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COMPTON DETECTOR OF NEUTRONS FOR THE ENERGY YIELDCONTROL IN THE ACTIVE ZONE OF WWER

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The article examines the processes of radiative neutron capture by nuclei of atoms of metallic rhodium and hafnium. Rhodium is used as the emitter in the direct charge detectors of neutrons, which are used to control the energy yield in the core of nuclear power reactors. A new Compton direct charge detector (DCD) (so-called SPND – Self Powered Neutron Detector) with the emitter of the metallic Hf is proposed. The specifics of the signal formation of the β-emission and Compton neutron detectors are considered. The signal lag time of the rhodium detector was calculated and the respective published data was studied, and the conclusion was made that the use of β-emission detectors for the detection of rapid changes of the neutron flux density is inappropriate. The main advantages of the hafnium emitter is a long "burnout" time of the emitter material and the instantaneous response to changes in the neutron flux in the reactor core. Therefore, the use of detectors with such emitters will simultaneously enhance the reliability and safety of operation of the reactors that are in use currently and those that are being developed.

INTRODUCTION

The technology of the electrical energy production in nuclear power field reached a very high level. The evidence to that, for example, may be the Tianwan Nuclear Power Plant (NPP) built in China, which can be characterized as a completely automated production of the electric energy with a new improved safety level. In this NPP all processes capable of provoking severe accidents and the destruction of the containment are excluded: the steam explosion in the reactor vessel; the hydrogen detonation; the repeated criticality of the active zone or melting; steam explosions outside of the reactor vessel. The level of the nuclear fuel burnout is more than 55 (MW·days)/kg U, the fuel campaign is ≥ 5 years, the technical resource of the reactor vessel is 60 years [1].

The further possibility of the improvement in the WWER-1000 (block 466) efficiency is to increase the energy yield in the core with respect to its calculated value, i. e., to achieve the maximum possible energy yield in the safe nuclear reactor operation mode.

The non-homogeneity of the energy yield profile related to the fluctuations of the technological parameters (pressure, temperature, flow level of the nuclear reactor coolant) changes the physical parameters of the active zone, influencing the process of neutron moderation, the flux density and the neutron spectrum. The listed processes do not allow to lower the safety margin (Fig. 1).

To control the profile of the energy yield, the direct charge detectors (DCD) (so-called in literature SPND) of neutrons with rhodium emitters are currently used. They are installed in the fuel assemblies (FA) of the core and act as the sensors of the core monitoring system (ICMS). Further, the information received from the DCDs is used in the reactor control and protection system, which controls the position of the control rods of the nuclear power reactor.

The article discusses in detail the processes of the radiative neutron capture by the atomic nuclei of the electron emitter made of the precious metal rhodium and of the emitter of gamma rays from the metallic hafnium.

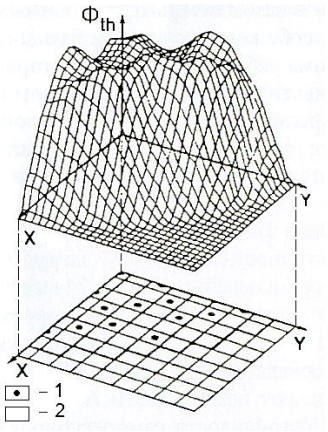

Fig. 1. Spatial distribution of the thermal neutrons flow $\Phi_{th}(\vec{r})$ in PWR reactor's active zone with the partially immersed Reactor Regulators:
1 – elementary cell of active zone;
2 – FA cell [2]

A Compton detector based on metal Hf is proposed to be used as a DCD. This detector has a very long time of the "burnout" of the emitter material and the instantaneous reaction to changes of the neutron flux density in the core of a nuclear reactor. The use of such detectors would simultaneously enhance the reliability and safety of operation of the reactors that are in use currently and those that are being developed.

1. SIGNAL FORMATION OF THE *β*-EMISSIVE NEUTRON DETECTOR

There are two DCD types: the β-emissive DCD, in which current is caused by the β-radiation of radioactive nuclides, and Compton DCD, in which current is created by the electrons, appearing due to the dispersion of the γ-quanta, which are emitted during the radiation capture of neutrons.

Fig. 2 shows the schematic diagram of a DCD. This detector has a set of advantages [3]: no power supply is

required; a simple design; compactness; good stability with respect to temperature and pressure; reproducibility and linearity of the generated signal; relatively low isotope (or isotopes) burnout in the emitter material.

In the WWER-1000 reactor (block B-320), the rhodium DCDs [4] numbering 448 pieces are used.

In the rhodium detector, the stream of electrons is registered by the collector of the device. The stream arises from the radiation during the β⁻-decay of $^{104}_{45}Rh$ ($T_{1/2}$ = 42.3 s), which is formed in the nuclear reaction of the radiative capture of neutrons by the stable mono isotopic $^{103}_{45}Rh$ nuclei of the emitter (see the schematic, Fig. 3).

During the irradiation of the stable isotope rhodium $^{103}_{45}Rh$ with neutrons, in the (n, γ)-reaction, the formation of the radioactive $^{104}_{45}Rh$ isotope and the metastable $^{104m}_{45}Rh$ isotope take place in the proportion of 93.1 and 6.9% respectively. The $^{104m}_{45}Rh$ in the metastable state, by emitting a γ-quantum, is transformed to the radioactive rhodium $^{104}_{45}Rh$, and only 0.13% of $^{104m}_{45}Rh$ emits the electrons and is transformed (as $^{104}_{45}Rh$) to the stable element $^{104}_{46}Pd$.

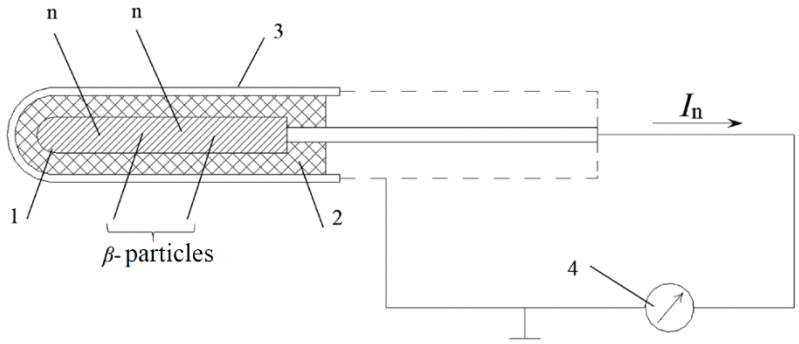

Fig. 2. The schematic of the electron-emissive direct charge detector of neutrons: 1 – emitter; 2 – insulator; 3 – collector; 4 – current measuring instrument

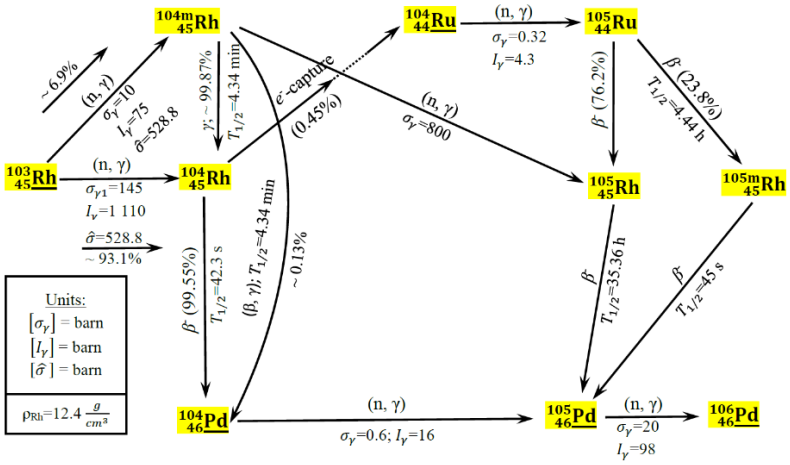

Fig. 3. The schematic of the transformation of daughter elements $^{104m}_{45}Rh$ ($T_{1/2}$ = 4.34 min) and $^{104}_{45}Rh$ ($T_{1/2}$ = 42.3 s), which are the products from radiation capture of neutrons, in the (n, γ) nuclear reaction, by $^{103}_{45}Rh$ atoms of a DCD emitter in the active zone of WWER-1000 reactor

In general, the unstable short-lived elements in the process of the radioactive decay are transformed to daughter elements with the rate of $\lambda = ln2/T_{1/2}$. Therefore, the concentration of the atomic nuclei of the unstable elements is the result of the competition of two processes – their formation with the rate of $\lambda_1 = \sigma_\gamma \cdot \Phi$, respectively (where σ_γ is the cross-section of the nuclear reaction of the radiative neutron capture (n, γ); Φ is the beam flux of thermal and decelerating neutrons) and their decay with the rate of $\lambda_2 = ln2/T_{1/2}$. The concentration of these atoms in a time point t is the difference between the number of the formed and the decayed atoms:

$$\frac{dN_2}{dt} = \lambda_1 N_1(t) - \lambda_2 N_2(t).$$

The solution of this differential equation looks as:

$$N_2(t) = \frac{\lambda_1 N_{10}}{\lambda_2 - \lambda_1} \cdot \left[e^{-\lambda_1 t} - e^{-\lambda_2 t} \right].$$

The atomic concentration of the radioactive elements of the emitter, in the beginning of the irradiation with neutrons, increases and reaches the saturation level, which corresponds to the balance between the mentioned above processes (the formation and the decay). A change of the neutron beam flux Φ results in the increase or decrease of the radioactive atoms formation rate and raises or lowers the level of their equilibrium concentration because the rate of the

radioactive decay remains a constant (Fig. 4). Due to this, the DCD output signal changes, registering the change of the electron stream according to the level of the equilibrium concentration N_2 of the radioactive β^--particle emitting ^{104}Rh and ^{104m}Rh atoms, which are formed by neutrons. This change of the DCD signal is delayed, relative to the moment in time when the neutron beam flux Φ changes, by the approximate time ~ 10 $T_{1/2}$. For a detector with the rhodium emitter (^{103}Rh), this delay is ~ 300 s for the instant neutron beam flux increase, and ~ 400 s for the instant decrease (see Fig. 4).

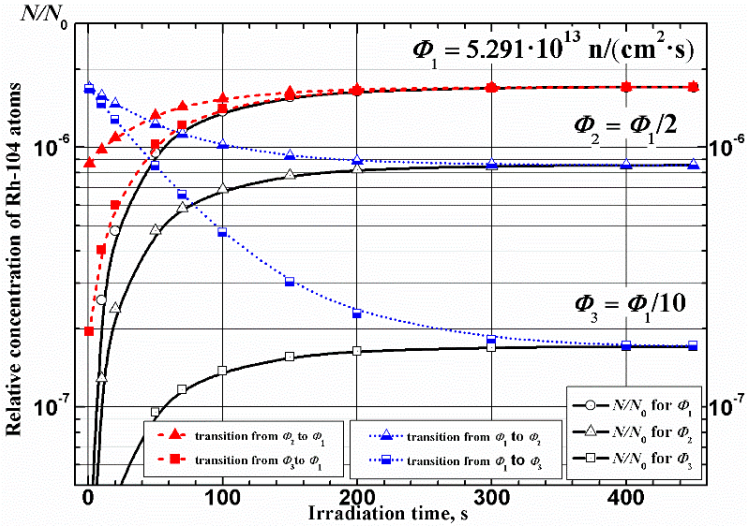

Fig. 4. The formation of the equilibrium concentrations of ^{104}Rh atoms in the nuclear reaction ^{103}Rh(n, γ)^{104}Rh ($T_{1/2}$ = 42.3 s), during their irradiation with neutrons of beam fluxes $\Phi_1 > \Phi_2 > \Phi_3$, taking into account their decay

There is a decrease in the unbalanced equilibrium concentration of the ^{104}Rh atoms with a sharp decrease of the neutron flux density from Φ_1 to Φ_2 and from Φ_1 to Φ_3. Also, there is an increase of the equilibrium concentration of the ^{104}Rh atoms with a sharp increase in neutron flux density from Φ_2 to Φ_1 and from Φ_3 to Φ_1 when transitioning from the respective equilibrium levels Φ_2 and Φ_3.

The analysis of the results leads to the conclusion that, due to rhodium DCD's high inertia, its output signal reflects the intensity of the neutron flux accurately only in the event of a slow flux change.

2. THE SIGNAL FORMATION OF THE COMPTON NEUTRON DETECTOR

The design of the Compton DCD does not differ from the design of the β-emissive detector type. The fundamental difference is the use of the instant γ-quanta radiation from the reaction of the radiation capture (n, γ) of the emitter atoms, which are emitted by the compound nuclei over the life-time $\tau = 10^{-14}$ s [5]. Further, the γ-quanta, during their interactions with the atoms of the emitter (and, partially, the insulator and the collector), generate electrons in the independent processes of the photoeffect, the Compton dispersion and the generation of electron-positron pairs.

The generated electrons are collected on the detector collector and form its electric signal (Fig. 5). The main role is played by the electrons from the following reactions that take place. 1). The reaction in the emitter $(n, \gamma)_e \rightarrow (\gamma, e)_e \rightarrow e_{ec}$, in which the electrons pass through the insulation layer to the collector. 2). The reaction of the γ-rays generated in the emitter with the insulator atoms $(n, \gamma)_e \rightarrow (\gamma, e)_i \rightarrow \Delta e_{ic}$. 3). The reaction $(n, \gamma)_i \rightarrow (\gamma, e)_{ei} \rightarrow e_{ic}$ of the generation of γ-rays during the interaction of neutrons with the atoms of the insulator, which in turn form the electrons in the process of their photoproduction on the atoms of the insulator. 4). The process of $(n, \gamma)_I \rightarrow (\gamma, e)_e \rightarrow \Delta e_{ec}$, in which γ-rays, generated in the isolator produce the electrons on atoms of the emitter, which also flow to the detector collector.

The main advantage of the Compton γ-emission DCD is its immediate response to rapid changes in the neutron flux density in the core of a nuclear reactor, making it preferable for the use in the automated control systems, the control and safety of nuclear reactors [6]. Fig. 6 shows the change in signals of the Compton γ-emission detector (1) and the β-emission detector (2). The immersion of the reactor power regulation rods (RR), which absorb neutrons in the fuel assembly, to the depth from 0 to 1.8 m does not cause the delay of the Compton detector signal within the time of the introduction and removal of the rod (Δt = 4.5 s). The time of the signal change of the Compton detector during the rapid change of the neutron flux density from high to the low (and vice versa) is 0.2…0.3 s. The inertia of the DCD was tested in the core of a nuclear reactor RBMK-1000 with the constant thermal power 2900 MW [6].

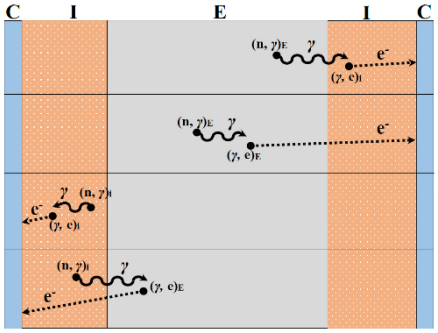

Fig. 5. The main processes of the electrons generation, which form the signal of the Compton emission detector of neutrons:
E – emitter; C – collector; I – insulator;
$(n, \gamma)_E$ – (n, γ)-reaction in emitter material;
$(n, \gamma)_i$ – (n, γ)-reaction in insulator material;
$(\gamma, e)_E$ – (γ, e)-reaction in emitter material;
$(\gamma, e)_i$ – (γ, e)-reaction in insulator material

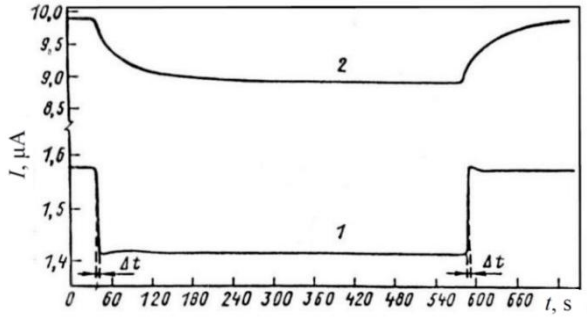

Fig. 6. Changes of the Compton and β-emissive detector signals in the course of the immersion of a RR from 0 to 1.8 and its subsequent return to the initial position [6].
Δt = 4.5 s is the calculated time of a RR movement;
1 – signal of the Compton detector;
2 – signal of the β-emissive detector

Such rapid response of the Compton γ-emission detector is related to the instant decay of and the γ-ray emission from the compound nucleus (lifetime ~ 10^{-14} s), which is formed as a result of the radiative neutron capture (n, γ) by the nucleus of the detector emitter atom or the isolation atom, as well as the time of the collision of γ-rays with the electrons of these nuclei in the process of the photoproduction (times for transitions of the electrons from a bound state in the atoms of the material to the continuum are ~ 10^{-20} s [7, 8]).

The electrons in a Compton *γ*-emission detector are formed in the sequence of two nuclear processes: the formation of γ-rays in the reactions of neutron capture by atoms (n, γ); and the formation of electrons by the generated *γ*-rays in the reactions of photoproduction (γ, e), which both occur with the probability of less than one. In this regard, the total probability of the production of the electrons, which is equal to the product of the probabilities of the sequential processes, is less than the probability of each process. Therefore, the signal of the Compton γ-emission detector is by ~ 5…6 times lower than that of the β^--emission DCD, as calculated per one neutron (see Fig. 6). Therefore, the materials of the emitter for the Compton γ-emission detector should have high enough cross-sections for the radiative capture of neutrons σ_γ and the values of the resonance integrals I_γ. At the same time, the emitter material must maintain its emissivity for quite a long time (years). The multi-isotope material hafnium practically meets the listed requirements.

In the developed Compton detectors [6, 9], which are described above, a HfO_2 powder emitter was used. These detectors were obtained mainly via the cable technology.

3. COMPTON DETECTOR OF NEUTRONS WITH THE EMITTER OF METALLIC Hf

The hafnium isotopes absorb thermal and resonance neutrons in the radiative capture reaction (n, γ). The maximum absorption cross-section for thermal neutrons has the ^{174}Hf isotope, whose content in the natural mixture of isotopes is 0.161%. The maximum resonance integral has the ^{177}Hf isotope (18.56%). During the irradiation of natural hafnium in the core of a nuclear reactor, its "burnout" takes place, and the isotopes are sequentially transformed one into another with increasing of the mass number. Simultaneously, during the irradiation by neutrons, the hafnium isotopes form metastable states with different half-lives of $T_{1/2}$, which, when turning into stable isotopes (isomeric transitions), mainly emit γ-rays. The exception is ^{180}Hf isotope, which is transformed through the capture of neutrons to the radioactive isotope ^{181}Hf with a half-life $T_{1/2}$= 42.4 days, and its decay results in the formation of a stable isotope of tantalum ^{181}Ta with the emission of β^--particles and γ-rays. The hafnium isotope ^{174}Hf during the neutron capture forms the radioactive isotope ^{175}Hf ($T_{1/2}$ = 70 days), the radioactive decay of which occurs with the capture of an electron from the orbit of the atom with the emission of a γ-quantum, which leads to the formation of a stable isotope of lutetium ^{175}Lu. With the growth of its concentration in the irradiated natural-composition hafnium, the isotope reacts with neutrons to form a stable isotope of lutetium ^{176}Lu and the metastable state ^{176m}Lu ($T_{1/2}$= 3.68 hours), a radioactive β^--decay of which leads to the formation of stable isotope ^{176}Hf. In turn, the^{176}Lu stable isotope interacting with a neutron in the capture reaction (n, γ) forms a radioactive isotope of lutetium ^{177}Lu ($T_{1/2}$ = = 6.71 days), whose β^--decay leads to the stable isotope ^{177}Hf.

Figs. 7 and 8 present the diagram of the successive transformations of hafnium isotopes in the steady neutron flux in the reactor core. The analysis of this diagram shows that all isotopes of hafnium are gradually and consistently turning into its heaviest isotope ^{180}Hf. The rate of ^{180}Hf transformation into the radioactive isotope ^{181}Hf (λ ~ 10^{-9} s^{-1}) is by 1–2 orders of magnitude lower than the transformation rate of other (lighter) Hf isotopes into the heavier ones.

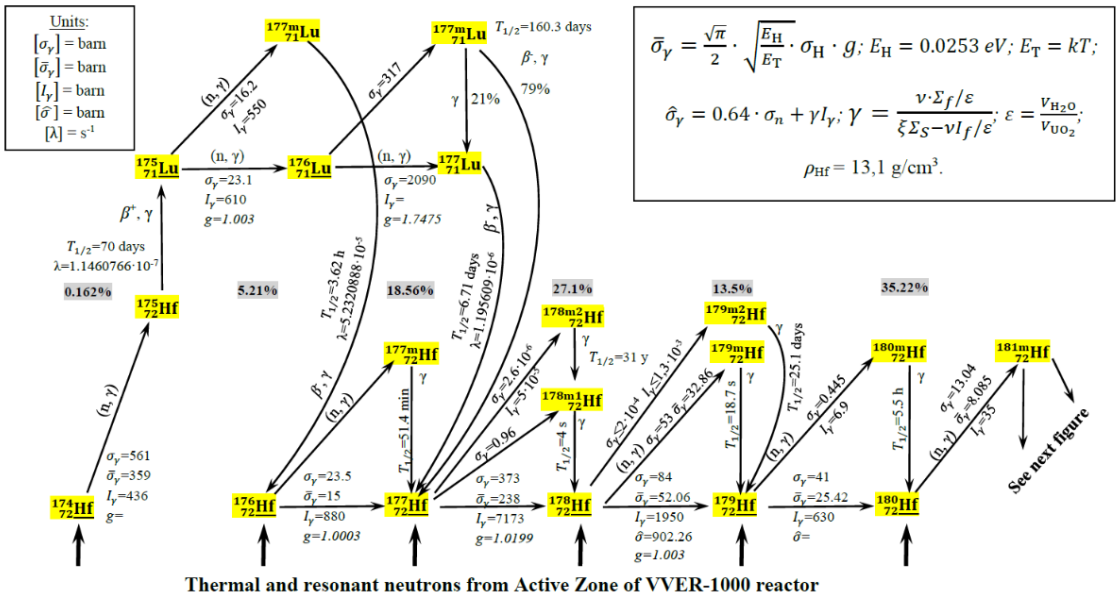

Fig. 7. The schematic of the consecutive transformations of hafnium isotopes under a steady stream of neutrons of the active zone of the WWER-1000 nuclear reactor (see the continuation of the chain in Fig. 8)

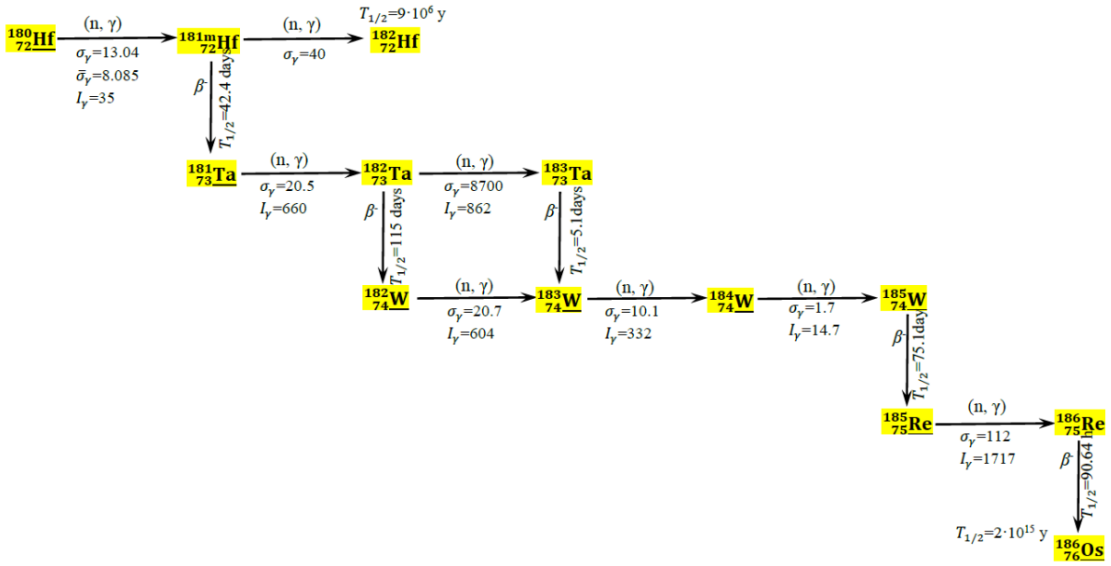

Fig. 8. The continuation of the transformation chain of Fig. 7

During a prolonged irradiation of hafnium of the natural isotopic composition, an equilibrium concentration of the $^{181}_{72}$Hf isotope is formed. This occurs as a result of the competition of the processes of its generation in the nuclear reaction (n, γ) and its β$^-$-decay into the stable isotope $^{181}_{73}$Ta ($T_{1/2}$ = 42.4 days).

The continuation of the transformations of hafnium isotopes in the steady neutron flux leads to the formation of the isotopes $^{182,\ 183}$Ta in the isomeric state ($T_{1/2}$ = 115 days and $T_{1/2}$ = 5.1 days respectively), and the stable isotopes of tungsten $^{182,\ 183,\ 184}$W (Fig. 9).

All these elements, as the hafnium isotopes, are also the capture emitters of γ-radiation [5].

The accumulation of W and Ta at a rate of 3% per year changes the elemental composition of the natural hafnium. This has little effect on the level of the emission of γ-quanta. Under the same conditions, the presently used DCD with the ^{103}Rh emitter burns out with the rate of about 14% per year. In this process, it loses its emissivity, which requires the continuous calibration adjustments while working in the core monitoring system.

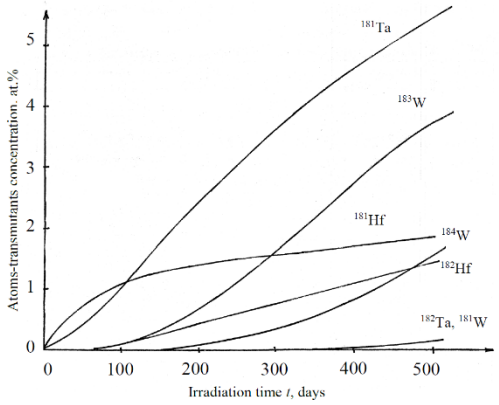

Fig. 9. The generation of atoms products of the natural hafnium transmutation under the irradiation by neutrons in the active zone of the WWER-1000 reactor [5]

4. THE SELECTION OF THE INSULATOR MATERIAL FOR THE COMPTON NEUTRON DETECTOR

The insulator resistance has a significant importance for the current generation process in the γ-emission detector [9, 10]. The reading of the γ-emission detector with an insulator made of magnesium oxide becomes unreliable when its resistance drops to $< 5 \cdot 10^5$ Ohm. The insulation resistance in a healthy Compton γ-emission detector, in the case of close-to-nominal power of the nuclear reactor and the temperature of ~ 650 °C, is $\geq 10^7 \ldots 10^8$Ohm.

During the operation at high temperatures (up to 700 °C) in the core of a nuclear reactor, all components of a DCD are subjected to the irradiation by neutrons, γ-quanta and electrons, which leads to changes in their main electrical and physical properties.

The radiation resistance of materials depends on their chemical composition and the type of valence bonds between the atoms.

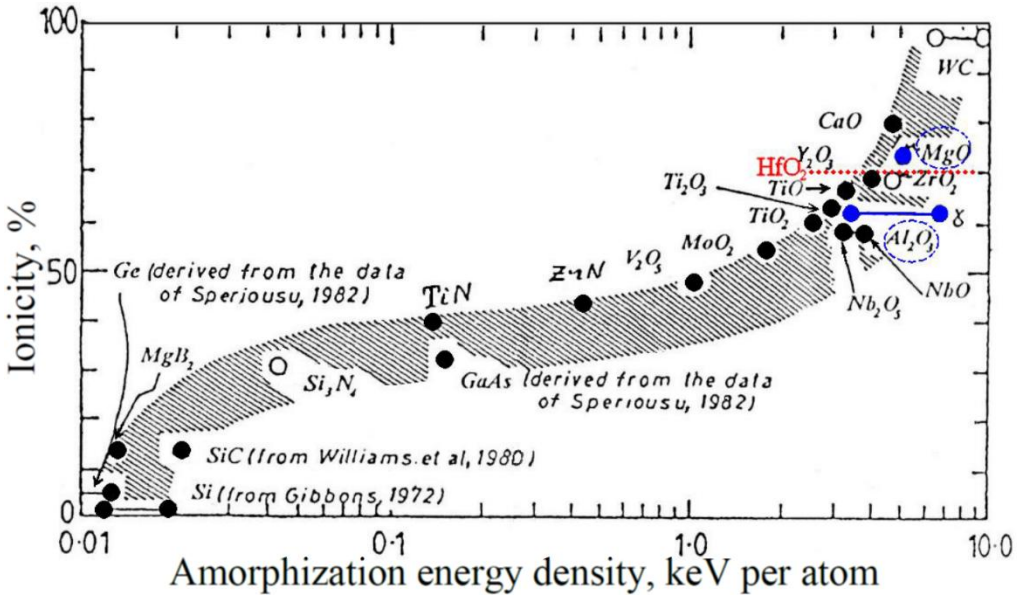

Fig. 10. The correlation of the density of the absorbed energy which leads to amorphicity (transition from crystalline state to amorphous) of various substances, and the characteristic of their valent bonds [13, 14]

To determine the type and direction of the bonds between the atoms, it is necessary to compare the values of electronegativity [11, 12]. The most chemically stable are the ionic structures, in which the atoms have a large difference in the electronegativity values. The atoms with a high value of the electronegativity attract the electron pair stronger. In other words, during the formation of the covalent bond between atoms of different elements, the total electron cloud moves toward the more electronegative atom. The portion of the ionic bonding between atoms is determined by the formula [13]:

$$i = 1 - exp[-0{,}25(X_A - X_B)^2],$$

where X_A and X_B are the electronegativities of the atoms of the molecule. Fig. 10 shows the evaluation of the radiation resistance of different materials taking into account the dependence of their ionic bond fraction on the specific absorbed radiation energy calculated per single atom of the irradiated material. It can be seen that the higher fraction of the ionic bond demands a significantly higher energy density of the bombarding particles to introduce the radiation damage.

The main materials for the DCD insulation are the compounds MgO and Al_2O_3 [3]. The values of the electronegativity of oxygen, magnesium and aluminum differ greatly: O – 3.5; Mg – 1.2; Al – 1.5 [14], and the values of the share of the ionic bonding of these oxides are great: MgO – 0.736; Al_2O_3 – 0.632 (see Fig. 10).

It can be seen that HfO_2 should have an intermediate ionicity value (dashed line in Fig. 10 at $i \approx 70\%$) between those of the commonly used insulators, which bares evidence in favor of its possible choice. Thus, HfO_2, alongside with the known oxides MgO and Al_2O_3, can be used as the insulator material in a DCD.

CONCLUSIONS

This work represents the initial stage of the research of a new Compton neutron detector with the emitter of the metal hafnium intended for use as a DCD in the energy yield control systems of NPP reactors.

The specifics of the signal formation in the β-emissive and Compton detectors of neutrons have been considered. The lag time of the rhodium detector signal was calculated and the literature data analyzed, and a conclusion was made that the use of the β-emission detectors for the detection of the rapid changes of the neutron flux density (for example, when adjusting the energy yield profile) is not advisable.

The use of a new detector with the emitter of metallic Hf and the insulator of HfO_2 was justified. Its main advantages are the long time of the emitter material burnout, and a high response speed in the rapidly changing neutron fluxes.

The use of the proposed detector for monitoring the energy yield profiles would simultaneously enhance the reliability and the safety of operation of different types of reactors (WWER, CANDU, and RBMK).

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КОМПТОНОВСКИЙ ДЕТЕКТОР НЕЙТРОНОВ ДЛЯ КОНТРОЛЯ ЭНЕРГОВЫДЕЛЕНИЯ В АКТИВНОЙ ЗОНЕ ВВЭР

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Рассмотрены процессы радиационного захвата нейтронов ядрами атомов металлических родия и гафния. Родий используется в качестве эмиттера в детекторах нейтронов прямого заряда, которые применяются для контроля энерговыделения в активной зоне ядерных реакторов АЭС. Предложен новый комптоновский детектор прямого заряда с эмиттером из металлического Hf. Рассмотрены особенности формирования сигналов β-эмиссионных и комптоновских детекторов нейтронов. Рассчитано время запаздывания сигнала родиевого детектора, также проведен анализ литературных данных и сделан вывод о нецелесообразности использования β-эмиссионного детектора в условиях быстрых изменений плотности потока нейтронов. Основными преимуществами гафниевого эмиттера являются: большое время «выгорания» материала эмиттера и мгновенная реакция детектора на изменение плотности потока нейтронов в активной зоне реактора. Следовательно, использование детекторов с такими эмиттерами позволит одновременно усилить надежность и безопасность эксплуатации современных и разрабатываемых реакторов.

КОМПТОНІВСЬКИЙ ДЕТЕКТОР НЕЙТРОНІВ ДЛЯ КОНТРОЛЮ ЕНЕРГОВИДІЛЕННЯ В АКТИВНІЙ ЗОНІ ВВЕР

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Розглянуті процеси радіаційного захоплення нейтронів ядрами атомів металів родію і гафнію. Родій використовується в якості емітера в детекторах нейтронів прямого заряду, які застосовуються для контролю енерговиділення в активній зоні ядерних реакторів АЕС. Запропоновано новий комптонівський детектор прямого заряду з емітером із металевого Hf. Розглянуто особливості формування сигналів β-емісійних і комптонівських детекторів нейтронів. Розраховано час запізнювання сигналу родієвого детектора, також проведено аналіз літературних даних і зроблено висновок про недоцільність використання β-емісійного детектора в умовах швидких змін щільності потоку нейтронів. Основними перевагами гафнієвого емітера є великий час «вигорання» матеріалу емітера і миттєва реакція детектора на зміну щільності потоку нейтронів в активній зоні реактора. Отже, використання детекторів з такими емітерами дозволить одночасно посилити надійність і безпеку експлуатації сучасних і розроблюваних реакторів.