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Role of (*n*,2*n*) Reactions in Transmutation of Long-Lived Fission Products

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Abstract—The conditions under which (n,γ) and (n,2n) reactions can help or hinder each other in neutron transmutation of long-lived fission products (LLFPs) are considered. Isotopic and elemental transmutation for the main long-lived fission products, ⁷⁹Se, ⁹³Zr, ⁹⁹Tc, ¹⁰⁷Pd, ¹²⁶Sn, ¹²⁹I, and ¹³⁵Cs, are considered. The effect of (n,2n) reactions on the equilibrium amount of nuclei of the transmuted isotope and the neutron consumption required for the isotope processing is estimated. The aim of the study is to estimate the influence of (n,2n) reactions on efficiency of neutron LLFP transmutation. The code TIME26 and the libraries of evaluated nuclear data ABBN-93, JEF-PC, and JANIS system are applied. The following results are obtained: (1) The effect of (n,2n) reactions on the minimum number of neutrons required for transmutation and the equilibrium amount of LLFP nuclei is estimated. (2) It is demonstrated that, for three LLFP isotopes (¹²⁶Sn, ¹²⁹I, and ¹³⁵Cs), (n,γ) and (n,2n) reactions are partners facilitating neutron transmutation. The strongest effect of (n,2n) reaction is found for ¹²⁶Sn transmutation (reduction of the neutron consumption by 49% and the equilibrium amount of nuclei by 19%).

Keywords: long-lived fission products, neutron transmutation, neutron consumption, equilibrium amount of nuclei

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1. INTRODUCTION

Operation of a nuclear power system is accompanied by radioactive waste production. Existing technologies of handling radioactive waste assume their immobilization in materials stable to the impact of the ambient medium and final burial in geological formations [1]. A passive strategy of protection of the population and environment via development of engineering barriers against penetration of radioactive waste from the burial grounds into the biosphere is implemented.

There exists an alternative approach to handling radioactive waste based on the strategy of waste processing by transforming it into short-lived or stable isotopes [2–7]. This transformation is called transmutation. Transmutation can be performed by irradiating radioactive waste with fluxes of elementary particles in specialized nuclear installations (transmuters) or power reactors, as a by-process. At present, the most efficient is transmutation under neutron irradiation (neutron transmutation).

The main long-lived fission products (LLFPs) are the following seven isotopes with half-lives from several tens of thousands to millions of years: ⁷⁹Se, ⁹³Zr, ⁹⁹Tc, ¹⁰⁷Pd, ¹²⁶Sn, ¹²⁹I, ¹³⁵Cs. One of the variants of neutron transmutation is the transformation of long-lived fission products into short-lived isotopes via neutron capture chains. By successively capturing several neutrons, an LLFP can be transformed into a short-lived isotope which then becomes a stable isotope of another chemical element. In this case, the chains of isotopic transformations can be written in the following short form:

⁷⁹Se(*n*,*γ*)⁸⁰Se(*n*,*γ*)⁸¹Se(
$$\beta^{-}$$
,18.5')⁸¹Br;
⁹³Zr(*n*,*γ*)⁹⁴Zr(*n*,*γ*)⁹⁵Zr(β^{-} ,64 days)⁹⁵Nb(β^{-} ,
35 days)⁹⁵Mo;
⁹⁹Tc(*n*,*γ*)¹⁰⁰Tc(β^{-} ,16")¹⁰⁰Ru;
¹⁰⁷Pd(*n*,*γ*)¹⁰⁸Pd(*n*,*γ*)¹⁰⁹Pd(β^{-} ,13.5 h)¹⁰⁹Ag;
¹²⁶Sn(*n*,*γ*)¹²⁷Sn(β^{-} ,2.1 h)¹²⁷Sb(β^{-} ,3.8 days)¹²⁷Te(β^{-} ,
9.3 h)¹²⁷I;
¹²⁹I(*n*,*γ*)¹³⁰I(β^{-} ,12.3 h)¹³⁰Xe; ¹³⁵Cs(*n*,*γ*)¹³⁶Cs(β^{-} ,
13 days)¹³⁶Ba.

If LLFPs are extracted from radioactive waste in the elemental form, they are accompanied by lighter and heavier isotopes. For example, in fragmentation selenium, the long-lived ⁷⁹Se isotope is accompanied by the lighter ⁷⁶Se, ⁷⁷Se, and ⁷⁸Se and heavier ⁸⁰Se and ⁸²Se isotopes. Here, the heavier isotopes play the role of parasitic neutron absorbers, while the lighter isotopes are also the sources of the main radionuclide via (n,γ) reactions. At the beginning of transmutation, it is necessary to spend neutrons for increasing, rather than reducing, the mass of the main LLFP via (n,γ) reactions of neutron precursors.

The dominating role of (n,γ) reactions in neutron transmutation of long-lived fission products can be challenged, although not too seriously, by (n,2n) reactions. Threshold (n,2n) reactions can be initiated only by high energy neutrons. The fraction of such above-threshold neutrons may turn out to be rather large in some regions of fast reactors and electronuclear and fusion installations.

Chains of (n,γ) reactions increase the atomic weights of isotopes. Therefore, we call such chains transmutation "to the right," toward larger atomic weights. Similarly, since (n,2n) reactions reduce the atomic weights of isotopes, we call these chains transmutation "to the left," toward lower atomic weights. The question of partnership or competition between transmutation "to the right" and "to the left" occurs.

2. NEUTRON CONSUMPTION FOR TRANSMUTATION WITHOUT CONSIDERATION OF (*n*,2*n*) REACTIONS

The feasibility of neutron transmutation is determined by neutron balance of the transmuter. It should have an excessive amount of neutrons suitable for transmutation. Traditionally, LLFP yields are measured in terms of the number of nuclei Y(LLFP) produced in one fission. Therefore, the number of neutrons ΔN_{tr} required for transmutation per one fission should not be smaller than the number of LLFP nuclei produced in this fission, i.e., $\Delta N_{\text{tr}} \ge Y(\text{LLFP})$. An LLFP isotope, capturing a neutron, can be transformed into other isotopes and transmutation products which can also absorb neutrons. The minimum number of neutrons required for LLFP transmutation is equal to its yield multiplied by the number of isotopic transitions to the nearest short-lived isotope.

In the case of elemental LLFP transmutation, the minimum number of neutrons is equal to the sum of yields of all isotopes multiplied by the number of neutron captures to the nearest short-lived isotope. For example, the number of neutrons required for elemental transmutation of 93 Zr can be estimated as

$$\Delta N_{\rm tr} = 5Y(^{90}{\rm Zr}) + 4Y(^{91}{\rm Zr}) + 3Y(^{92}{\rm Zr}) + 2Y(^{93}{\rm Zr}) + Y(^{94}{\rm Zr}) + Y(^{96}{\rm Zr}).$$
(1)

There exists a more formalized approach to estimation of neutron consumption for elemental LLFP transmutation. A certain amount of an LLFP element characterized by the vector \vec{Y} (isotope yields) is produced in fission. If these LLFP nuclei are placed in the neutron field of the transmuter, the evolution of this number of nuclei is described by the equation

$$d\vec{G}(t)/dt = \hat{A} \cdot \vec{G}; \qquad \vec{G}(0) = \vec{Y}; \tag{2}$$

where $\vec{G}(t)$ is the vector of the number of nuclei of LLFP isotopes; \vec{Y} is the vector of LLFP yields, nucleus/fission; and \hat{A} is the matrix of isotopic transitions.

The solution to Eq. (2) can be written in the form

$$\vec{G}(t) = \exp(\hat{A}t) \cdot \vec{Y}$$
 (nuclei per fission).

The neutron radiative capture rate for all LLFP isotopes is found as the sum

$$N_c(t) = \sum_i \sigma(n, \gamma)_i \cdot \phi \cdot G_i(t) = (\vec{\sigma}_{n, \gamma} \cdot \phi, \vec{G}).$$

One neutron is lost in each radiative capture event. Therefore, the number of neutrons required for complete elimination of LLFPs is equal to the total capture rate integrated from zero to infinite time,

$$\Delta N_{\rm tr} = \int_{0}^{\infty} N_{c}(t) dt = \left(\vec{\sigma}_{n,\gamma} \cdot \phi, \int_{0}^{\infty} \exp(\hat{A} \cdot t) dt \vec{Y}\right)$$

(neutrons per fission).

The integral can be calculated analytically,

$$\Delta N_{tr} \left[\frac{\text{neutrons}}{\text{fission}} \right] = -(\vec{\sigma}_{n,\gamma} \cdot \phi, \hat{A}^{-1} \cdot \vec{Y})$$

$$= (\vec{\sigma}_{n,\gamma} \cdot \phi, \vec{G}_{eq}),$$
(3)

where $\vec{G}_{eq} = -\hat{A}^{-1} \cdot \vec{Y}$ is the equilibrium amount of nuclei of isotopes determined by the balance between the LLFP production rate in the nuclear power system and the rate of LLFP burning in the neutron transmuter from the equation $d\vec{G}(t)/dt = \vec{Y} + \hat{A} \cdot \vec{G}$. It can be shown that, in the case of elemental transmutation of ⁹³Zr, Eq. (3) is transformed into Eq. (1).

3. NEUTRON CONSUMPTION AND EQUILIBRIUM AMOUNT OF NUCLEI WITH CONSIDERATION OF (*n*,2*n*) REACTIONS

Taking into account (n,2n) reactions, first, considerably changes the matrix of isotopic transitions \hat{A} and, as a consequence, the vector of equilibrium numbers of isotope nuclei $\vec{G}_{eq} = -\hat{A}^{-1} \cdot \vec{Y}$. Second, a neutron source appears, since in each (n,γ) reaction one neutron is lost, while in each (n,2n) reaction one absorbed neutron is replaced by two new ones. As a result, formula (3) determining the minimum neutron consumption for transmutation takes the following form:

$$\Delta N_{\rm tr} \left[\frac{\rm neutrons}{\rm fission} \right] = \left[\left(\vec{\sigma}_{n,\gamma} - \vec{\sigma}_{n,2n} \right) \cdot \phi, \, \vec{G}_{\rm eq} \right].$$
(4)

Thus, (n,2n) reactions can reduce the number of neutrons required for LLFP transmutation.

Let us consider the example of two isotopes connected with each other by (n,γ) and (n,2n) reactions. Let isotope 1 be an LLFP to be transmuted, and let isotope 2 be its stable heavier "mate." The formulas for the equilibrium amounts of isotope nuclei have the form

$$G_{1,eq} = \frac{1}{\sigma_{a.1} \cdot \varphi} \cdot \left[\frac{Y_1}{1 - \varepsilon_{12} \cdot \varepsilon_{21}} + \frac{Y_2 \cdot \varepsilon_{21}}{1 - \varepsilon_{12} \cdot \varepsilon_{21}} \right],$$

$$G_{2,eq} = \frac{1}{\sigma_{a.2} \cdot \varphi} \cdot \left[\frac{Y_2}{1 - \varepsilon_{21} \cdot \varepsilon_{12}} + \frac{Y_1 \cdot \varepsilon_{12}}{1 - \varepsilon_{21} \cdot \varepsilon_{12}} \right],$$
(5)

where $\sigma_{a,i} = \sigma(n,\gamma)_i + \sigma(n,2n)_i$ is the total neutron absorption cross section, $\varepsilon_{12} = \sigma(n,\gamma)_1/\sigma_{a,1}$ is the probability for isotope 1 nucleus to transform into isotope 2 nucleus via (n,γ) reaction, and $\varepsilon_{21} = \sigma(n,2n)_2/\sigma_{a,2}$ is the probability for isotope 2 nucleus to transform into isotope 1 nucleus via (n,2n) reaction.

The terms in formulas (5) have a clear physical meaning. For example, in the formula for $G_{1,eq}$, the first term $Y_1/(1 - \varepsilon_{12} \cdot \varepsilon_{21})$ is the sum of geometric progression describing multiple transformation of isotope 1 nuclei into isotope 2 nuclei and back. The second term $Y_2 \cdot \varepsilon_{21}/(1 - \varepsilon_{12} \cdot \varepsilon_{21})$ is the sum of the geometric progression describing multiple transformation of isotope 2 nuclei into isotope 1 nuclei, back into isotope 2 nuclei, and then again into isotope 1 nuclei. The terms in the formula for the equilibrium amount of isotope 2 nuclei have a similar meaning.

The neutron consumption for transmutation can be determined as follows:

$$\Delta N_{\rm tr} = \frac{\sigma(n,\gamma)_1 - \sigma(n,2n)_1}{\sigma_{a,1}} \left[\frac{Y_1 + Y_2 \cdot \varepsilon_{21}}{1 - \varepsilon_{21} \cdot \varepsilon_{12}} \right] + \frac{\sigma(n,\gamma)_2 - \sigma(n,2n)_2}{\sigma_{a,2}} \left[\frac{Y_2 + Y_1 \cdot \varepsilon_{12}}{1 - \varepsilon_{21} \cdot \varepsilon_{12}} \right].$$

In order to estimate the role of (n,2n) reactions, let us consider the change in the neutron consumption and the equilibrium amount of LLFP nuclei if (n,2n) reactions are taken into account. It can be easily shown that, if $\sigma(n,2n)_1 = \sigma(n,2n)_2 = 0$, then

$$G_{1,eq}$$
(without $n, 2n$) = $Y_1/(\sigma(n, \gamma)_1 \cdot \phi)$;

$$\Delta N_{\rm tr}$$
 (without $n, 2n$) = $2Y_1 + Y_2$.

Then the equilibrium amount of LLFP nuclei with (n,2n) reactions taken into account has the form

$$\Delta G_{1,eq} = G_{1,eq} (\text{with } n, 2n)$$

- $G_{1,eq} (\text{without } n, 2n) = (a \cdot Y_1 + b \cdot Y_2)/\varphi;$

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where

$$a = \frac{\sigma(n,\gamma)_1 \cdot \varepsilon_{21} - \sigma(n,2n)_1}{\sigma_{a,1} \cdot \sigma(n,\gamma)_1 \cdot (1 - \varepsilon_{12} \cdot \varepsilon_{21})};$$

$$b = \frac{\varepsilon_{21}}{\sigma_{a,1} \cdot (1 - \varepsilon_{12} \cdot \varepsilon_{21})}.$$

The coefficient *b* is always positive, and the coefficient *a* can change sign depending on the spectral conditions of the transmuter; i.e., the (n,2n) reaction is capable of both increasing and reducing the equilibrium amount of LLFP nuclei. Without an (n,2n) reaction, isotope 2 does not make any contribution to the equilibrium amount of LLFP nuclei. If the (n,2n) reaction of isotope 2 is taken into consideration, some part of nuclei of this isotope can be transformed into LLFP nuclei with the probability ε_{21} . Therefore, if there exists the heavier "mate" with the higher yield Y_2 , the equilibrium amount of LLFP nuclei can be larger.

The estimate of neutron consumption for transmutation with (n,2n) reactions taken into account gives a more encouraging result. Indeed, it can be shown that

$$\delta(\Delta N_{\rm tr}) = \Delta N_{\rm tr}(\text{with } n, 2n) - \Delta N_{\rm tr}(\text{without } n, 2n)$$
$$= -\frac{3\sigma(n, 2n)_1 \cdot (\sigma_{a,2} \cdot Y_1 + \sigma(n, 2n)_2 \cdot Y_2)}{\sigma_{a,1} \cdot \sigma_{a,2} \cdot (1 - \varepsilon_{12} \cdot \varepsilon_{21})}.$$

Therefore, (n,2n) reactions reduce the neutron consumption for transmutation.

Another example of two isotopes connected by (n,γ) and (n,2n) reactions is as follows. Let isotope 1 be an LLFP to be transmuted, and let isotope 2 be its stable lighter "mate." It can be shown that the formula for the equilibrium amount of LLFP nuclei has the form

$$G_{1,eq} = \frac{1}{\sigma_{a,1} \cdot \varphi} \cdot \left[\frac{Y_1}{1 - \varepsilon_{12} \cdot \varepsilon_{21}} + \frac{Y_2 \cdot \varepsilon_{21}}{1 - \varepsilon_{12} \cdot \varepsilon_{21}} \right],$$

where $\varepsilon_{12} = \sigma(n, 2n)_1 / \sigma_{a,1}$ is the probability for isotope 1 nucleus to transform into isotope 2 nucleus via (n, 2n) reaction and $\varepsilon_{21} = \sigma(n, \gamma)_2 / \sigma_{a,2}$ is the probability for isotope 2 nucleus to transform into isotope 1 nucleus via (n, γ) reaction.

The equilibrium amount of LLFP nuclei with (n,2n) reactions taken into account has the form

$$\Delta G_{l,eq} = G_{l,eq} (\text{with } n, 2n) - G_{l,eq} (\text{without } n, 2n)$$
$$= (a \cdot Y_1 + b \cdot Y_2) / \varphi;$$

where

$$a = -\frac{\sigma(n, 2n)_1 \cdot (1 - \varepsilon_{21})}{\sigma_{a,1} \cdot \sigma(n, \gamma)_1 \cdot (1 - \varepsilon_{12} \cdot \varepsilon_{21})};$$

$$b = -\frac{\sigma_{a,1} \cdot (1 - \varepsilon_{21})}{\sigma_{a,1} \cdot \sigma(n, \gamma)_1 \cdot (1 - \varepsilon_{12} \cdot \varepsilon_{21})}.$$

Taking into account (n,2n) reactions results in a smaller equilibrium amount of LLFP nuclei.

Without consideration of (n,2n) reactions, all nuclei of the lighter isotope transform into LLFP nuclei via the (n,γ) channel. If (n,2n) reactions are taken into account, part of nuclei of this light isotope first transform into LLFP nuclei via the (n,γ) channel and then return back via the (n,2n) channel with the probability ε_{12} . The lightest isotope can also leave the transmutation chain via the (n,2n) channel. This additionally reduces the equilibrium amount of LLFP nuclei.

The estimate of neutron consumption for transmutation with (n,2n) reactions taken into account also gives an encouraging result. It can be shown that

$$\delta(\Delta N_{\rm tr}) = \Delta N_{\rm tr}(\text{with } n, 2n) - \Delta N_{\rm tr}(\text{without } n, 2n)$$
$$= -\frac{3\sigma(n, 2n)_2 \cdot (\sigma(n, 2n)_1 \cdot Y_1 + \sigma_{a,1} \cdot Y_2)}{\sigma_{a,1} \cdot \sigma_{a,2} \cdot (1 - \varepsilon_{12} \cdot \varepsilon_{21})}.$$

In this case, (n,2n) reactions reduce the neutron consumption for transmutation.

Thus, in both cases, i.e., in the presence of both the heavier and the lighter neighbors, taking into account (n,2n) reactions reduces the neutron consumption for LLFP transmutation. Regarding the equilibrium amount of LLFP nuclei when (n,2n) reactions are taken into account, the presence of the lighter neighbor is favorable (lower number of LLFP nuclei) for any spectral conditions of the transmuter. The presence of the heavier neighbor can both reduce and increase the equilibrium amount of LLFP nuclei depending on the spectral conditions and yields of fission products.

4. PARTNERSHIP OF (n,γ) AND (n,2n) REACTIONS

Obviously, (n,γ) and (n,2n) reactions are partners if the LLFP has short-lived "mates" on both sides. Then both kinds of transmutation ("to the left" and "to the right") are irreversible. Nuclei of long-lived fission products, being transformed into the heavier isotope via the (n,γ) channel, cannot return back via the (n,2n)channel because of the short half-life of the neighbor "on the right." Similarly, nuclei of long-lived fission products, being transformed into the lighter isotope via the (n,2n) channel, cannot return back via the (n,γ) channel for the same reason.

Among the seven LLFP isotopes mentioned above, two of them, ¹²⁶Sn and ¹²⁹I, have short-lived "mates" on both sides. The nearest "left" neighbor of ¹²⁶Sn is ¹²⁵Sn with a half-life of 9.64 days, and the nearest "right" neighbor is ¹²⁷Sn with a half-life of 2.1 h. For ¹²⁹I, the nearest "left" neighbor is ¹²⁸I with a half-life of 25 min, and the nearest "right" neighbor is ¹³⁰I with a half-life of 12.3 h. In [4] and a number of other publications devoted to the project BREST, isotope ⁹⁹Tc is considered as one of the candidates for transmutation because of its radiotoxicity and migration capacity. It is assumed that ⁹⁹Tc can be transmuted in the peripheral region of the fast reactor BREST. The relatively soft neutron spectrum at the periphery of the reactor allows one to neglect threshold (n,2n) reactions. The low neutron flux level at the periphery of the reactor, however, may be a stimulus for moving the ⁹⁹Tc transmutation region closer to the center. Numerical estimates show that the rates of (n,γ) and (n,2n) reactions for ⁹⁹Tc increase as the transmutation region shifts from the periphery to the center.

Fragmentation technetium consists of ⁹⁹Tc (100%); i.e., there is no difference between the isotopic and elemental transmutation. Capturing neutrons, ⁹⁹Tc transforms into ¹⁰⁰Tc with a half-life of 16 s, which eliminates backward transformation of ¹⁰⁰Tc into ⁹⁹Tc via the (n,2n) channel. ⁹⁹Tc, however, has two lighter long-lived neighbors, ⁹⁸Tc with a half-life of 4.2 million years and ⁹⁷Tc with a half-life of 2.6 million years. It was shown above that the lighter neighbors cannot hamper LLFP transmutation either with respect to the equilibrium amount of nuclei or with respect to the neutron consumption for transmutation. Nonetheless, shifting the transmutation region toward the center of the reactor for increasing the ⁹⁹Tc burning rate may result in the production of long-lived radioactive isotopes 97 Tc and 98 Tc via (n, 2n) reactions whose intensity increases in a harder neutron spectrum.

A similar situation takes place for ¹³⁵Cs transmutation; it has no heavier stable neighbors. The nearest "right" neighbor is the short-lived isotope ¹³⁶Cs with a half-life of 13.2 days, and the nearest "left" neighbor is the isotope ¹³⁴Cs with an intermediate half-life of 2 years, which does not eliminate reverse ¹³⁵Cs feed via the (n,γ) channel. Lighter neighbors reduce the equilibrium amount of LLFP nuclei and the neutron consumption for transmutation.

5. COMPETITION OF (n,γ) AND (n,2n) REACTIONS

The remaining three LLFPs, ⁷⁹Se, ⁹³Zr, and ¹⁰⁷Pd, have both lighter and heavier stable neighbors. Heavier neighbors with high yields in fission can increase the equilibrium amount of LLFP nuclei via (n,2n) reactions. For ⁷⁹Se, the nearest "right" neighbor is ⁸⁰Se with the yield in fission higher by approximately a factor of 2.5 than the LLFP yield. For ⁹³Zr, the nearest "right" neighbor is ⁹⁴Zr, whose yield in fission exceeds by approximately 40% that of the LLFP. For ¹⁰⁷Pd, the nearest "right" neighbor is ¹⁰⁸Pd, but its yield in fission is lower by approximately 40% than that of the LLFP. So, the corresponding hope and apprehension can be proved or disproved by particular numerical estimates.

6. NUMERICAL ESTIMATES OF THE INFLUENCE OF (*n*,2*n*) REACTIONS ON LLFP TRANSMUTATION

The numerical estimates were performed for a 1D cylindrical model of the fast reactor BREST-OD-300 [8] using the code TIME26 [9] in the framework of the 26-group diffusion approximation. The energy dependences of the microscopic cross sections of (n,γ) and (n,2n) reactions for LLFPs and accompanying isotopes were obtained from the libraries of evaluated nuclear data JEF-PC and from the system JANIS [10]. The equilibrium amount of LLFP nuclei and the neutron consumption for its transmutation obtained with and without consideration of (n,2n) reactions were compared. The results of this comparison given below correspond to the spatial point at which the difference is maximal.

For the three LLFPs (⁹⁹Tc, ¹²⁶Sn, and ¹²⁹I), the difference between isotopic and elemental transmutation is not large. Fragmentation technetium consists of the isotope ⁹⁹Tc alone. The isotope ¹²⁶Sn has seven lighter "mates," but none can influence the equilibrium amount of ¹²⁶Sn nuclei because of fast decays of intermediate isotopes. They can change the neutron consumption for elemental transmutation of ¹²⁶Sn, however. A similar situation takes place for ¹²⁹I. The only lighter "mate," ¹²⁷I, cannot influence the equilibrium amount of ¹²⁸I nuclei because of fast decay of the intermediate isotope ¹²⁸I, but is capable of influencing the neutron consumption.

The estimates for ⁹⁹Tc transmutation showed that taking into account (n,2n) reactions did not influence the equilibrium amount of its nuclei or the neutron consumption. This is explained by the fact that (n,2n)reactions result in the production of long-lived isotopes ⁹⁸Tc and ⁹⁷Tc, which are not present in nonirradiated technetium. In the course of irradiation, ⁹⁹Tc nuclei, first transforming into ⁹⁸Tc and ⁹⁷Tc nuclei via (n,2n) reactions, then return back via higher intensity (n,γ) reactions. The rates of (n,γ) reactions of these isotopes are higher by approximately three orders of magnitude than those of (n,2n) reactions.

The estimates for ¹²⁶Sn transmutation were more encouraging. In the case of isotopic and elemental transmutation, taking into account (n,2n) reactions noticeably (by 19%) reduces the equilibrium amount of ¹²⁶Sn nuclei. As regards neutron consumption, elemental transmutation of ¹²⁶Sn differs strongly from the isotopic one. For isotopic transmutation, taking into account (n,2n) reactions reduces the neutron consumption by 47%, and for elemental transmutation, by just 10% because of additional neutron absorption by light tin isotopes. The estimates for ¹²⁹I transmutation showed that taking into account (n,2n) reactions does not result in noticeable changes in the equilibrium amount of nuclei or neutron consumption. Taking into account (n,2n) reactions reduces the equilibrium amount of ¹²⁹I nuclei by just 0.35% and neutron consumption by 0.7% in both transmutation modes.

The estimates for isotopic transmutation of ⁷⁹Se showed that taking into account (n,2n) reactions hardly changes the neutron consumption, but increases the equilibrium amount of ⁷⁹Se nuclei by approximately 1.2%. For elemental transmutation, taking into account (n,2n) reactions reduces the neutron consumption by 7% and the equilibrium amount of ⁷⁹Se nuclei by 3%.

The estimates for 93 Zr transmutation showed that taking into account (n,2n) reactions hardly changes the neutron consumption in both transmutation modes. The equilibrium amount of 93 Zr nuclei for isotopic transmutation increases by 5%, and for elemental transmutation, it increases by 7% if (n,2n) reactions are taken into account.

The estimates for 107 Pd transmutation showed that taking into account (*n*,2*n*) reactions hardly changes the neutron consumption but increases the equilibrium amount of 107 Pd nuclei by 1.2% in both transmutation modes.

The estimates for isotopic transmutation of ¹³⁵Cs showed that taking into account (n,2n) reactions does not change the equilibrium amount of ¹³⁵Cs nuclei or the neutron consumption. For elemental transmutation, taking into account (n,2n) reactions insignificantly (by 0.3%) reduces the equilibrium amount of ¹³⁵Cs nuclei and noticeably (by 4%) reduces the neutron consumption.

7. CONCLUSIONS

1. Taking into account (n,2n) reactions always reduces the minimum number of neutrons required for LLFP transmutation, irrespective of the spectral conditions and the presence of lighter or heavier LLFP neighbors with any nuclear yields in fission.

2. Taking into account (n,2n) reactions can noticeably influence the equilibrium amount of LLFP nuclei, either increasing or reducing this amount depending on the spectral conditions and mainly the presence of heavier neighbors with high nuclear yields in fission.

3. The strongest effect from (n,2n) reactions is observed in transmutation of the ¹²⁶Sn and ¹²⁹I isotopes, since these two isotopes have both "right" and "left" short-lived "mates."

4. For the three LLFPs (¹²⁶Sn, ¹²⁹I, and ¹³⁵Cs), (n,γ) and (n,2n) reactions are partners contributing to neutron transmutation of LLFPs. The strongest contribu-

tion is made by (n,2n) reactions in ¹²⁶Sn transmutation (reduction of the neutron consumption by 49% and equilibrium amount of nuclei by 19%). This weakly absorbing isotope is characterized by comparable rates of (n,γ) and (n,2n) reactions in the core of the reactor BREST-OD-300.

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