PHYSICAL GROUND FOR RADIOACTIVE WASTE TRANSMUTATION FACILITY USING THERMONUCLEAR NEUTRONS

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Two variants of radioactive waste transformation system using thermonuclear neutrons were considered. In one case transuranium elements and fission products are irradiated together and in second one they are irradiated separately. Advantages and disadvantages of each case were analyzed. Second variant was chosen. Physical basing of facility construction for radioactive waste elements transmutation using thermonuclear neutrons for such variant was developed.

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1. INTRODUCTION

By 2010 year from the world nuclear reactors fleet with total power more then 400 GWt will produce more then 300 thousand tons of spent fuel. Composition and amount of spent nuclear fuel from Ukraine reactors is shown in Table 1. In Russian Federation amount of spent fuel by 2003 is equal to 8740 tons (17.39×10^{19} Bq). Composition data and main properties of long-lived activity fission products of LWR reactor after 40 years life time period are represented in Table 2.

Table 1. *Composition and amount of radioactive waste of Ukrainian NPP*

Element	Content, %	Amount per year, tons	Amount per 40 years, tons
Uranium	94.771	264.536	10581.420
Neptunium	0.059	0.165	6.165
Pluthonium	0.951	2.663	106.512
Americium	0.092	0.257	10.304
Curium	0.00217	0.006	0.243
Fission products	4.124	11.547	461.888

Table 2. *Composition and properties of LWR fission products after 40-yr life-time*

Element	Initial mass,kg	Half-life period, yr	I_{res}, Barn	Daughter nucleus	Half-life period, yr	Final element	Activity,curie	Disposal volume, m^3
^{99}Tc	843	2.11×10^5	310	^{100}Tc	15 s	^{100}Ru	14455	48181
^{129}I	196.0	1.57×10^7	26.5	^{130}I	12.36 h	^{130}Xe	34.7	4.327
^{127}I	59.4	Stab	149	^{128}I	25 min	^{128}Xe	34.7	4.327
^{93}Zr	810.4	1.53×10^6	15.2	^{94}Zr	Stab	^{94}Zr	2040	583
^{90}Zr	257.8	Stab	0.17	^{91}Zr	Stab	^{91}Zr	2040	583
^{91}Zr	670.4	Stab	6.8	^{92}Zr	Stab	^{92}Zr	2040	583
^{92}Zr	724.6	Stab	0.68	^{93}Zr	1.5×10^6 yr	^{94}Zr	2040	583
^{94}Zr	838.4	Stab	15.4	^{95}Zr	64.02 days	^{95}Mo	2040	583
^{96}Zr	896.8	Stab	5.8	^{97}Zr	16.9 hours	^{97}Mo	2040	583
^{135}Cs	442.2	2.3×10^6	60.2	^{136}Cs	13.2 d	^{136}Ba	510.1	510
^{133}Cs	1228	Stab	393	^{134}Cs	2.06 yr	^{134}Ba	510.1	510
^{137}Cs	832.2	30.1	0.616	^{138}Cs	32.2 m	^{138}Ba	510.1	510
^{126}Sn	29.48	1.0×10^5	0.139	^{127}Sn	2.1 h	^{127}I	838.1	239
^{79}Se	6.57	6.5×10^5	56	^{80}Se	Stab	^{80}Se	458.6	131

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At present time high intensity sources of thermal neutrons by TD reactions are developed in Russia, France and other countries. Such sources can be used for transmutation of the transuranium elements (TRE) or fission products (FP). Transuranium elements and fission products have long-life half-time periods (more then 10^5 years). Thus nuclear-waste disposal requires transmutation of this kind of waste into short-lived or even stable elements. Transmutation problem is very actual for Ukraine [1]-[7].

In our paper physical backgrounds for long-lived nuclear waste transmutation facility are considered. Analysis of cross-section data for physical processes at various neutron energies from different International Databases is carried out. Results of neutron transition modelling through different materials are also discussed.

2. TRANSMUTATION OF TRANSURANIUM ELEMENTS

Transmutation of transuranium elements results from fission. Dependencies of fission cross-section from neutron energy for plutonium and corresponding actinides (^{237}Np, ^{241}Am, ^{244}Cm) are shown in Fig. 1.

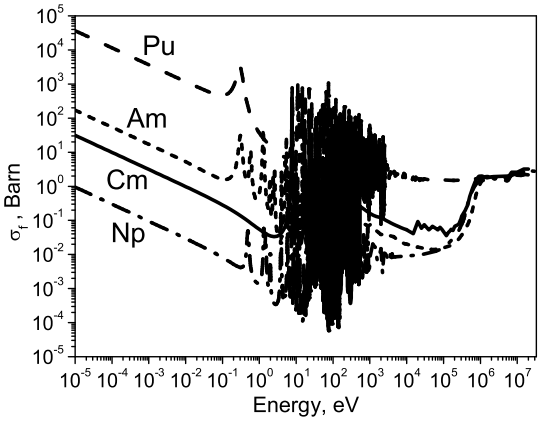

Fig.1. *Fission Cross-Section σ_f for Plutonium (^{239}Pu) and radioactive daughters (^{237}Np, ^{241}Am, ^{244}Cm) versus neutron energy*

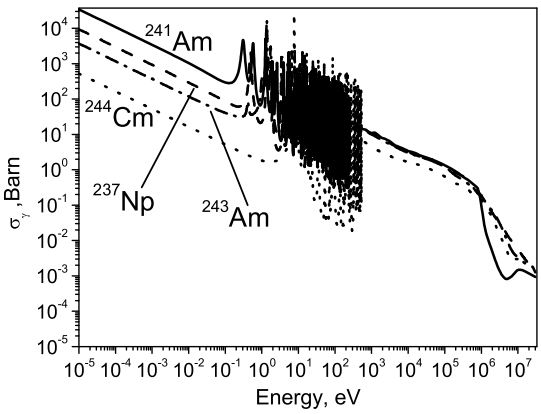

Fig.2. *Capture Cross-Section for radioactive daughters*

Capture cross-sections are shown in Fig. 2. These figures are based on Brookhaven National Laboratory database [8] and on IAEA database [9]. From Fig. 1 and Fig. 2 follows that fission cross-sections are significant in neutron energy range 1-15MeV and they decrease rapidly for neutron energy of few keV. For lower energies cross-sections increase extremely due resonance neutron capture. Thus TE transmutation into other isotopes is possible by means of fast neutron irradiation. 14 MeV thermonuclear neutrons obtained from deuterium-tritium reaction could be used for such transmutation process. In other hand transuranium elements can be transmutated into short-lived or stable isotopes by irradiation of neutrons in resonance energy range.

3.TRANSMUTATION OF THE FISSION PRODUCTS

FP constitute a significant part in nuclear waste. As they have a long half-life periods as it is shown in Table 1 and Table 2 transmutation is necessary. Disposal of FP in geological formation is't optimal because of enormous disposal volumes. At the same time neutron irradiation provides means of FP conversion into other isotopes. For example technetium has one isotope and could be easy extracted for transmutation process. ^{129}I isotope transmutation could be carried out without isotopic separation, though impurity of ^{127}I isotope increases neutron absorbtion to 23% compared to pure isotope ^{129}I. Transmutation of ^{93}Zr is possible but due to high expenses and low ecological loading this process is still doubtful. Isotopic separation of ^{135}Cs is embarrassed by radiation hazards. Transmutation in this case is possible after waste radiation lowering down to recycling or hand on levels. FP transmutation into short-lived or stable isotopes is possible only through (n,γ) reaction. ^{99}Tc, ^{127}I, ^{135}Cs nuclei cross-sections energy dependencies are shown in Fig.3. It uses data from Brookhaven National Laboratory database [8] and from IAEA database [9].

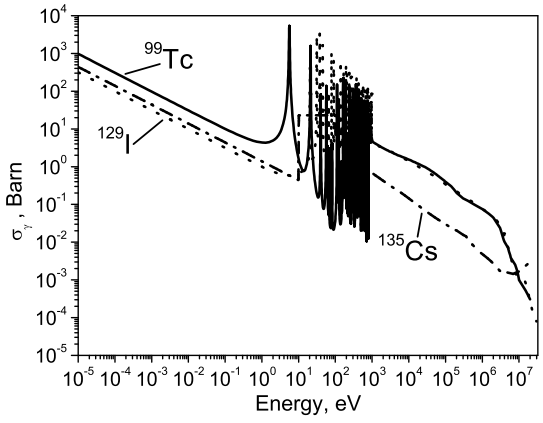

Fig.3. *Capture Cross-section for ^{99}Tc,^{127}I,^{135}Cs versus neutron energy*

These dependencies reveal negligible FP transmutation when $14\,MeV$ neutrons are used. This process is possible only after neutron moderation down to $10\,...\,10000\,eV$ energy range. In this range resonance capturing occurs. Transmutation by means of Adiabatic Resonance Crossing (ARC) method give considerably increases efficiency of neutron capture. In this case probability of neutron capture is proportional to resonance integral: $I_{res}(E_{min}, E_{max}) = \int \sigma(n,\gamma)E^{-1}dE$. Values of resonance integrals for some nuclei are shown in Table 2. ^{99}Tc resonance integral is 310 b while cross-section at thermal/epithermal neutron energies (En $\leq$ 1 eV) is only of the order of 20 b. Neutron capture on ^{99}Tc ($t_{1/2}$ = 2.11 $\times 10^3$ yr) produces ^{100}Tc ($t_{1/2}$ = 15.8 s) with then decays to stable ^{100}Ru isotope. Thus, the radiotoxicity can be eliminated in a single neutron capture and, since ^{100}Ru has small neutron capture cross-section and both ^{101}Ru and ^{102}Ru are essentially stable, no new radioactive elements are produced. The similar processes take place for ^{135}Cs and ^{129}I isotopes. Neutron capture in ^{129}I ($t_{1/2}$ = 1.57×10^5 yr) produces ^{130}I ($t_{1/2}$ = 12 h) decaying to stable ^{130}Xe isotope. And neutron capture on ^{135}Cs ($t_{1/2}$ = 2.3 $\times 10^6$ yr) produces ^{136}Cs ($t_{1/2}$ = 13 d) decaying to ^{136}Ba. Therefore ARC is most efficient for elements with strong capture resonances, such as ^{99}Tc, ^{135}Cs and ^{129}I constituting 95% of the total LLFF(long-lived fission fragment) radiotoxicity inventory.

Large capture cross-section of the given element could be achieved converted by an efficient use of resonances. This is possible due very small lethargic steps of neutrons down in lead. Neutrons have a peculiar behavior in lead:

1. a small average lethargy ξ due to the high atomic mass of lead:

$$\xi \equiv 1 + \frac{\alpha}{1-\alpha} ln(\alpha)\,,$$

where

$$\alpha \equiv \frac{(m_{Pb} - m_n)^2}{(m_{Pb} + m_n)^2}\,;$$

2. a high and nearly energy-independent elastic scattering cross-section;

3. a long 'storage'time because, below the capture resonances (En $\leq$1 keV) and down to epithermal energies, the elastic scattering process is nearly isotropic and the transparency to neutrons is very high (it takes about 3 ms, 1800 scatterings and a path in lead of 60 m to thermalize a 1 MeV neutron).

The corresponding neutron spectra is presented in Fig.4.

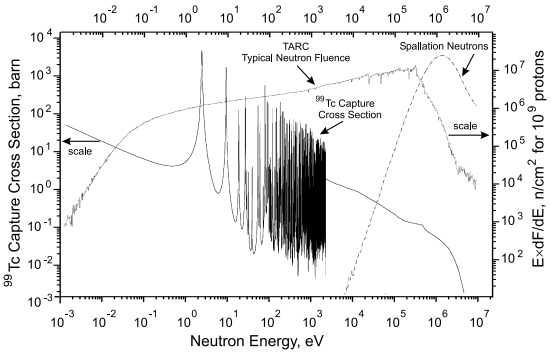

Fig.4. *^{99}Tc neutron capture cross-section (JENDL-3.2 database [9]), as a function of neutron energy (left-hand scale); typical neutron fluence energy distribution in TARC [3], as a function of neutron energy in isolethargic bins, for 3.5 GeV/c protons (right-hand scale). Energy distribution of neutrons from the spallation process is shown in arbitrary units*

Spectra for neutrons (Fig.4.) produced by spallation at relatively high energy (few MeV), after having been quickly moderated by (n,xn), (n,n') reactions down to energies of a few hundred keV, will slow down quasiadiabatically with small isolethargic steps and reach the capture resonance energy of an element to be transmuted where it will have a high probability of being captured. The resonance width is usually larger than the average lethargic step. Typical example is ^{99}Tc where strong neutron capture resonance at $5.6\,eV$ (4000 b) (see Fig.4) covers four average lethargy steps.

4. TRANSMUTATION FACILITY ANALYSIS AND SIMULATION

For our simulation of neutron transport through material we have used cross-section data bases [8],[9] to consider elastic and inelastic collision processes. Corresponding cross-sections are presented in Fig.5.

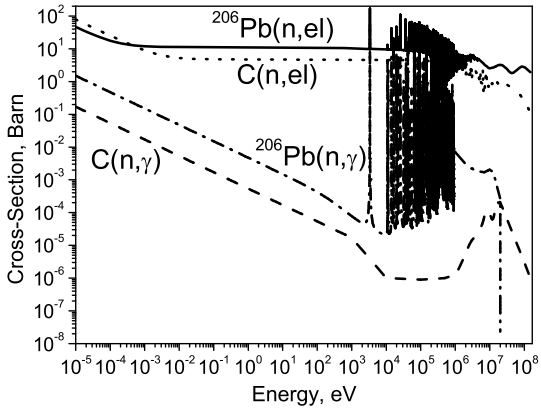

Fig.5. *Cross-Sections for ^{208}Pb and ^{12}C versus neutron energy*

This figure shows ^{12}C to act as neutron moderator dye to elastic collisions. So for initial $14\,MeV$ neutrons interacting with lead or carbon we have

obtained spectra in wide range from few eV to few MeV. This kind of spectra fits requirements for transmutation process for FP and TRE. In present article we considers two variants of transmutation facilities construction. First one uses neutrons from external source irradiating mixture of TRE and FP. In the second one neutrons from external neutron irradiate TRE and secondary neutrons irradiate FP. In the first case TRE and FP mixture can be placed into container filled with lead or with melted lead for better wettability of the mixture. Neutron spectra obtained by Monte-Carlo simulation from lead target irradiated by $14\,MeV$ neutrons is presented in Fig.6. It follows when lead target thickness increases from 20 to $120\,cm$ corresponding neutron spectra undergo significant modification. The high energy part of spectra decreases while low energy (resonance range) part increases.

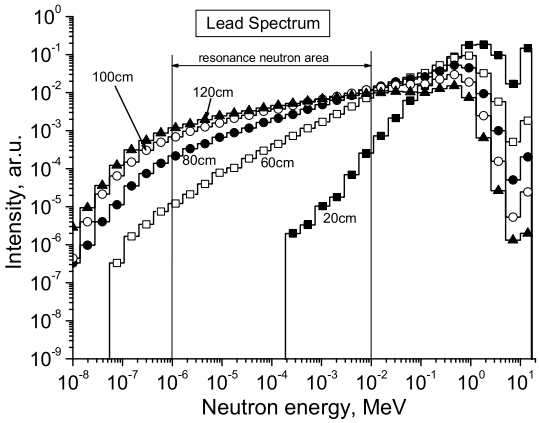

Fig.6. *Neutron spectra for lead layers*

During irradiation of mixture of TRE and FP by fast ($\geq 1\,MeV$) neutrons fission is dominant as TRE and FP capture cross-section is small in this energy range. Next neutron slows down to the resonance energy range. Both TRE fission and FP transmutation process are dominant.

Let us consider a case with TRE and FP placed separately. Effectiveness of such separation could be readily understood from Fig.7. Here ^{241}Am fission cross-section and ^{99}Tc capture cross-section are presented. It follows for fast neutron FP placement in TRE area is ineffective. In the other hand for resonance neutron TRE placement in FP area is also ineffective.TRE can be placed into carbon-carbon matrix. In this case we have more resistant compound with additional helium cooling required due to additional heat deposition by fission. We have provided some Monte Carlo simulation intended to obtain detail information about carbon thicknesses appropriate in this case. For simulation of TRE and FP placed in carbon matrix density of this matrix set up to 10% of usual carbon. Simulated neutron flux after transition through carbon for different thicknesses is shown in Fig.8.

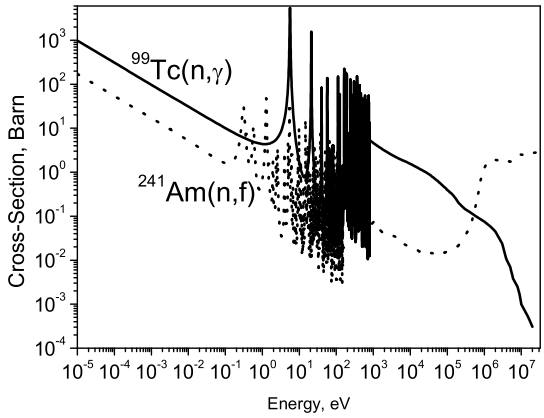

Fig.7. *Fission cross-section of ^{241}Am and capture cross-section of ^{99}Tc versus neutron energy*

This figure shows that increase of carbon thickness decreases neutron energy. For 10 cm carbon thickness almost all neutrons are fast. Therefore transmutation of TRE is a result of fission. For resonance neutrons TRE transmutation results from both fission and capture processes.

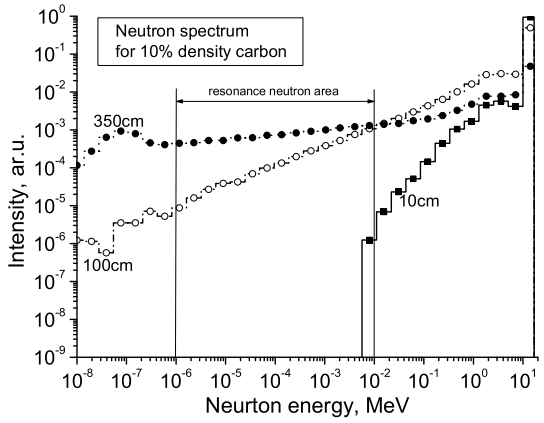

Fig.8. *Neutron spectra for 10% carbon*

The transmutation system design with separated TRE and FP placement has an advantage because between two cells with TRE and FP we can set up cell with Lithium. Such cell can contain ^{6}Li and ^{7}Li isotopes in the form of Lithium oxide. As first cell contains TRE fast neutron flux is necessary for it. Hence carbon thickness for this cell can be about 10 cm. Here we consider transmutation facility with $14\,MeV$ initial neutron flux. It is necessary to develop optimal construction for TRE transmutation in the first cell, FP transmutation in the second one, and also with optimal Beryllium and Lithium usage. For TRE placed in a 10 cm cell with 10% carbon matrix base neutron flux is formed by fast neutrons. So such elements as Am, Cm, Np are subjected to transmutation through fission with cross-section about $4\,...\,6\,barns$. This transmutation process goes

with relatively high intensity. A neutron spectra from such cell is shifted to low energies due to fission neutrons. Moreover we expect some increase of neutron flux due to fission multiplication.

For our model we setup TRE as a mixture of Am, Cm, and Np. It corresponds to irradiated VVER-1000 fuel containing ^{237}Np 44.5%, ^{241}Am 48.6%, and ^{243}Am 6.9% after a 10 yr holding period. [10] Cell has a 10 cm thickness and compsises 10% carbon matrix, 10% TRE and 80% helium. Comparison of neutron spectrum after such cell with TRE and with lead is shown in Fig.9. Lead has atomic number close to that of TRE and elastic cross-sections are also close to each other. Therefore we have obtained difference between neutron spectra of this cell due to fission activity. The ratio of initial transmitted neutron flux to initial flux is 1.12. Addition of 20% TRE increases ratio up to 1.36 i.e. almost to 40%. Additional neutron flux makes allows to setup beryllium cell after the TRE cell. In this cell additional growth of neutron flux due to (n,2n) reaction is expected.

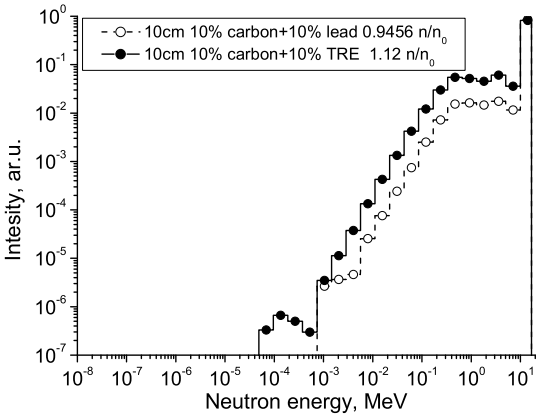

Fig.9. *Neutron spectra for carbon-lead and for carbon-TRE cells*

Cross-section of Be(n,2n) reaction is shown in Fig.10. It follows that such reaction has high intensity only in neutron energy range above 5 MeV.

We expect that the thickness of Beryllium layer will be about 1 ... 2 cm due to high neutron remission in thick layers. Neutrons with such spectra are appropriate for Tritium production. This can be attained placing Lithium cell filled with Lithium oxide after beryllium cell. Due to reaction we have Tritium production. It is necessary to mention that natural Lithium consists of 92.5% ^{7}Li and 7.5% ^{6}Li. Cross-sections for ^{7}Li and ^{6}Li isotopes are presented in Fig.10. This cross-section has a maximum in energy range about 250 keV therefore neutrons after Beryllium cell are appropriate for Tritium production. Thickness of such a cell will be about 10 ... 15 cm. After this layer neutron spectrum is still too fast for FP transmutation so addition moderation is necessary. This can be achieved by placement of the pure carbon cell after Li oxide cell.

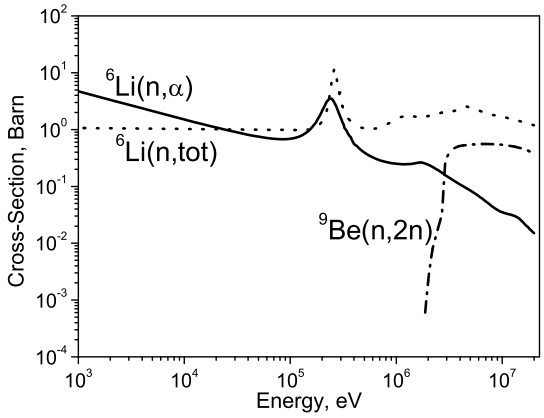

Fig.10. *Cross-sections for ^{7}Li and ^{6}Li isotopes*

Thickness of such cell will be about 15 ... 30 cm The neutron energy distribution obtained by simulation of such cell sequence is shown in Fig.11.

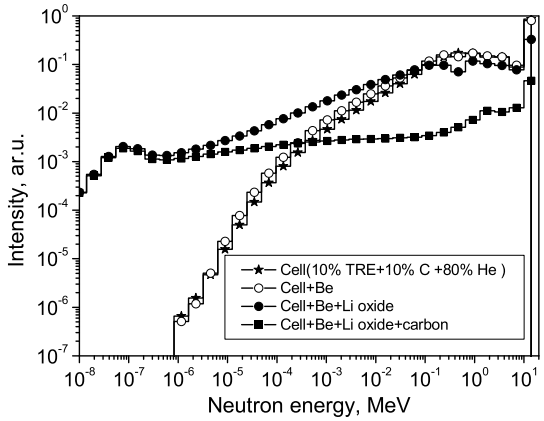

Fig.11. *Neutron spectra after our cells sequence*

Neutron spectra from this cell sequence irradiates cell with carbon matrix and FP. The thickness of this cell will be strongly depended from FP composition. Optimization of volumes and thicknesses of our cell sequence by Monte-Carlo simulation allows detailed design of transmutation facilities and evaluation of transmutation efficiency for TRE and FP. At present moment a lot scientific groups in the world focus efforts on such problems and we hope that this problem will be solved.

References

1. A.V. Bogoslovskiy. Unichtozhenie radioaktivnyh othodov s pomosch'yu bystryh neytronov kak al'ternativa geologicheskomu zahoroneniyu (Istoriya voprosa i proekt K. Rubbia). Obzor // *Atomnaya tehnika za rubezhom.* 1998, v.1, p.3-10 (in Russian).

2. V.P. Dmitrievskiy. Elektroyadernyy metod polucheniya energii // *Physics of Elementary Particles and Atomic Nuclei (PEPAN).* 1997, v.28, p.815-836 (in Russian).

3. S. Andriamonje, et al. *Proposal: Experimental Study of the Phenomenology of Spallation Neutrons in a Large Lead Block.* CERN/SPSLC 95–17, SPSLC/P291, 5 May 1995.

4. V.A. Bronnikov. Novye issledovaniya yadernyh harakteristik nekotoryh produktov deleniya. Obzor // *Atomnaya tehnika za rubezhom.* 2003, v.12, p.3-6 (in Russian).

5. V.A. Mahova, I.D. Sokolova, N.A. Shul'ga. Issledovaniya po frakcionirovaniyu i transmutacii dolgozhivuschih radionuklidov. Obzor //*Atomnaya tehnika za rubezhom.* 2003, v.3, p.3-10 (in Russian).

6. C. Rubbia, S. Buono, Y. Kadi and J.A. Rubio. *Fast neutron incineration in the energy amplifier as alternative to geological storage: the case of Spain.* CERN internal report, CERN/LHC/97–01 (EET), Feb. 1997.

7. S.A. Bznuni, V.S. Barashenkov, V.M. Zhamkochyan, A.N. Sosnin, A. Polyanski, A.G. Hudaverdyan. Perspektivnye elektroyadernye sistemy // *Physics of Elementary Particles and Atomic Nuclei (PEPAN).* 2003, v.34, p.977-1030 (in Russian).

8. http://www.nea.fr/html/dbdata/eva/evaret.cgi

9. http://www.nea.fr/html/dbdata/eva/evaret.cgi

10. B.A. Gabaraev, I.H. Ganev, A.V. Lopatkin, et al. Vliyanie glubiny vygoraniya i vyderzhki obluchennogo yadernogo topliva na otnoshenie massy Am, Cm, Np i delyaschegosya plutoniya // *Atomnaya energiya.* 2004, v.96(2), p.126-132 (in Russian).

ФИЗИЧЕСКОЕ ОБОСНОВАНИЕ КОНСТРУКЦИИ УСТАНОВКИ ДЛЯ ТРАНСМУТАЦИИ ОТДЕЛЬНЫХ ЭЛЕМЕНТОВ РАО С ИСПОЛЬЗОВАНИЕМ ТЕРМОЯДЕРНЫХ НЕЙТРОНОВ

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Рассматривается два варианта системы трансмутации радиоактивных отходов с помощью термоядерных нейтронов. В одном - трансурановые элементы и продукты деления не отделены друг от друга, в другом варианте - облучаются раздельно. Проанализированы преимущества и недостатки каждого из вариантов. Предпочтение отдано второму варианту и для него выполнено физическое обоснование элементов конструкции разрабатываемой установки.

ФІЗИЧНЕ ОБҐРУНТУВАННЯ КОНСТРУКЦІЇ УСТАНОВКИ ДЛЯ ТРАНСМУТАЦІЇ ОКРЕМИХ ЕЛЕМЕНТІВ РАВ З ВИКОРИСТАННЯМ ТЕРМОЯДЕРНИХ НЕЙТРОНІВ

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Розглядається два варіанти системи трансмутації радіоактивних відходів за допомогою термоядерних нейтронів. В одному - трансуранові елементи та продукти ділення не відокремлені один від одного, в іншому варіанті - опромінюються роздільно. Проаналізовано переваги та недоліки кожного з варіантів. Перевага віддана другому варіанту і для нього виконано фізичне обґрунтування елементів конструкції установки, що розробляється.