STUDIES OF THERMONUCLEAR NEUTRON USAGE MEANS FOR RADIOACTIVE WASTE TRANSMUTATION

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Two variants of radwastes transmutation schemes using thermonuclear neutrons are considered. In the first case transuranium elements and fission products are not separated while in the second case these are irradiated separately. Advantages and drawbacks of both cases were analyzed. Simulation of radwastes transmutation systems was performed. Analysis of radwastes transmutation efficiency for all cases was carried out. Physical backgrounds for radwaste transmutation by thermonuclear neutrons were prepared.
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INTRODUCTION

Nowadays intensive thermonuclear neutron sources with above 10^{14} n/s fluxes are developed in a number of countries (France, Russia and etc.). Such neutron fluxes could be used for efficient radwastes transmutation. This is a topical problem because up to 2010 from the world nuclear reactor fleet with total power of 400 GW above 300 thousand tons of spent fuel should be removed. The problem is actual for Ukraine too. During radwastes transmutation before burial transuranium elements (TRU) and fission products (FP) with long half-decay periods (hundreds and thousands years) are to be converted into short-lived or stable isotopes. At present considerable attention is paid to a problem of radwastes transmutation [1].

In the presented paper means of thermonuclear neutron usage for radwastes transmutation were studied employing mathematical simulation methods. Two cases of transmutation systems were considered. In the first case transuranium elements and fission products are not separated and are irradiated together while in the second case these are irradiated separately.

MATHEMATICAL SIMULATION RESULTS

In our work we have studied transmutation of the basic transuranium elements ^{237}Np, ^{241}Am, and ^{244}Cm and fission products ^{99}Tc, ^{127}I, ^{135}Cs by the thermonuclear neutron beam with 10^{15} n/cm^2 flux density. Firstly, we have used database [2] and have obtained fission (nf) and capture (nγ) cross-sections versus neutron energies of the above mentioned isotopes. Neutron energy range from 10^{-5} to 10^{8} eV was considered.

In the Fig. 1 an example dependencies of fission cross-section (nf) for transuranium element ^{241}Am and capture cross-section (nγ) for fission product ^{99}Tc are shown. For other elements cross-sections are similar but differ in magnitude.

The figure reveals sharp difference in magnitude and behavior of cross-section energy dependence for neutron energies above 1 MeV [3]. While transuranium elements undergo intense fission by neutrons with En = 1...15 MeV, almost no transmutation of fission products (^{99}Tc) occur.

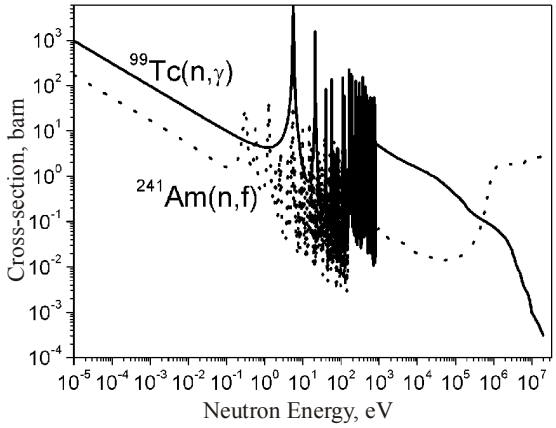

Fig. 1. ^{241}Am fission cross-section (σ_f) and ^{99}Tc capture cross-section (σ_γ) versus neutron energy

This peculiarities lead to two cases of transmutation systems (joined and separated irradiation). From Fig. 1 it is evident that fission products will undergo intense transmutation provided neutron energy decelerate down to E_n = 10…10000 eV where resonance capture occur. In this case capture probability is proportional to resonance integral

$$I_{рез}(E_{min}, E_{max}) = \int_{E_{min}}^{E_{max}} \sigma(ny) E^{-1} dE .$$

Resonance integral for FP nuclei neutron capture is significantly larger than thermonuclear neutrons cross-sections. For instance for ^{99}Tc nucleus I_{res}=300 barn while cross-section is about 20 barn.

As a materials where non-separated TRU+FP materials to be placed we have considered lead and carbon, and for separated TRU and FP only carbon was considered. We have simulated cells with various radwastes components concentration in lead or carbon for neutrons with initial energy of 14 MeV. Cell volume averaged spectra for neutrons perform radioactive isotopes transmutation were calculated. Calculations of isotopes concentration variation in dependence of irradiation time and mode were performed. For the separated irradiation case we have determined parameters of moderator placed between cells where TRU and FP are irradiated separately in carbon matrices. Moderator consists of three layers: 1 cm beryllium, 10 cm lithium

oxide and 20 cm carbon. Material with detailed description of simulation method and results were sent to "The Journal of Kharkov National University" [4]

After additional calculations the most significant results could be presented as following. Firstly let us consider the case of non-separated TRU and FP are placed into two identical cells of 100 cm length and 50 cm diameter. In one cell 20% TRU (^{237}Np – 44.5%, ^{241}Am – 48.6%, ^{243}Am – 6.9%) and 10% FP (^{99}Tc – 57.7%, ^{135}Cs – 28.9%, ^{129}I – 13.4%) are placed into 70% lead matrix, in another cell TRU + FP with the same composition are placed into 70% carbon matrix. The cell's bottoms are targeted by 14 MeV neutron beam 6.9 cm in diameter. Volume averaged neutron spectra are presented in the Fig. 2. From the figure it follows for carbon matrix inside energy range 1 keV…1 MeV a number of neutrons is larger than for lead matrix. This is due to higher TRU fission efficiency in carbon matrix compared to lead matrix.

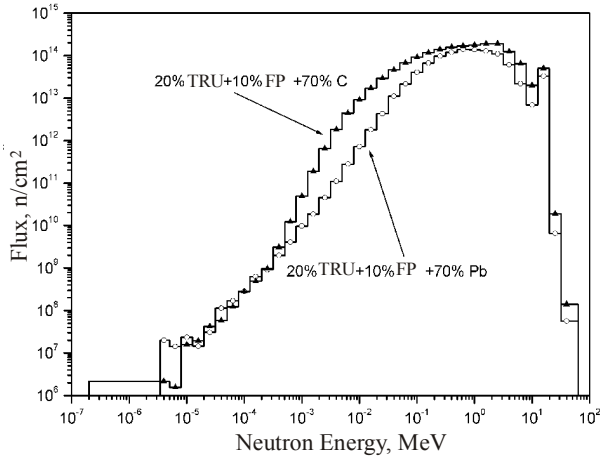

Fig. 2. Neutron spectra for (20% TRU + 10% FP + 70% C) and (20% TRU + 10% FP + 70% Pb). Spectra are averaged over cell volume

Comparison of spectra from Fig. 2 with similar spectra for matrices contenting 100% lead or carbon shows neutron number resulting from (nf) reaction on TRU essentially exceeds number of initial 14 MeV neutrons (more than 9 times for lead).

For transmutation calculations we have used FISPACT code [5]. It provides solution of balance equations using iteration methods and modern cross-sections databases. Concentration change in time for various isotopes, such as minor actinides and FP for different matrices and varying radwastes content were studied.

In the Fig. 3 concentration changes in time are shown for 20% TRU and 10% FP placed inside 70% lead matrix.

Fig. 4 presents ^{99}Tc concentration change versus irradiation time for matrices with different carbon content.

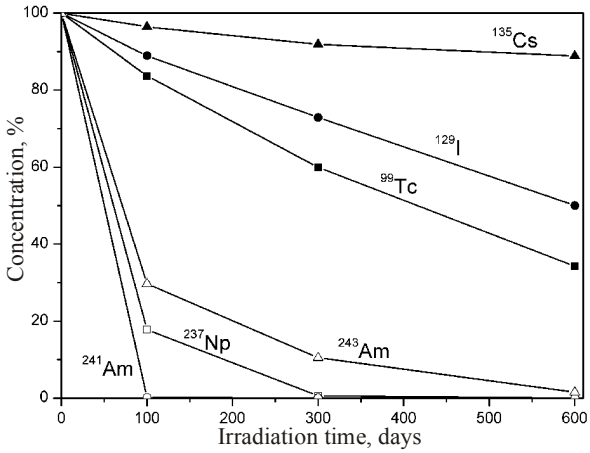

Fig. 3. Isotope concentration versus time

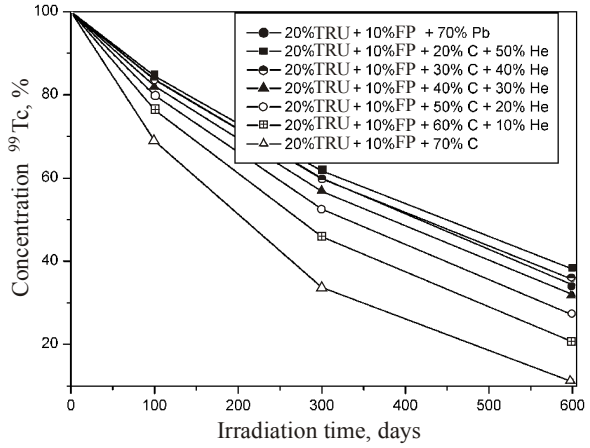

Fig. 4. ^{99}Tc concentration change versus irradiation time for different matrices

Evidently transmutation in carbon matrix (70% C) is more intense than that in lead matrix (70% Pb). Also increase of carbon concentration from 20% to 70% provides more intensive ^{99}Tc transmutation.

Concentration of ^{135}Cs changes in a similar way while for ^{129}I there are some differences. For ^{129}I concentration change 70% lead matrix is same as in 70% carbon matrix. These peculiarities rely on energy dependence of (nγ) reactions for such isotopes.

From Fig. 2 follows for resonance region with neutron energies E_n=10…10000 eV where resonance integrals for FD neutron capture are essential a number of neutrons is small. This impedes FP transmutation. We have calculated averaged neutron spectrum inside cell with FP in carbon matrix for separated TRU and FP irradiation with moderator (Be, LiO and C) placed between cells (Fig. 5).

Obviously a number of neutrons in resonance region increase. One can expect this to be more suitable for FP transmutation but due to neutron absorption in moderator total number of neutrons hit FP cell in carbon decreases by about hundred of times. Hence amount of FP isotopes under transmutation will be lower than that for cell with non-separated TRU+FP. It is clear from Fig. 6 where ^{99}Tc

concentration change versus time is shown for various irradiation conditions.

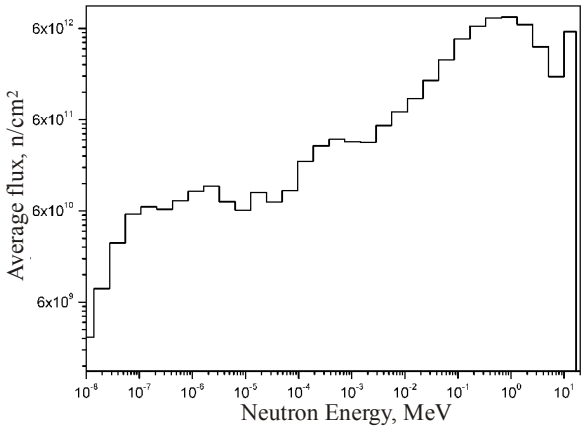

Fig. 5. Neutron spectrum in carbon cell with FD averaged over cell volume

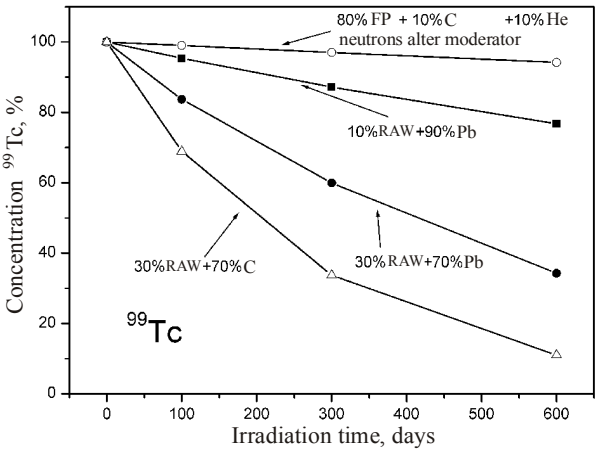

Fig. 6. ^{99}Tc concentration change versus time for various irradiation conditions

Evidently for separated irradiation (80% FP + 10% C+ 10% He) ^{99}Tc undergoes almost no transmutation. The same dependencies were obtained for ^{129}I and ^{135}Cs.

CONCLUSIONS

For the given geometry and using mathematical simulation we have determined optimal conditions for minor actinides and FP transmutation under joined and separated irradiation by 14 MeV neutrons. Further investigations are necessary to find optimal conditions for separated irradiation avoiding strong attenuation of neutron flux.

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ИССЛЕДОВАНИЕ ВОЗМОЖНОСТИ ИСПОЛЬЗОВАНИЯ ТЕРМОЯДЕРНЫХ НЕЙТРОНОВ ДЛЯ ТРАНСМУТАЦИИ РАО

Е.В. Рудычев, Р.П. Слабоспицкий, М.А. Хажмурадов

Рассматривается два варианта системы трансмутации радиоактивных отходов (РАО) 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. Выполнено моделирование систем трансмутации РАО. Проанализирована эффективность трансмутации РАО для каждого из вариантов. Подготовлено физическое обоснование для трансмутации РАО термоядерными нейтронами.

ДОСЛІДЖЕННЯ МОЖЛИВОСТІ ВИКОРИСТАННЯ ТЕРМОЯДЕРНИХ НЕЙТРОНІВ ДЛЯ ТРАНСМУТАЦІЇ РАВ

Є.В. Рудичев, Р.П. Слабоспицький, М.А. Хажмурадов

Розглядається два варіанти системи трансмутації радіоактивних відходів (РАВ) за допомогою термоядерних нейтронів. В одному – трансуранові елементи і продукти ділення не відокремлені один від одного, в іншому варіанті – опромінюються роздільно. Проаналізовано переваги і недоліки кожного з варіантів. Виконано моделювання систем трансмутації РАВ. Проаналізовано ефективність трансмутації РАВ для кожного з варіантів. Підготовлено фізичне обгрунтування для трансмутації РАО термоядерними нейтронами.