

THE BURN-UP PROFILE OF THE COMPTON NEUTRON DETECTOR Hf-EMITTER IN THE WWER-1000 REACTOR

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Several properties of the Compton neutron detector with Hf emitter have been studied in the article. Using the MCNPX program it was shown how the absorption properties can degrade during the 4 fuel campaigns in WWER-1000 core. The nuclide composition of the emitter material was calculated for several coaxial layers in order to find out the burn-up profile. To assess the unevenness of the neutron irradiation, the neutron losses within the emitter layers were calculated.

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INTRODUCTION

In the WWER-1000 reactors, for the neutron field monitoring the in-core Rh-based Self Powered Detectors (SPD) are widely used. Along with the advantages, these SPDs have a significant drawback, a specific delay of the signal due to the emission of an electron in the (n, β) -reaction [1, 2]. To ensure the safety of the reactor operation during transient or emergency modes, other tools are also used, for instance, external ionization chambers. At the same time, inertialess SPDs (also they are called the Compton detectors) could provide additional safety and possibly to improve the economic performance of nuclear reactors by reducing their excessive conservatism. A detector based on metallic Hf, where the (n, γ) , and (γ, e) processes are used, can be one of the prospective inertialess SPD.

So the main interest in the Hf-based SPD is the possibility of the in-core tools modification, which is necessary because of the constant trend of achieving better safety and efficiency of the reactor. Usage of the prompt-response Compton detector together with a high-sensitive β -emissive one can significantly expand the power range, which can be precisely and safely controlled in the reactor.

The main idea of investigation was to find out how burn-up processes influence the neutron absorption properties of the metallic Hf, which is used as an emitter material in the Compton SPD. Therefore, one should estimate the number of neutrons that disappear within the emitter due to the (n, γ) -reaction. Obviously, such a process has a non-uniform character depending on the thickness of the irradiated material and its composition which in its turn also changes in time. So the main purpose of this study was to investigate the neutron losses depending on the depth of the material and the degradation of its absorption properties directly related to the evolution of nuclide composition. Both of them will give the information on trends in the Hf emitter operation and provide all needed data for further studying the detector response features (generation of the γ -quanta, and electrons).

The current work is performed based on the previous articles [3, 4] and reflects new specific features of the

Hf as the emitter material. Particularly, different approach was used concerning the nuclide composition calculations. The Bateman solution was used for previous studies, but here we applied the MCNPX program to deal with the burn-up problem. It allows to reproduce a real-life sources distribution within the volume and provides the working conditions that are close to the actual ones. As a result, the current calculation model of the examined Hf SPD corresponds to the real conditions of WWER-1000 core and allowed us to get more precise results if to compare with previous analytical studies mentioned above.

1. RESEARCH METHODS

1.1. MCNPX MODEL

The subject of the study is the Compton SPD, where the emitter is Hf rod (radius is $r_{Em} = 0.075$ cm), the insulator is MgO (thickness is $d_{Ins} = 0.0414$ cm), the collector is the Inconel-600 tube (wall thickness is $d_{Col} = 0.02$ cm). The detector length is 25 cm. The detector has a coaxial geometry in which an insulator fills the space between the electrodes. The detector model is based on the real prototype described in the [5] work. In the MCNPX model, the Hf detector is located within the central channel in the fuel assembly (FA) Fig. 1, in the standard place for one of the Rh detectors.

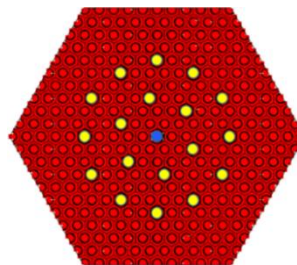


Fig. 1. Fuel assembly cross-section:

- – central channel;
- – guide channel

In order to speed-up the calculations, only FA contains of the SPD was taken into account, and it was just a part of an assembly which corresponds to the detector length. As a result, for the “burn” option in the MCNPX program, we needed to indicate the real power value of this FA part. For the chosen 25 cm part of FA we have

the power 1.8 MW. This value corresponds to real FA which parameters and neutron sources distribution have already been used in [6].

1.2. DATA COLLECTING

Burn-up steps were chosen quite large (144.5 days that is a half of the one fuel campaign) but, as it will be shown in the following chapter, this was done in order to clearly reflect the trends of the nuclide composition changes. It was done just to obtain the difference between the specific points of time. In the real operation, each FA is used for 289 days, so to investigate the Hf emitter material modifications during 4 campaigns of irradiation, it was necessary to use the following approach.

The burn-up process was executed 8 times with a step of 144.5 days (0.5 of the one fuel campaign) as it was mentioned above. The time between the consequent campaigns is 76 days during which the FA has a power equal to 0 in order to use in the next campaign the correct concentration values for all nuclides within the emitter material (it concerns radioactive nuclides that appear in Hf transformation chain during the neutron irradiation). As a result, we got a situation where the Hf emitter had been burning-up for 4 fuel campaigns in the same conditions (SPD burns-up 4 times with a “fresh” FA (2 steps within each fuel campaign), but each time has a new composition of its emitter material that was taken from the previous step).

In order to consider the burning-up process within the emitter, the one was divided into axial layers. From the MCNPX program point of view, it was implemented as burning-up in separate cells with their own composition. Such an approach allows to track the changes in nuclide composition within the emitter over time.

Knowing the concentration $N_k(t)$ for each nuclide and its microscopic cross-section σ_{ny} in the (n, γ) -reaction, one can obtain the macroscopic cross-section, which reflects the absorption properties of the material

$$\Sigma_a = \sum_k \sigma_k^a \cdot N_k, \quad (1)$$

where σ_k^a is the microscopic cross-section for the absorption of neutron by the k^{th} kind of nuclei; N_k is the concentration of the k^{th} kind of atom nuclei. The index “a” in (1) corresponds to the absorption in the (n, γ) -reaction. In order to take into consideration the absorption within the resonant and thermal ranges¹, the values of cross-section σ_k^a used in (1) were corrected. This analytical approach for such correction is based on several studies by different authors [7 - 13] and was precisely described in [4].

The emitter layers mentioned above are shown in the Fig. 2. Thus there were 5 cells limited by cylindrical surfaces, and, according to the aims of this study, it was necessary to find out the neutron losses in each cell.

Neutrons move in all directions, so it is needed to count the difference between how many of them come into the cell and how many come out. To find out this difference, the current integrated over the surface can be used. In the MCNPX program it is so called “F1-tally” [14] (applying to neutrons for surfaces that limit particular cell). As it can be seen in Fig. 2, $\Delta\Phi_n$ is necessary

difference. The arrows in the figure reflect the fact that neutrons are coming into and out of the cell. Thus, the value of $\Delta\Phi_n$ can be described as

$$\Delta\Phi_n = \Phi_n^{\text{INTO the cell}} - \Phi_n^{\text{OUT the cell}}, \quad (2)$$

where $\Phi_n^{\text{INTO the cell}}$ for i^{th} cell is a sum of neutrons that come from the $(i - 1)^{\text{th}}$ and $(i + 1)^{\text{th}}$ cells, and $\Phi_n^{\text{OUT the cell}}$ for i^{th} cell is a sum of neutrons that go to the $(i - 1)^{\text{th}}$ and $(i + 1)^{\text{th}}$ cells. As it has been mentioned, the final measure of neutron losses is being assessed as $\Delta\Phi_n/V_{\text{cell}}$. Results for all “F1-tallies” were normalized to the volumes of the cells limited by the concentric surfaces. So, finally, there were neutron losses per unit volume within each of the emitter layers. Such an approach was used in order to get a specific feature that concerns only the material properties to absorb neutrons.

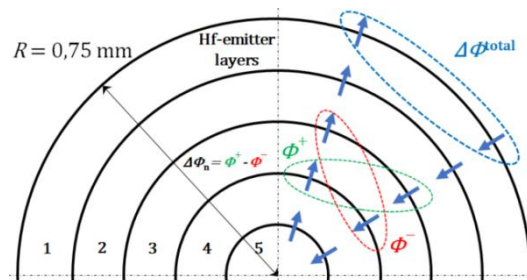


Fig. 2. The emitter layers and the neutron counting

Φ^+ is $\Phi_n^{\text{INTO the cell}}$; Φ^- is $\Phi_n^{\text{OUT the cell}}$

$\Delta\Phi^{\text{total}}$ is a total neutron loss within the emitter

2. RESULTS AND DISCUSSION

2.1. EMITTER MATERIAL COMPOSITION

Due to the irradiation process, all nuclides from the emitter are within a really wide range, from 10^{-5} to 10^2 % at. In terms of the absorption properties, the most important nuclides are those whose portion is at least a few percent. As it can be seen in Fig. 3, after 4 fuel campaigns, ^{179}Hf and ^{180}Hf prevail on other ones. Such a picture takes place for all emitter layers, with a small deviation due to the difference in the neutron flux, which obviously depends on the irradiation depth.

The behaviour of nuclides that are located before ^{180}Hf in the transformation chain has a similar character, it is a permanent depletion. However, it is important to say that the concentration of ^{179}Hf goes up first and then goes down. This effect arises due to the sequential transformations of predecessor nuclides, such as ^{177}Hf , ^{178}Hf , and ^{179}Hf .

Having the highest value of σ_{ny} (3392 b)², ^{177}Hf is transformed into ^{178}Hf at a quite high rate. The ^{178}Hf has σ_{ny} value higher for about 40% than ^{179}Hf (434 b and 300 b, accordingly), that is why the composition spike of ^{179}Hf nuclide takes place. So ^{177}Hf quite quickly disappears, and generation of ^{178}Hf is stopped. Then, as a result, the production of ^{179}Hf also cut off, and it starts to deplete without any regeneration. The cross-section σ_{ny} of ^{180}Hf is quite low (22 b), so the following transformation is limited, and from the point of view of the chemical composition, Hf remains the primary element for the emitter material.

¹ It's about taking into account the resonant integral, the neutron spectrum hardness, and the Westcott g-factor.

² Here and below the cross-sections values from [4] were used where the absorption within resonant and thermal ranges were taken into account.

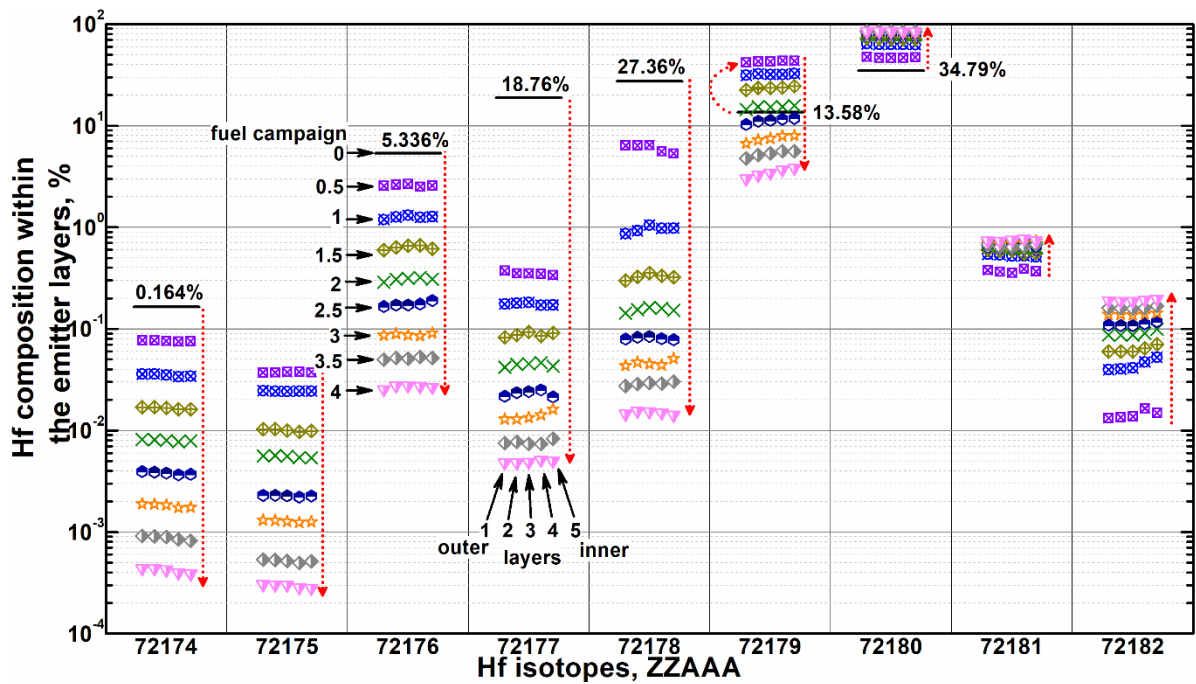


Fig. 3. Nuclide composition within Hf-emitter layers (during 4 fuel campaigns)

In the process of the irradiation by neutrons, the Hf transformation chain expands significantly [15]. There are many transmutants, some of which are of the interest from the standpoint of preserving absorbing properties because of their cross-section values.

It can be seen in Fig. 4, that in layers 1 and 2, there is the highest concentration of the transmutants (Lu, Ta, W, Re). But actually the difference between the layers is insignificant (for the 4th campaign of irradiation, it's about 9.6% for 5th layer and about 10.3% for 1st). It happens due to the shielding effect, obviously; the deeper layers are under the irradiation with lower flux values of neutrons than the outer ones.

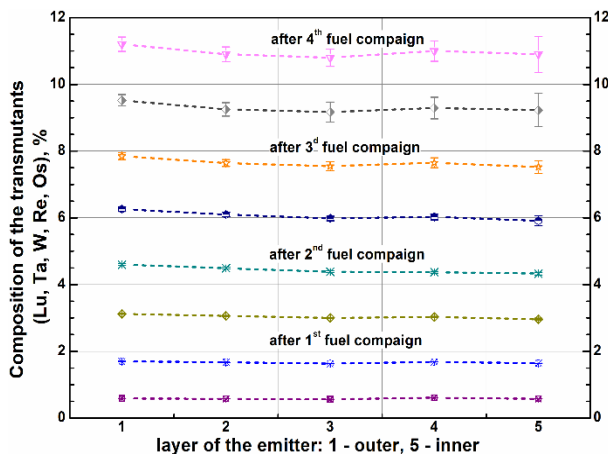


Fig. 4. Transmutants composition within Hf emitter layers (during 4 fuel campaigns)

From the whole set of transmutants, the most important and, as a result, valuable are the nuclides of Ta and W (especially ¹⁸¹Ta and ¹⁸³W). They have about 11% at. of the composition at the end of the irradiation (after 4 fuel campaigns), whereas the rest of the transmutants (Lu, Re, Os) have in a summary no more than 0.15% at. That's why only Ta and W isotope behaviour is shown in Fig. 5.

Concerning the composition within the Hf emitter, the following summary can be done:

- the contribution of Ta and W isotopes continuously goes up leading to the value of their concentration of about 11% at. after the irradiation during 4 fuel campaigns;
- the major nuclides in the transmutants composition are ¹⁸¹Ta and ^{182, 183, 184}W;
- there are some deviations among the concentration behaviour within the different layers because of the shielding effect. But practically, the composition difference between the layers isn't significant and can't seriously affect the material behaviour in terms of its using as an emitter of SPD.

In Figs. 3 and 5 the composition changes within specific emitter layers is shown. In Fig. 6 all that data is put together and one can see how the overall composition of the emitter material changes in time. In general, similar behaviour of nuclide concentrations is observed here. Among the Hf nuclides, the 180th isotope gradually becomes the most significant, while the rest gradually burns-out. Among transmutants, nuclides ¹⁸¹Ta and ¹⁸⁴W are dominant ones.

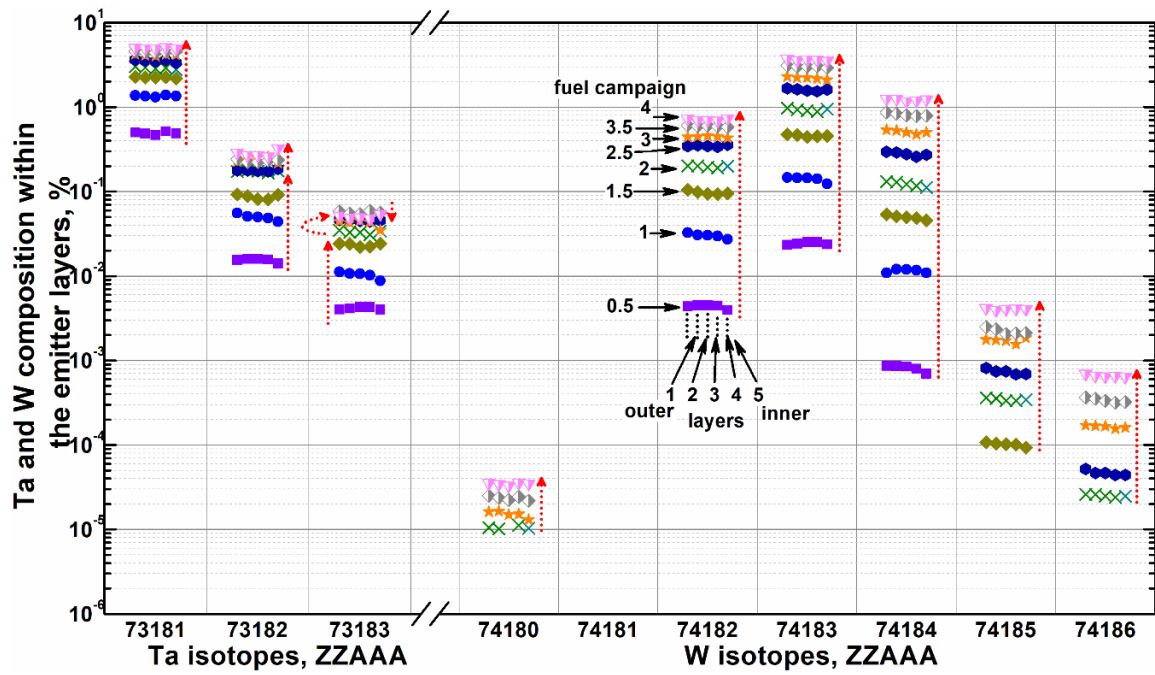


Fig. 5. Ta and W isotopes composition within Hf-emitter layers (during 4 fuel campaigns)

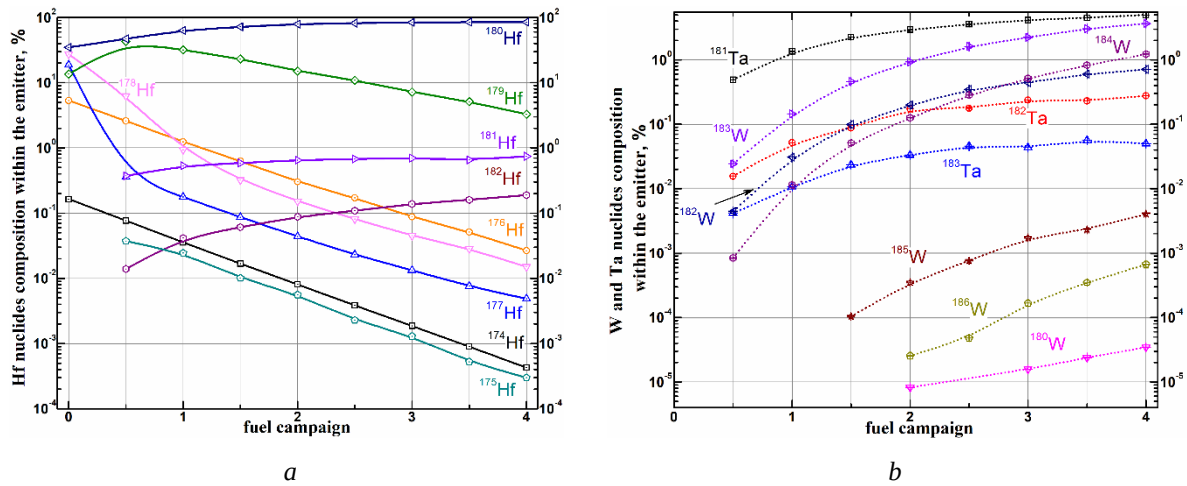


Fig. 6. Composition changes within Hf-emitter (during 4 fuel campaigns):
Hf nuclides (a); Ta and W nuclides (b)

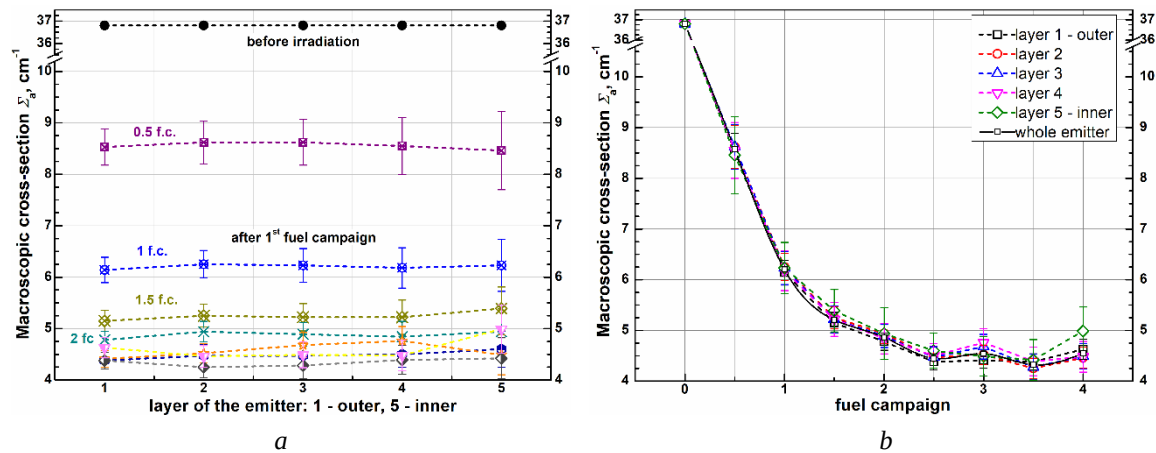


Fig. 7. Macroscopic cross-section Σ_a of an emitter material within specific layers depending
on burn-up time (4 fuel campaigns): changing within layers (a); changing in time (b)

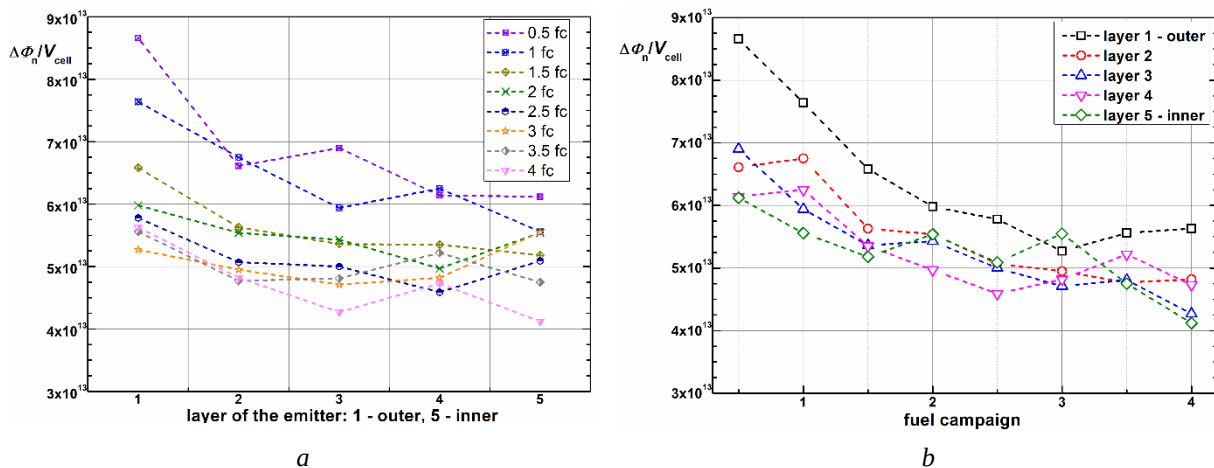


Fig. 8. Neutron losses per unit volume within specific layers depending on the burn-up time (0...4 campaign): changing within layers (a); changing in time (b)

2.2. MACROSCOPIC CROSS-SECTION

The elemental composition of the emitter material mostly consists of Hf. As it can be seen from Fig. 7, the macroscopic cross-section Σ_a (see expression (1)) significantly decreases. The reason of such behaviour is the burning-up of particular Hf isotopes (such as, ^{177}Hf , and ^{178}Hf). The nuclide ^{180}Hf just limits the following burning-up of Hf as an element.

In Fig. 7,a, the data is arranged according to the layers of the emitter, so each line corresponds to the specific burning-up campaign (1...4 with a step of 0.5 fuel campaign). Here one can see the deviations of the Σ_a within each layer. It is obvious that the deepest layer (5) remains with the highest values of the macroscopic cross-sections as it's shielded by the previous ones. During the first 2 years of irradiation, the macroscopic cross-section decreases approximately 7 times. All subsequent changes of Σ_a are within the calculation error, while the macroscopic cross-section remains approximately at the same level.

In Fig. 7,b, the data is arranged according to burn-up years, where each line is for a specific layer within the emitter. It's clear that each layer material has its own Σ_a , but in the meantime it is obvious that macroscopic cross-sections values barely differ from each other, as it is shown by the error bars. Of course, such a picture takes place due to the small size of the irradiated emitter ($r_{\text{Em}} = 0.075 \text{ cm}$), where the self-shielding process can't be as important as for more massive objects (like the control rods, for instance).

2.3. THE NEUTRON LOSSES

Despite minor differences in the macroscopic cross-sections values of the material for the each layer of the emitter, the neutron losses in these layers still differ significantly. As it can be clearly seen in Fig. 8, at the initial stages of the irradiation, the differences in the values of $\Delta\Phi_n/V_{\text{cell}}$ are up to 50% between the inner and outer layers. However, over time, these differences have been smoothed out. At the end of the irradiation, neutron losses in the outer layer are higher than in other layers. Such a picture is formed due to two directly linked effects. Firstly, this is the shielding of the inner

layers, and secondly, this is the longer burn-out of the key Hf nuclides since, due to the shielding process, they are irradiated with a weaker neutron flux.

It can be seen that the efficiency of the emitter material in the (n, γ) -reaction over a period of operation equal to 4 fuel campaigns is significantly reduced. This can be observed both in the degradation of the macroscopic cross-section (see Fig. 7) and in $\Delta\Phi_n/V_{\text{cell}}$ (see Fig. 8). Comparison can only be made in the range of 0.5...4 fuel campaigns. For both $\Delta\Phi_n/V_{\text{cell}}$, comparable rates of attenuation is observed (approximately 1.5-2 times depending on the emitter layer, if we compare the points that correspond to 0.5 of the 1st campaign and the end of the 4th campaign).

In general, the deterioration in the efficiency of absorbing neutrons has negative consequences in terms of the detector reliability. This is so due to the fact that part of the signal that is formed in successive processes (n, γ) and (γ, e) is reduced, and the role of the scattering of the prompt (γ) -quanta emitted during the fission of fuel nuclei and delayed (γ) -quanta emitted by fuel fission products) γ -radiation inevitably arises.

CONCLUSIONS

The data obtained in this study demonstrate how the nuclide composition changes within the Hf emitter of the Compton SPD. During the first 2 campaigns of the irradiation, the absorption capacity of the emitter degrades significantly, decreasing by approximately 7 times. Further irradiation doesn't lead to any significant changes that would somehow affect the performance of the detector. Confirming earlier works, it was shown that the most significant transmutants are Ta and W nuclides.

It was considered how the macroscopic cross-section of the emitter material changes depending on the depth (along the radius). It was shown that there are minor differences, but they barely exceed the calculation errors. However, in this case, considering neutron losses, it is clearly seen that these values differ significantly from layer to layer. And yet, by the end of the chosen irradiation period (4 fuel campaigns), this difference virtually disappears. This effect is based on the process of self-shielding as well as on the extension of the burn-

up period of Hf nuclides, which have sufficiently high cross-sections in the (n, γ) -reaction. However, it is worth noting that the rate of decrease in neutron losses is comparable to the rate at which the macroscopic cross section degrades.

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ПРОФІЛЬ ВИГОРЯННЯ Нf-ЕМІТЕРА КОМПТОНІВСЬКОГО НЕЙТРОННОГО ДЕТЕКТОРА В РЕАКТОРІ ВВЕР-1000

В. Ганн, П. Кузнєцов, О. Улибкін

Досліджено ряд властивостей нейтронного комптонівського детектора з Нf-емітером. За допомогою програми MCNPX було показано, як можуть погіршуватися властивості поглинання протягом 4 паливних кампаній в активній зоні ВВЕР-1000. Розраховано нуклідний склад матеріалу емітера для кількох коаксіальних шарів з метою визначення профілю вигорання. Для оцінки нерівномірності нейтронного опромінення було розраховано втрати нейтронів у окремих шарах емітера.