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Original Article

Radiation-induced transformation of Hafnium composition

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ABSTRACT

The safety and efficiency of nuclear reactors largely depend on the monitoring and control of nuclear radiation. Due to the unique nuclear-physical characteristics, Hf is one of the most promising materials for the manufacturing of the control rods and the emitters of neutron detectors. It is proposed to use the Compton neutron detector with the emitter made of Hf in the In-core Instrumentation System (ICIS) for monitoring the neutron field. The main advantages of such a detector in comparison the conventional β-emission sensors are the possibility of reaching of a higher cumulative radiation dose and the absence of signal delays. The response time of the detection is extremely important when a nuclear reactor is operating near its critical operational parameters. Taking Hf as an example, the general principles for calculating the chains of materials transformation under neutron irradiation are reported. The influence of ^{179m1}Hf on the Hf composition changing dynamics and the process of transmutants' (Ta, W) generation were determined. The effect of these processes on the absorbing properties of Hf, which inevitably predetermine the lifetime of the detector and its ability to generate a signal, is estimated.

1. Introduction

The most important measurements which ensure the required level of the nuclear power plants' safety and efficiency are the monitoring and the control of the neutron flux spatial density and the level of energy release in the reactor core. These measurements are directly related to the operating parameters of the reactor and its power [1].

Hafnium is one of the most promising materials for the elements of the ICIS. This metal has unique nuclear-physical properties: effective absorption of thermal and epithermal neutrons; high corrosion resistance and thermal conductivity; dimensional stability in radiation conditions; partial recovery of absorbing properties during prolonged irradiation in neutron fluxes [2,3]. It is due to these advantages that Hf is one of the promising materials for the manufacture of various reactor core elements.

At present, there is a task of selecting a candidate neutron-sensitive element for an in-core neutron detector. Such detector should be operated reliably for more than 4 years (it is the maximum lifetime of the neutron detectors in the WWER), and also be able to instantly respond to any fluctuations in the neutron field. In light of this, the search for materials, which are capable of retaining their properties under unconventional irradiation conditions (higher irradiation dose and higher temperature), is highly relevant.

The widely used β-emission Self Powered Neutron Detectors (SPND) are not able to satisfy the requirements presented above [4–7]. Therefore, instead of inertial SPND, it is proposed to use Compton (prompt response) SPND, and to take the metallic Hf as the emitter material of such detector. In connection with this choice, it is important to understand how the neutron irradiation and the subsequent composition changes affect the ability of Hf to form the SPND's signal.

From the standpoint of the nuclear reactor's sustainable and safe operation, the primary problem that needs to be solved for any used materials is the calculation (or prediction) of the isotopic and elemental compositions. In the literature sources, one can find many papers, which are dedicated to the transmutation and burn-out calculation methods. Unfortunately, these methods are often presented in such a way that it is difficult to apply in practice. The

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reason for this is the authors' desire to summarize the information as much as possible, without going into details. The most frequently encountered demonstration of the nuclear transformations' calculation method is reduced to the consideration of a three-nuclides chain (both for serial transformations and for the case of branching) and usually has the format as in work [8]. Sometimes the authors show only the result, withholding the details of how it was obtained [9].

At present, there are a sufficient number of certified software products that allow to calculate any burn-out and transmutation processes with high accuracy. Such codes are very expensive, and not all researchers have access to them. Therefore, the method of "manual" calculation is a convenient, acceptable and even necessary tool that allows you to study fine details of the occurring processes.

In this paper, the basic principles of the materials' (which exposed to neutrons) composition calculation are demonstrated step by step. This is done on the example of natural Hf.

This article is a continuation of the works [4,10–14] dedicated to the development of the SPND with an emitter of Hf, as well as the study of the possibility of using such sensor in the ICIS.

The Hf SPND is a neutron detector of Compton type, which has an instantaneous (prompt) response. Typically, prompt response detectors are used in an emergency protection system. The signals of the ICIS detectors should have a wide enough range of proportionality to the neutron flow (i.e. the detector needs to have a sufficient level of the neutron flux sensitivity). The sensitivity of the Compton SPNDs with Hf emitters is at an acceptable level [15–17]. This allows using of such detector for the monitoring and control of the energy release in the reactor core.

As mentioned above, the change in Hafnium's ability to absorb neutrons is important from the standpoint of the SPND's signal formation. The efficiency of the (n, γ)-reaction that causes the subsequent (γ, e)-process is implied. The evaluation of the composition changes and absorption ability will be implemented by using the "manual" calculation.

2. The Hafnium transformation chain

The burning-out of the initial nuclide and the formation of a new one in the nuclear reaction involving a neutron can be represented by the following schematic (all chain members are irradiated with neutrons):

$$\overset{n}{\downarrow}X_1 \rightarrow \overset{n}{\downarrow}X_2 \rightarrow\ldots\rightarrow \overset{n}{\downarrow}X_{k-1} \rightarrow \overset{n}{\downarrow}X_k \rightarrow \overset{n}{\downarrow}X_{k+1} \rightarrow\ldots\rightarrow \overset{n}{\downarrow}X_n \rightarrow \quad (1)$$

For members from (1), the change in concentration over time can be described by a system of n ordinary differential equations.

$$\begin{cases} dN_1 = -r_1 \cdot N_1 dt; \\ dN_2 = r_1 \cdot N_1 dt - r_2 \cdot N_2 dt; \\ dN_3 = r_2 \cdot N_2 dt - r_3 \cdot N_3 dt; \\ \ldots \\ dN_n = r_{n-1} \cdot N_{n-1} dt - r_n \cdot N_n dt, \end{cases} \quad (2)$$

where r [s^{-1}] is a nuclear transformation rate (a decay (λ) or a nuclear reaction (r_x, x – a reaction type).

The solution of such a system was obtained by Bateman more than 100 years ago [18] and has the next form:

$$N_n(t) = N_1^0 \cdot \left(\prod_{k=1}^{n-1} r_k\right) \cdot \sum_{k=1}^{n} \frac{e^{-r_k \cdot t}}{\prod_{m \neq k}^{n} (r_m - r_k)}, \; N_1^0 = N_1(t=0). \quad (3)$$

where $m = 1\ldots n$; $k = 1\ldots n$; n – is the number of chain members; r can be represented as λ, r_x or $(\lambda + r_x)$, it depends on the process which is demanded to describe.

If there is a branching in the chain (for example, the chain has a division into two branches A and B, Fig. 1), then, applying the Bateman solution to it, the r_k in the product with $k = [1, n\text{-}1]$ after the branching point, must be changed to partial values, according to the branching ratio ($q_i = r_i/(r_A + r_B)$, i = A; B). In this case, each branch must be calculated independently. If the branches are further connected again, then the number of atoms at such a point (as well as "below" this point) can be obtained by summing up the transformations along both chains [8].

The use of the Bateman's solution requires compliance to the following rules: 1) when setting up the linear chains, for the original nuclides, one should select only those nuclides that existed at the initial moment; 2) the splitting of a complex chain into linear independent components (branches) is applicable to any nuclear transformation, in which a change in the substance concentration occurs over time; 3) the concentration of the nuclides located in the chain after ("below") the branching point is calculated by taking into account the branching coefficients for each branch (in the equation for the concentration, you need to add a multiplier corresponding to the branching coefficient); 4) the concentration of the nuclides below the point, where the branches are reconnected again, is calculated by adding the concentration portions formed for each of the branches taking into account their branching ratios.

The nuclides that are part of the natural Hf undergo nuclear transformations under the influence of a neutron flux (Fig. 1). The schematic in the figure is considerably simplified, as it does not show the metastable states of the formed isotopes (you can see all isomers in Ref. [4]). For example, due to the small fraction of ^{174}Hf (0.16%) in the natural mixture, the corresponding transformation chains (with participation of ^{176m,177m}Lu) (Fig. 1) are not considered. This is done with the aim to demonstrate the above principles for the concentrations calculation as clearly as possible.

As known, the natural Hf composition includes 6 stable isotopes. Therefore, it is necessary to consider 6 initial sources (parent) nuclides, which simultaneously burn-out in (n, γ)-reactions of radiative neutron captures. Each of the chain members has its own reaction cross-section (Table 1) and makes its own contribution to the concentration change of all following nuclides. For example, if you want to calculate the $N(t)$ for ^{178}Hf, you have to account 4 portions of atoms Fig. 2: contributions from ^{174,176,177}Hf and a depletion of initial ^{178}Hf concentration. As a result, it is necessary to sum up all portions, which can be obtained by formula (3):

$$N_{Hf-178}(t) = N^{from\ Hf-174}_{Hf-178}(t) + N^{from\ Hf-176}_{Hf-178}(t) + N^{from\ Hf-177}_{Hf-178}(t) + N^{0}_{Hf-178}(t) \quad (4)$$

The Bateman's solution allows you to separate and study any contributions. For example, counting the change in concentration of ^{183}W, it is necessary to separately calculate which portion

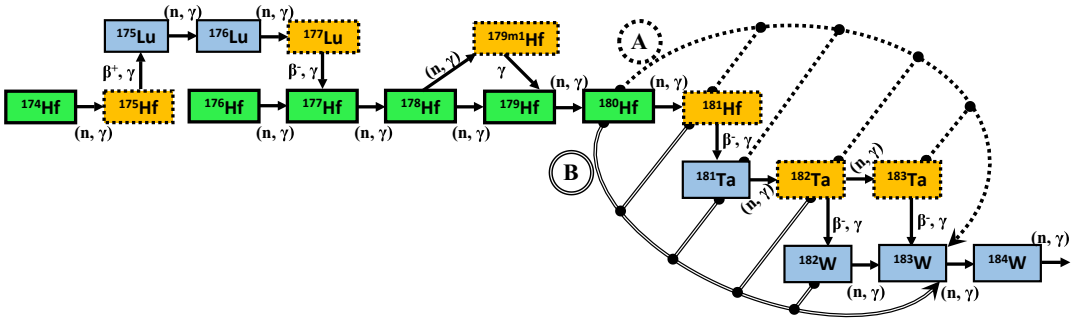

Fig. 1. A simplified schematic for the Hf isotope transformation under the influence of the neutrons flux.

Table 1
The thermal and the resonant neutron capture cross-sections and the half-lives of nuclides from the chain on Fig. 1 taking into account: the resonance integral I; the $k{\cdot}T$ (600 K); the spectrum hardness $k_{res/th} = 0.436$ and the Westkott g-factor.

Element	Hf								Lu			Ta			W		
A, a. m. u	174	175	176	177	178	179	180	181	175	176	177	181	182	183	182	183	184
$T_{1/2}$, d	–	70	–	–	–	–	–	42.4	–	–	6.65	–	115	5.1	–	–	–
$\widehat{\sigma}_{(n,\gamma)}$, b	496	–	318	3395	888	246	21	107	283	5116	–	301	17291	–	286	154	8.3

A is the atomic mass; $T_{1/2}$ is the half-life of the radionuclides; $\widehat{\sigma}_{(n,\gamma)}$ is the cross-section which accounts such factors like T of the moderating medium, the resonant absorption and the hardness of the neutrons spectrum ($\widehat{\sigma}_{(n,\gamma)}$ was obtained by formula (6)).

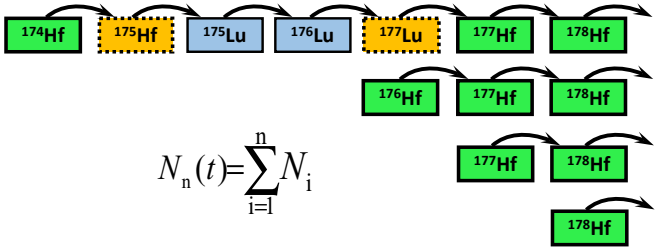

Fig. 2. The linear chains for the calculation of ^{178}Hf concentration.

(fraction) of W atoms is formed by the branches "A" and "B" from each of the 6 original Hf nuclides. In particular, if you look at the contribution from ^{180}Hf (see. Fig. 1), then we get following chains:

$$\begin{aligned} &\text{A: } {}^{180}\text{Hf} \rightarrow {}^{181}\text{Hf} \rightarrow {}^{181}\text{Ta} \rightarrow {}^{182}\text{Ta} \rightarrow {}^{183}\text{Ta} \rightarrow {}^{183}\text{W} \rightarrow; \\ &\text{B: } {}^{180}\text{Hf} \rightarrow {}^{181}\text{Hf} \rightarrow {}^{181}\text{Ta} \rightarrow {}^{182}\text{Ta} \rightarrow {}^{182}\text{W} \rightarrow {}^{183}\text{W} \rightarrow; \end{aligned} \tag{5}$$

By calculating the ^{183}W concentrations from each of the branches and then adding them, we get the fraction of the full concentration that is formed due to the transformation of ^{180}Hf. To find out the true behavior of $N(t)$, we need to make up chains for all Hf isotopes. These chains will be similar to (5). Then we need to describe each of the chains with an equation like (3), and in the end we need to sum up all contributions.

3. The Hafnium composition change

The nuclear reaction rate r [s^{-1}] depends of the neutron flux, the neutron energy and the interaction cross-sections of the material atoms subjected to the irradiation. The dependence of the cross-sections of the (n, γ)-reactions for the nuclei of heavy elements has a complicated character because they have a large number of resonances, related to the excitation of the energy levels of the compound nuclei [19].

The equation for the r includes 2 components and has the following look [20,21]:

$$r_{(n,\gamma)} = \left(\overline{\sigma} + I{\cdot}K_{res/th}\right){\cdot}\Phi_n = \widehat{\sigma}{\cdot}\phi_n \tag{6}$$

where $k_{res/th}$ is the neutron spectrum hardness; I is the resonant integral; Φ_n is the neutron flux density; the cross-section $\overline{\sigma}$ equals to Refs. [20,21].

$$\overline{\sigma} = \frac{\sqrt{\pi}}{2}{\cdot}\sqrt{E_n/E_T}{\cdot}\sigma(E_n){\cdot}g(T) \tag{7}$$

$E_n = 0.0253$ eV; $E_T = k{\cdot}T$; T is the temperature of the medium where the neutrons are moderated; k is the Boltzmann constant; $\sigma(E_n)$ is the microscopic interaction cross-section; $g(T)$ is the Westcott g-factor [22].

In the case when a nuclide, besides its participation in the (n, γ)-reaction, also undergoes the radioactive decay (for example, ^{182}Ta, see. Fig. 1) one should also take into account the concentration decrease in the decay process of the nuclide

$$\lambda_{dec} = \ln 2 / T_{1/2} \tag{8}$$

Thus, for the total rate of disappearance of a nuclide we obtain the following equation

$$r = \lambda_{dec} + r_{(n,\gamma)} \tag{9}$$

The calculation results show that, under the influence of neutron irradiation, the composition of natural Hf is constantly changing. (Figs. 3 and 4). The concentrations of ^{174,176,177,178}Hf isotopes decrease relatively quickly. It is quite clear that the ^{180}Hf concentration is constantly growing. The reason for this is the low value (by more than 10 times) of its cross-section in the (n, γ)-reaction (see. Table 1).

The algorithm described in Section 1, as well as the equations for the obtaining r given earlier, were used to obtain the results in Figs. 3 and 4. The calculation used some average magnitudes of the neutron flux ($5{\cdot}10^{13}$ n/(cm$^2{\cdot}$s)) and the neutron spectrum hardness ($k_{res/th} = 0.436$). The nuclear data for the calculations are taken from the sources [23–25].

It should be noted that we did not take into account the self-

shielding effect. That is, the situation was considered when all particles of the irradiated material are in the same conditions. Software codes allow to take into account the mentioned phenomenon, which, of course, allows you to make a more accurate calculation (the ability to obtain a spectrum of values, and not one averaged characteristic). However, a "manual" calculation is sufficient for the qualitative evaluation and the use of averages.

4. The influence of nuclear isomer on the composition changing

Metastable states (isomers) are an important object of research of radioactivity and play a significant role in understanding of the nuclear structure details. Isomers differ from ordinary excited nuclei states in that they have certain lifetimes [26]. It is important to know how this phenomenon affects the Hf properties. Let's add the ^{179m1}Hf metastable state to the chain (Fig. 1) (the ^{179m1}Hf state has the largest σ, Table 2 [27]). In this case, for example, the chain

$$^{176}\mathrm{Hf} \rightarrow {}^{177}\mathrm{Hf} \rightarrow {}^{178}\mathrm{Hf} \rightarrow {}^{179}\mathrm{Hf} \rightarrow {}^{180}\mathrm{Hf} \rightarrow \qquad (10)$$

turns into the chain.

$$\begin{array}{ccccccccc} & & & & {}^{179m1}\mathrm{Hf} & & & & \\ & & & \nearrow & \downarrow & & & & \\ {}^{176}\mathrm{Hf} & \rightarrow & {}^{177}\mathrm{Hf} & \rightarrow & {}^{178}\mathrm{Hf} & \rightarrow & {}^{179}\mathrm{Hf} & \rightarrow & {}^{180}\mathrm{Hf} \rightarrow \end{array} \qquad (11)$$

Next, the chain (11) should be separated into 2 linear independent components, one of which has the form (10), and the second one

$$^{176}\mathrm{Hf} \rightarrow {}^{177}\mathrm{Hf} \rightarrow {}^{178}\mathrm{Hf} \rightarrow {}^{179m1}\mathrm{Hf} \rightarrow {}^{179}\mathrm{Hf} \rightarrow {}^{180}\mathrm{Hf} \rightarrow \qquad (12)$$

Now, using the algorithm described above, it is possible to calculate the concentration of any chain member. When calculating, for example, the concentration of ^{179}Hf or ^{180}Hf, it is necessary to take into account the branching ratio for each component (10) and (12).

The calculation shows that the appearance of the ^{179m1}Hf state, complicates the chain, but its influence on the composition change nature is small. However, the presence of the isomer has some effect on the ^{178}Hf и ^{179}Hf isotopes (Fig. 5): the 178th starts burn-out faster (because it is necessary to account for the formation of

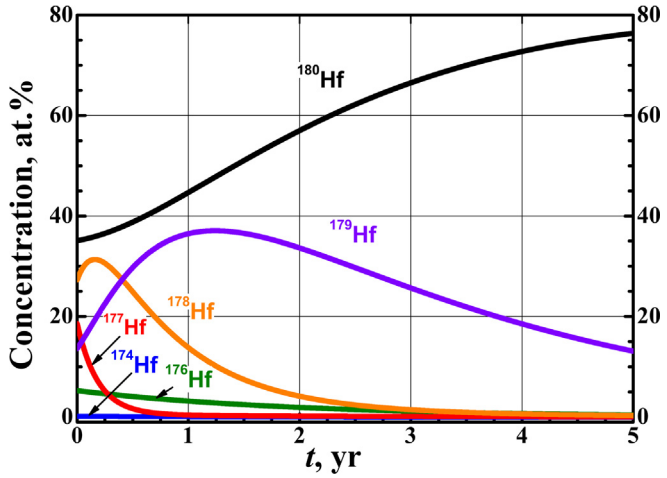

Fig. 3. The composition change of Hf isotopes under the WWER-1000 neutron flux irradiation.

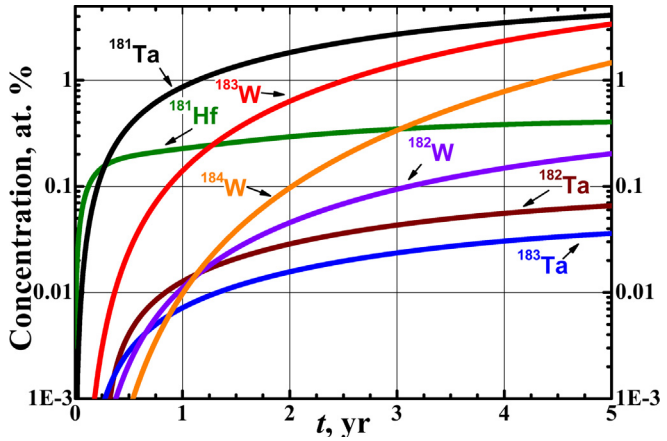

Fig. 4. The transmutants accumulation into Hf under the WWER-1000 neutron flux irradiation.

Table 2
The thermal and the resonant neutron capture cross-sections for some Hafnium isomers [27].

Reaction	σ, b	I, b
^{177}Hf(n, γ)^{178m2}Hf	$2.6\cdot10^{-6}$	$5\cdot10^{-5}$
^{177}Hf(n, γ)^{178m1}Hf	0.96	–
^{178}Hf(n, γ)^{179m2}Hf	$\leq 2\cdot10^{-4}$	$\leq 1.3\cdot10^{-3}$
^{178}Hf(n, γ)^{179m1}Hf	53	–
^{179}Hf(n, γ)^{180m}Hf	0.45	6.9

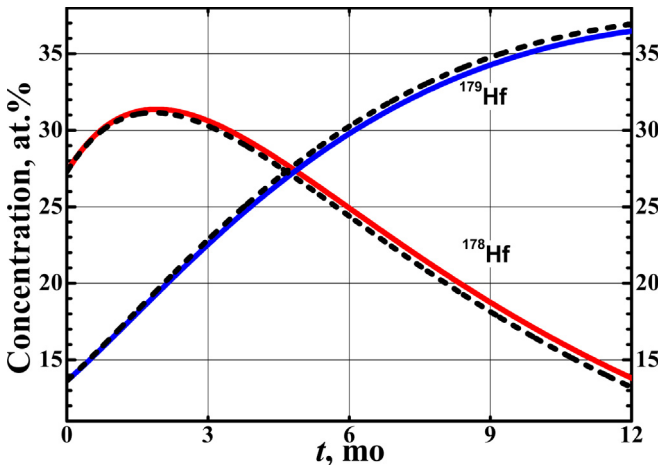

Fig. 5. ^{178}Hf and ^{179}Hf concentration dependence $N(t)$ on the time taking into account the influence of ^{179m1}Hf (dotted curves) and without it (solid curves).

^{179m1}Hf, and it leads to the additional concentration decrease of the parent nuclide); the concentration of the 179th begins to accumulate quicker. It can be seen that, during the entire time, for which the calculation was done, the "difference" between the curves is < 1 at. %. For the concentrations of the transmutants ^{181}Hf, Ta, W, this difference is 2 orders of magnitude smaller. In the case of Hf, the effect of the metastable state on the concentration changes of the initial isotopes and the produced transmutants is insignificant. But in general, one should always check all nuclides, which are present (or will appear) in the irradiated material on the subject of isomers' generation.

5. The evaluation of Hafnium burn-up life

As known, the neutron irradiation changes physical and mechanical properties of materials. These changes determine the service life depending on the exposure dose, its accumulation rate and the environment temperature. These factors are decisive for the materials burn-up life.

The burn-up life can be characterized by the change of the absorption ability (for neutrons) on time. The ability has the following outlook

$$R(t) = \Sigma(t)/\Sigma(0) \tag{13}$$

where $\Sigma(t)$ is the macroscopic cross-section of the nuclides "mixture" at the irradiation time t; $\Sigma(0)$ is the macroscopic cross-section of the nuclides "mixture" before the irradiation start.

The macroscopic cross-section $\Sigma[\mathrm{cm}^{-1}]$ has the meaning of the number of interactions per unit length of the neutron path, i.e. it is the linear attenuation coefficient into the irradiated material. If the material consists of a mixture of nuclei, then the macroscopic section with respect to the process "i" is determined by the equation

$$\Sigma_i = \sum_k \sigma_i^k \cdot N_k \tag{14}$$

where σ_i^k is the microscopic cross-section of ith type of the neutron interaction with the kth kind of nuclei; N_k is the number of kth kind of nuclei per unit volume. The macroscopic cross-section is a material's characteristic and depends on its composition [28,29].

Knowing the behavior of the concentrations of Hf isotopes' and the transmutants accumulated in it (see. Figs. 3 and 4), let's estimate how Hf absorption ability changes with time. It is clear (curves corresponding to $k_{res/th} = 0.436$, Fig. 6) that after the first year of irradiation and later, the absorption ability decreases more slowly, because it begins to largely recover due to the transmutants' accumulation (as calculated for the number of particles corresponding to 1 cm^3 of substance).

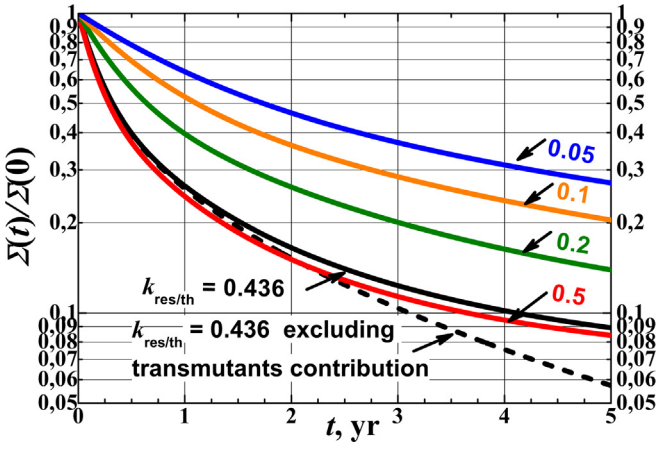

Fig. 6. The absorption capacity change of Hf: $k_{res/th} = 0.436$ corresponds to the calculation result obtained here; $k_{res/th} = 0.05$; 0.1; 0.2; 0.5 corresponds to the results from Ref. [21].

6. Discussion

The results obtained in this work demonstrate the promise of using Hf in the ICIS. With the help of "manual" calculation, the advantages of Hf as a structural material for control rods and emitters of Compton detectors are estimated. This is especially important in the context of a constant increase in the level of Power Plants safety and efficiency, as well as the search for new materials for energy management systems. Hafnium is very well suited for this use. This is demonstrated even by the qualitative prediction of its nuclide composition (see Figs. 3 and 4).

The unique nuclear-physical properties of Hf create a good prospect of its use in pressurized water reactors (PWR). Sufficiently high absorption cross-sections in the thermal range, significant absorption in the epithermal region, high thermal conductivity and mechanical properties are the most important advantages of this material.

The quite complex chain of nuclear transformations (see Fig. 1), having many isomers, does not interfere with the use of Hf. Moreover, successive isotopes 176–180 ensure the preservation (and partial regeneration) of the absorbing properties. The transmutants arising in the process of neutron irradiation (see Fig. 4) also contribute to the partial regeneration of the ability to absorb neutrons (see Fig. 6). The appearance of radioactive nuclides (^{181}Hf, $T_{1/2} \approx 42$ d; ^{182}Ta, $T_{1/2} \approx 115$ d; ^{183}Ta, $T_{1/2} \approx 5$ d) doesn't affect SPND's prompt response ability. This is because the concentration of beta-radioactive transmutants, which are responsible for the slow response, doesn't exceed 0.5% after 5 years operation. Besides this "0.5%" influences only the (n,γ)-portion of the signal, doesn't influence the (γ,e)-portion of the signal. What is important is that the new nuclides (^{181,182,183}Ta, ^{182,183,184}W) have a large Z and can absorb γ-quanta effectively.

From the view point of using Hf as the Compton SPND's material, the partial regeneration of the absorption capacity is an extremely important phenomenon. It slows down the signal level decrease over time, and also allows to increase the detector service life. The compensation occurs due to the Hf isotopes transforming one into another and the accumulation of transmutants, which are capable of working as sources of instantaneous γ-radiation in the (n, γ)-reactions as effectively as the Hf nuclides.

Thus, the Hf's nuclear-physical properties ensure the prospects of this metal as a material for the manufacture of SPND's emitters and control rods of ICIS. This is possible primarily due to the efficiency of Hf in (n-γ-e$^-$)-processes and its high radiation burn-up life.

7. Conclusions

The use of Compton SPND instead of β-emission detectors in the ICIS of the PWR has a good prospect. This proposal arises in light of the trend of increasing the dose and temperature of the irradiated materials (increase in power density). This inevitably brings the technological parameters corresponding to the normal operation of fuel rods and other elements of the core to the critical (limit, threshold) values adopted to prevent the damage (especially fuel rods) in various operating conditions. That is why there is a need for accurate and prompt measurements, as well as the regulation of the energy distribution in the reactor core volume.

It is proposed to use the metallic Hf as an emitter of the Compton SPND. In this regard, the calculation of the change in the metal composition in time was carried out. The obtained results allow us to conclude that Hf is a good candidate for the role of the material for the manufacture of both control rods and emitters of neutron detectors. The influence of the most probable isomeric state on the dynamics of the Hf composition changes was considered separately. The calculation shows that the isomer's influence is insignificant.

The methods for calculating the transmutation and burn-out, presented in the literature, seem quite difficult to put into practice. Therefore, in the framework of this study, the literature sources were analyzed, and the algorithm that allows to estimate the nuclides' concentration changes in nuclear transformations that occur under the influence of neutron irradiation was summarized.

In the future, our research will focus on the Compton SPND's signal level prediction depending on the received radiation dose. In addition, it is planned to carry out the simulation tests by using an accelerator of charged particles.

Declarations of interest

None.

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