RESEARCH OF TRANSMUTATION OF PRODUCTS OF NUCLEAR CYCLE AT THE ELECTRON ACCELERATOR

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The analysis of the use of linear electron accelerators for transmutation of fission products and actinides from nuclear power plants is carried out. The results of irradiation of isotopes ^{90}Sr, ^{241}Am and ChNPP fuel containing materials with the neutron flow from the linac are described. The variant of the test stand for investigation of transmutation of fission products and actinides is proposed on the basis of the powerful linear electron accelerator.

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INTRODUCTION

Research of waste management by the technologies and their practical implementation under optimum safety conditions continues to be active. Together with disposal in geological rocks a promising direction in solving the problems of the waste manipulation is the use of transmutation.

In this connection the new approaches to reducing the waste recycling are developed. As is known at low concentration of neutrons in the reactor the operating time of isotopes of the higher nuclear weight in the first approximation equal to [1]:

$$N_A=((\sigma_{A-1}\cdot N_o\cdot n\cdot v)/(n\cdot v(\sigma_{A-1}+\sigma_f)+\lambda_A))\times$$

$$\times(1-\exp(-n\cdot v(\sigma_{A-1}+\sigma_f)-\lambda_A)\cdot t)$$

N_o, N_A are the content of nuclei with atomic number A–1 and A, σ_{A-1}, σ_f are the cross-section of neutron-capture and fission for nuclei with atomic numbers A-1 and A, respectively, n is concentration of neutrons, v is average neutron speed, λ_A is decay constant of nuclei with atomic number A.

Table 1. Equilibrium distribution of transuranium elements at high dozes in the reactor on thermal and fast neutrons (on the left) and the charge of neutrons Dj on division (Dj < 0 means the excess of own manufacture n, Dj > 0 means the necessity of an external source of neutrons)

Isotope	Thermal n spectrum	Fast n spectrum
^{237}Np	5.51	0.75
^{238}Pu	4.17	0.89
^{239}Pu	23.03	66.75
^{240}Pu	10.49	24.48
^{241}Am	0.54	0.97
^{241}Pu	9.48	2.98
^{242}Cm	0.18	0.40
^{242m}Am	0.02	0.07
^{242}Pu	3.89	1.8
^{243}Am	8.11	0.44
^{243}Cm	0.02	0.03
^{244}Cm	17.85	0.28
^{245}Cm	1.27	0.07
^{246}Cm	11.71	0.03
^{247}Cm	0.75	2.E-3
^{248}Cm	2.77	6.E-4
^{249}Bk	0.05	1.E-5
^{250}Cf	0.03	7.E-6
^{251}Cf	0.02	9.E-7
^{252}Cf	0.08	4.E-8
Total	100.0	100.0

Values D_j (neutron consumption per fission)				
Isotope (or fuel type)	10^{15} n/cm^2 fast n	10^{14} n/cm^2, n of PWR	10^{16} n/cm^2, n of PWR	10^{16} n/cm^2, highly thermalized
Th with extraction of ^{238}Pa	-0.39	-0.24	-0.24	-0.27
Th without extraction of ^{238}Pa	-0.38	0.20	1.22	1.14
^{238}U	-0.62	0.07	0.05	0.1
^{238}Pu	-1.36	0.17	0.042	-0.13
^{239}Pu	-1.46	-0.67	-0.79	-1.07
^{240}Pu	-0.96	0.44	0.085	0.14
^{241}Pu	-1.24	-0.56	-0.91	-0.86
^{242}Pu	-0.44	1.76	1.10	1.12
^{237}Np	-0.59	1.12	0.53	-0.463
^{241}Am	-0.62	1.12	0.076	-0.54
^{243}Am	-0.60	0.82	0.16	0.21
^{244}Cm	-1.39	-0.15	-0.53	-0.48
^{245}Cm	-2.51	-1.48	-1.46	-1.37
D_{TRU} (discharge from a PWR)	-1.17	-0.05	-0.35	-0.54
$D_{TRU+Pu+Np}$ (discharge from a PWR)	-0.70	1.1	0.3	0.4

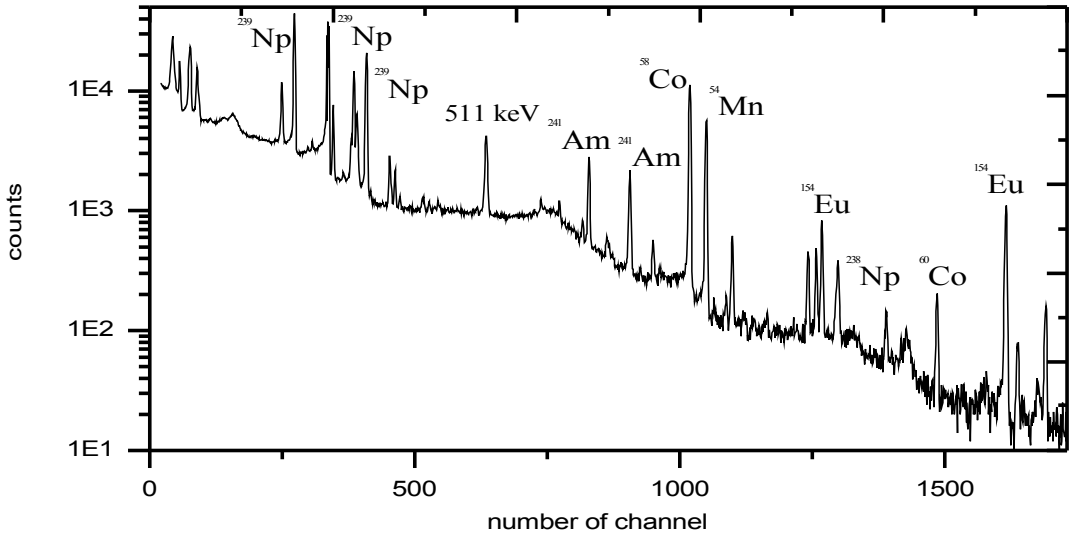
As is seen, during the reactor operating time the content of elements with a large A decreases with neutron concentration increasing. Therefore the increase of neutron flow density in the reactor results in the change of the actinide content in the reactor.

The approach ADS (Accelerator Driven Systems) has many supporters in the world. The main advantage of ADS is the significant decrease of fission products and TRU, and also burning out of new transuraniums. The second feature of ADS is repeated recirculation of

fuel or waste. It allows one to essentially reduce the danger of the spent fuel from usual nuclear power plants after updating with the help of ADS (tabl.1 (on the left)).

From table 1 (on the right) it can be seen that for fast neutrons the flow 10^{15} n/s·sm^2 practically completely results in burning out of transuraniums. Their radiotoxicity is prevalent. From table 2 it is seen that asymptotic spectra lead to isotopes with A>238 with the result being more acceptable by toxicity. Therefore, third advantage of ADS systems is the fast neutron spectrum. As is seen from the given tables during operating time the low level of transuranium elements and fission products in reactors with a flow 10^{15} n/s sm^2 on fast neutrons is attained. The projects of use of powerful linear proton accelerators [2] and deuteron accelerators [3] are known. The use of linear electron accelerators for transmutation is promising [4].

The plutonium isotopes contribute for about 90% to the radiotoxic inventory, the rest 10% is contributed by americium and curium. In the high-level waste arising from reprocessing activities, during which 99.88% of the plutonium and uranium are recovered, americium and curium are the main contributors. To reach a considerable reduction of the radiotoxicity, at least 90% of the americium is need to be fissioned. This can be achieved by multiple recycling in fast neutron flux or once-via transmutation in high-thermal or epidermal neutron flux.



Spectrum of gamma radiation from the sample ^{241}Am after irradiation with neutrons for one year

RESULTS AND DISCUSSION

Samples of ^{241}Am, ^{90}Sr and FCM 4 block ChNPP were investigated. The irradiation of samples was carried out by neutrons from the aluminium-silicon converter during year [5]. The irradiation time was 2·10^7 s. Through the target 4·10^{14} neutrons penetrated. The estimated neutron fluence was 5·10^{18} neutrons/cm^2. The instant flow of neutrons is about 10^{13} n/s cm^2. In the sample ^{90}Sr the activity ^{89}Sr caused by the nuclear reaction ^{90}Sr(n,2n)^{89}Sr was detected. The low yield of radiation with an energy of 1210 keV did not allowed to measure the content of ^{91}Sr → ^{91}Y (reaction ^{90}Sr(n,γ)^{91}Sr, I_{res} = 0,3 b). The spectrum of gamma radiation from the sample ^{241}Am is given in Fig. In the sample ^{241}Am the activity of isotopes $^{238, 239}$Np, ^{233}Pa, $^{241, 243}$Am is detected. The radiation of fission products of ^{103}Ru, ^{154}Eu, ^{95}Nb and isotopes $^{57, 60}$Co, ^{59}Fe, ^{51}Cr caused by the activity of a material of the container is detected. Besides the traces of isotopes of americium, the isotopes of europium-154 were observed.

The feature of irradiation of samples from FCM 4 block ChNPP during year is the absence of fission products of ^{103}Ru, ^{95}Nb.

The powerful linear electron accelerator (P=20 kw in the beam) can be the tool for the study of transmutation of fission products and actinides. 2 10^{13} n/s from the tantalum converter when using the water moderator will have a maximum of distribution of a neutron flow nv at a distance from the centre of a beam of about 7.6 cm. With the use of the moderator with low cross-section of capture (D_2O) the fast neutron flow (≈ 1…5 eV) in a maximum will have 10^{15} n/s·cm^2, that allows one to solve the majority of research tasks on the management with the spent fuel of nuclear power plants.

Table 2. Radiotoxicity of isotopes (CD=Cancer Dose Hazard)

Isotope	Toxicity factor CD/Ci	Half-life, years	Toxicity factor CD/g
^{210}Pb	455	22.3	34800
^{223}Ra	15.6	0.03	799000
^{226}Ra	36.3	$1.6\cdot10^{3}$	35.9
^{227}Ac	1185	21.8	85800
^{229}Th	127.3	$7.3\cdot10^{3}$	27.2
^{230}Th	19.1	$7.54\cdot10^{4}$	0.394
^{231}Pa	372	$3.28\cdot10^{4}$	0.176
$^{2\,34}$U	7.59	$2.46\cdot10^{5}$	0.047
^{235}U	7.23	$7.04\cdot10^{8}$	$1.56\cdot10^{-5}$
^{236}U	7.5	$2.34\cdot10^{7}$	$4.85\cdot10^{-4}$
^{238}U	6.97	$4.47\cdot10^{9}$	$2.34\cdot10^{-6}$
^{237}Np	197.2	$2.14\cdot10^{6}$	0.139
^{238}Pu	246.1	87.7	4220
^{239}Pu	267.5	$2.41\cdot10^{4}$	16.6
^{240}Pu	267.5	$6.56\cdot10^{3}$	60.8
^{242}Pu	267.5	$3.75\cdot10^{5}$	1.65
^{241}Am	272.9	433	936
^{242m}Am	267.5	141	28000
^{243}Am	272.9	7370	54.5

Isotope	Toxicity factor CD/Ci	Half-life, years	Toxicity factor CD/g
^{242}Cm	6.9	0.45	22900
^{243}Cm	196.9	29.1	9960
^{244}Cm	163	18.1	13200
^{245}Cm	284	8500	48.8
^{246}Cm	284	4800	86.7
	Shot-lived	Fission	Products
^{90}Sr	16.7	29.1	2280
^{90}Y	0.6	0.0073	326000
^{137}Cs	5.77	30.2	499
	Long-lived	Fission	Products
^{99}Tc	0.17	$2.13\cdot10^{5}$	$2.28\cdot10^{-3}$
^{129}I	64.8	$1.57\cdot10^{7}$	0.0115
^{93}Zr	0.095	$1.5\cdot10^{6}$	$2.44\cdot10^{-4}$
^{135}Cs	0.84	$2.3\cdot10^{6}$	$9.68\cdot10^{-4}$
^{14}C	0.2	$5.93\cdot10^{3}$	0.892
^{59}Ni	0.08	$7.6\cdot10^{4}$	$6.38\cdot10^{-3}$
^{63}Ni	0.03	100	1.7
^{126}Sn	1.7	$1\cdot10^{5}$	0.0483

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ИССЛЕДОВАНИЕ ТРАНСМУТАЦИИ ПРОДУКТОВ ЯДЕРНОГО ЦИКЛА НА УСКОРИТЕЛЕ ЭЛЕКТРОНОВ

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Проведен анализ использования линейных ускорителей электронов для трансмутации продуктов деления и актиноидов атомных электростанций. Описаны результаты облучения потоком нейтронов на ЛУЭ изотопов ^{90}Sr, ^{241}Am и топливосодержащих масс ЧАЭС. Предложен вариант стенда для исследования трансмутации продуктов деления и актиноидов на базе мощного линейного ускорителя электронов.

ДОСЛІДЖЕННЯ ТРАНСМУТАЦІЇ ПРОДУКТІВ ЯДЕРНОГО ЦИКЛУ НА ПРИСКОРЮВАЧІ ЕЛЕКТРОНІВ

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Проведено аналіз використання лінійних прискорювачів електронів для трансмутації продуктів ділення й актиноїдів атомних електростанцій. Описано результати опромінення потоком нейтронів на ЛПЕ ізотопів ^{90}Sr, ^{241}Am і паливовміщуючих мас ЧАЕС. Запропоновано варіант стенда для дослідження трансмутації продуктів ділення й актиноїдів на базі потужного лінійного прискорювача електронів.