	Sum
VVER-1000	13.8	15.2	0.024	2.8	0.6	32.4
SPX-1200	4.6	70.7	0.25	5.8	0.45	81.8

The results of calculations of continuous irradiation of minor actinides in different reactors-transmuters are presented in the figures 1 and 2 in the form of dependence of total radiotoxicity of minor actinides and plutonium formed from them upon the irradiation time. Data are normalized by the feed from one power reactor VVER-1000 (fig.1) or SPX-1200 (fig.2).

The main parameters of the transmutation modes are given in the tables II and III. Under the condition of continuous feed, some kind of equilibrium appears in a reactor-transmuter when the rate of incineration of transmuted nuclides becomes equal to the rate of feed. Plutonium formed in the process of transmutation is also included into the equilibrium mass of minor actinides. The last lines of the tables II and II present the period of time τ which is used for comparison of accumulation of radiotoxicity in a reactor-transmuter and in a storage facility without transmutation. So τ is the period of time for which the radiotoxicity of minor actinides accumulated in a reactor-transmuter becomes equal to radiotoxity of neptunium, americium, and curium accumulated in a long-term storage facility.

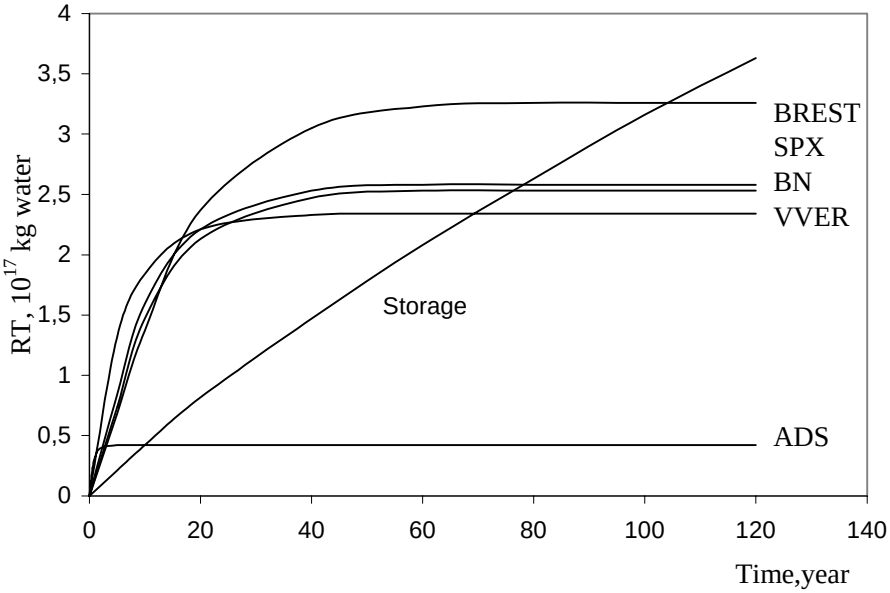

Fig.1. Radiotoxicity R accumulated in storage and transmutation facilities with ADS-800, VVER-1000, BN-800, SUPER-PHENIX-1200, and BREST-1000 type reactors at feed by actinides extracted from VVER-1000 type reactor

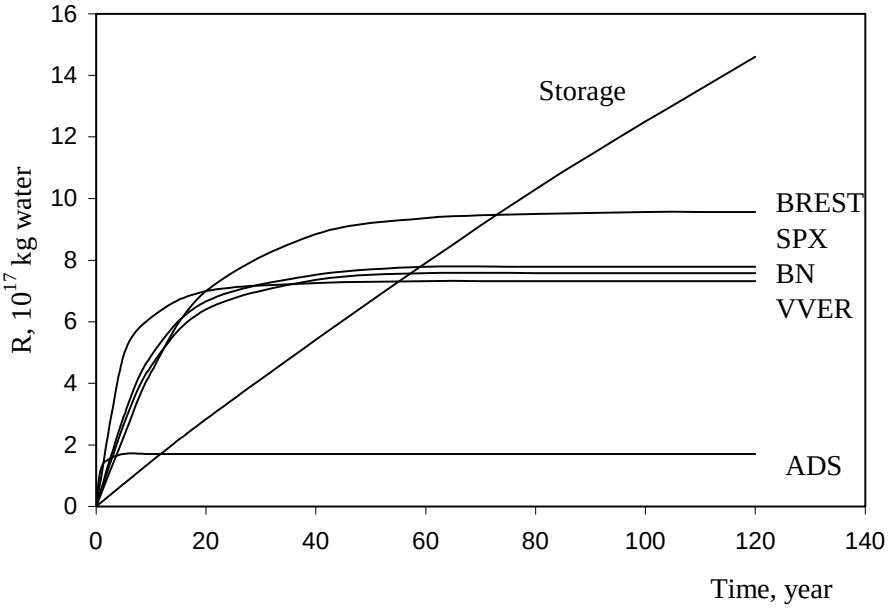

Fig.2. Radiotoxicity R accumulated in storage and transmutation facilities with ADS-800, VVER-1000, BN-800, SUPER-PHENIX-1200, and BREST-1000 type reactors at feed by actinides extracted from SUPER-PHENIX-1200 type reactor

Table II. Transmutation of minor actinides extracted from VVER-1000 type reactor

Reactor-transmuter	VVER-1000	BN-600	SPX-1200	BREST-1000	ADS-800
Total neutron flux density, n/cm^2s	$2.4 \cdot 10^{14}$	$3.0 \cdot 10^{15}$	$3.0 \cdot 10^{15}$	$2.0 \cdot 10^{15}$	$2.6 \cdot 10^{15}$
Thermal neutron flux density, n/cm^2s	$2.8 \cdot 10^{13}$	$1.4 \cdot 10^{6}$	$4.9 \cdot 10^{5}$	$3.8 \cdot 10^{5}$	$2.0 \cdot 10^{15}$
Time of the transition to equilibrium, years	30	60	60	60	1
Equilibrium mass of MA, kg	300	460	460	670	17
Time period τ, years	70	80	80	110	10

Table III. Transmutation of minor actinides extracted from SPX-1200 type reactor

Reactor-transmuter	VVER-1000	BN-600	SPX-1200	BREST-1000	ADS-800
Total neutron flux density, n/cm^2s	$2.4 \cdot 10^{14}$	$3.0 \cdot 10^{15}$	$3.0 \cdot 10^{15}$	$2.0 \cdot 10^{15}$	$2.6 \cdot 10^{15}$
Thermal neutron flux density, n/cm^2s	$2.8 \cdot 10^{13}$	$1.4 \cdot 10^{6}$	$4.9 \cdot 10^{5}$	$3.8 \cdot 10^{5}$	$2.0 \cdot 10^{15}$
Time of the transition to equilibrium, years	30	45	45	70	1
Equilibrium mass of MA, kg	690	1200	1200	1750	53
Time period τ, years	55	60	60	75	12

The obtained results are the evidence of the following.

1. Accumulation of the radiotoxicity in a reactor in the process of transmutation takes place mainly due to transformation of the long-lived isotopes ^{237}Np, ^{241}Am and ^{243}Am into comparatively short-lived alpha-radiators: ^{238}Pu, ^{242}Cm, ^{244}Cm as a result of the reactions:

$$^{237}\text{Np}+n \rightarrow {}^{238}\text{Np}\ (\beta,\ 2{,}12\ \text{days})\ {}^{238}\text{Pu}\ (\beta,\ 87\ \text{years})\ldots,$$

$$^{241}\text{Am}+n \rightarrow {}^{242}\text{Am}^{g}\ (\beta,\ 16\ \text{hours})\ {}^{242}\text{Cm}\ (\beta,\ 161\ \text{days})\ {}^{238}\text{Pu}\ldots,$$

$$^{243}\text{Am}+n \rightarrow {}^{244}\text{Am}\ (\beta,\ 10\ \text{hours})\ {}^{244}\text{Cm}\ (\beta,\ 18{,}1\ \text{years})\ldots$$

During the first 10-20 years after beginning of transmutation, the rate of accumulation of radiotoxicity in all fast reactors is approximately equal (in the thermal reactors it is some higher) and is essentially higher than the rate of accumulation in a long-term storage facility without transmutation.

2. The equilibrium radiotoxicity essentially depends upon the type of the reactor-transmuter and in some rate less upon the composition of feed. The lowest equilibrium radiotoxicity is reached in the facility ADS-800. It is by 4 - 5 times less than in the reactors-transmuters VVER-1000, BN-600, SPX-1200, BREST-1000 in the case of feed with minor actinides extracted from the reactors of VVER-1000 type. In the case of feed from the reactors of SPX-1200 type, the equilibrium radiotoxicity in the ADS-800 is by 6 - 8 times less than in other reactors-transmuters. The highest level of the equilibrium radotoxicity is reached in the reactor of BREST-1000 type.

3. Comparison of formation of the radiotoxicity in the reactors-transmuters and of accumulation of radiotoxicity in the storage facilities (the period of time τ) shows that accumulation in the storage facilities is safer during the first 60 - 110 years than in the reactors of the VVER, BN-600, SPX, and BREST-1000 types. The reactors-transmuters turn out to be more favorable than accumulation in storage facility only after 60 - 100 years after beginning of transmutation. This period of time for the high flux facility ADS-800 is much less, it is only 10-12 years.

4. Long operation of the reactors-transmuters with solid fuel is possible only with repeated refueling and processing of the spent nuclear fuel. That is why the period of time τ may be essentially longer than the data obtained with the aid of the model of continuous irradiation. At the same time, multiple chemical reprocessings increase the potential risk of contamination of the environments during the process of transmutation [8].

5. The values of the parameters presented in the tables II and III are the evidence of the fact that the process of transmutation is the least effective in the reactors of the type BREST-1000 because of relatively low fuel power density. This property is organically inherent to similar facilities.

6. The equilibrium mass and radiotoxicity of minor actinides (including plutonium formed in them) in the reactors-transmuters reach the values of similar orders (or higher) as in the loaded plutonium and its radiotoxicity in the core of a fast reactor of electric power 1 GW. This shows that the potential risk of possible contamination of the environments by minor actinides in the process of transmutation is comparable with the risk of contamination by plutonium loaded in the cores of fast reactors.

7. The process of transmutation is the most effective in the subcritical facilities of the type ADS-800. Because of use of liquid fuel, high flux of thermal neutrons, and continuous removal of the fission products such facility may operate in the mode of equilibrium with relatively low content of long-lived radiotoxicity in the blanket.

CONCLUSIONS

The presented above results of our calculations allow to give the answer to the question put on at the title of the article. This answer is negative. Transmutation of long-lived radiotoxicity in the fast reactors is not effective in comparison with the analogous processes in other facilities. A subcritical facility with liquid fuel (molten salts) with high flux of thermal neutrons and continuous mode of operation seems most effective for transmutation.

Specialists in nuclear technologies, who propose the atomic power industry as the large scale and long-term method of production of energy, must develop and substantiate effective methods of extermination of the harmful wastes of this industry. Transmutation in the reactors is an alternative to long-term storage of the radionuclides in serviced and guarded buildings with stabilized temperature and pressure. Placing of the most dangerous radionuclides in the reactors-transmuters (high temperatures and pressures) is today uncommensurably much more dangerous in comparison with storing of them in specially equipped buildings. But similar reasons should not be considered as a ground for absence of perspectives of investigations in transmutation. It is not excluded that practical application of transmutation of the radionuclides will be necessary only in the future. However, it is necessary just now to carry out investigations, which would allow to demonstrate a real possibility of incineration of the long-lived radionuclides.

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