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# HAFNIUM IN NUCLEAR POWER INDUSTRY: THE EVOLUTION OF INCREASING OF THE ECONOMIC INDICATORS AND THE OPERATION SAFETY OF PRESSURIZED WATER NUCLEAR REACTORS

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A further increase of the reliability, safety and efficiency of nuclear reactors, the extension of the nuclear fuel campaign and a better control of the fuel burning-out level demand to search for and use new materials. New materials are also necessary for new energy release management elements in the active zone. In this paper, the potential place that hafnium could occupy in the solution of the mentioned tasks is discussed. The high values of the absorption cross-section for thermal and epithermal neutrons, high corrosion resistance, good mechanical properties and heat conduction of hafnium allow for its use, without any barrier coatings, as the absorbing and structural material for regulation rods of nuclear reactors. Another application of high purity hafnium in the nuclear power industry can be in the emitter material of Compton-type Self-Powered Neutron Detectors (SPND). Such SPND will have instant response to neutron flux changes, which will allow to reach positive effects on the reactor safety and efficiency: increase fuel burning-out times; control the crisis boiling coordinates and time; exclude the processes, which could cause heavy accidents).

## INTRODUCTION

Modern light water nuclear reactors WWER, PWR and BWR have high performance characteristics. Nevertheless, there is room for further improvement in the designs of their active zones (AZ) and fuel assemblies of fuel rods (FA). The goals of the improvement are the following: 1) reaching the maximum burn-up fraction of fuel; 2) implementation of more economical and safe ways of managing reactor power generation; 3) using fuels with sufficient multiplication factor *K*, which defines the reactivity reserve $\rho = (K - 1)/K$, which, in turn, determines fuel work during predetermined time at predetermined (nominal *N*) power of reactor; 4) ensuring reliable cooling at nominal power in any possible operating modes; 5) ensuring the possibility of compensating for excessive reactivity ($\rho_{\text{del}}$) in the begin of a fuel campaign and the reactor management during the consumption of reactivity reserve in the process of burning out of $^{235}$U, and formed isotopes $^{239}$Pu and $^{241}$Pu (from $^{238}$U) and poisoning of fuels by the fission products (isotopes Xe и Sm, which absorb neutrons).

The designs of the AZ and the FA determine the neutron-physical properties of the fuel grid and the nature of their changes during the variation of technological parameters (temperature, coolant pressure) as well as the economics and safety of nuclear reactor operation [1]. The value of multiplication factor *K* depends of the AZ parameters and the composition of the used materials (their cross-sections of neutron absorption) in such a way, so as to maintain the balance of neutrons, which is necessary for maintaining a continuous chain reaction of uranium nuclei fission [2]. The determination of the remaining parameters of the reactor and its AZ is only meaningful after ensuring the required value of the neutron multiplication factor *K*. It is obvious that the selected materials should have corrosion resistance in the radiation conditions of AZ; high thermal conductivity; dimensional stability. The above engineering parameters, although they are secondary, determine the economics and safety of reactor operation.

The intensity of heat release of FA in nuclear reactors is theoretically unlimited. This feature is used to select the maximum permissible heat dissipation (energy release) *q* with a reserve for possible deviations from the nominal level:

$$q \le q_{\text{crit}} - \Delta q_{\text{del}}. \qquad (1)$$

The thermal load *q* on a surface is a very important parameter and is uniquely determined by the design of the fuel elements, the concentration of fissile isotopes in the fuel, and the neutron flux density profile. The temperature and its gradients (in the heat-releasing layer of the coolant, which cools the fuel element (FE)) depend on *q*. The critical thermal load $q_{\text{crit}}$ determines the energy release level of the FE, at which the bubble boiling of the coolant on its surface transitions to the film boiling [3, 4]. Above the starting point of the surface boiling, the bulk density of the coolant decreases due to the formation of vapor bubbles in it. During the surface boiling, the collapse of these bubbles occurs. The lifetime of the bubbles depends on the temperature and pressure of the coolant. The change in the coolant density affects the neutron deceleration factor. In this case, the amplitude of the coolant density oscillations increases substantially, which leads to an increase in the noise in the energy spectrum of the neutron flux. There is a spatial instability of energy release, which should be eliminated. Therefore, normal operation and safety of a nuclear reactor is impossible without systems for monitoring and controlling the processes taking place in its AZ.

## EVOLUTION OF WWER AZ DESIGNS

The requirements for increasing reliability, safety, efficiency, increasing fuel burn-up and prolonging its campaign in nuclear power plants lead to the need to use new materials for the elements of the structures of the energy management system. First of all, this concerns the control and protection systems (CPS) and systems for physical control of energy release. The tendency of expansion of the used instrumental measurement systems is quite natural, taking into account the processes described above. The most important measurements in the nuclear power reactors are the determination of the spatial density of the neutron flux and the generated energy in the AZ. The importance of these measurements is in the fact that they are directly linked to the operation parameters of nuclear reactors and their power [5].

Therefore, a tendency of the demand of the increase in the operation safety of newly built light-water reactors, of the increase in nuclear fuel burning out fraction and efficiency requires the development and use of the increasing quantity of measurement systems in the AZ.

In today's conditions, to ensure the safety of the power plant operation, it is necessary to have accurate and timely information on the energy release spatial distribution and the parameters that determine its magnitude.

An example is the system of in-reactor control (SVRK-M) containing 448 Self-Powered Neutron Detectors (SPND) used in the reactor of WWER-1000 of Kalininskaya NPP [6]. Tests of this system were carried out at the 3rd block of the Kalininskaya NPP, the 1st and the 2nd blocks of the Tianwan NPP, the 5th and the 6th blocks of the Kozloduy NPP, the 2nd block of the Balakovo NPP.

Gradual implementation of these efforts led to success. Two units of WWER-1000 (NPP-91) of Project B-428 were built at Tianwan NPP. The project of NPP-91/99 with the B-466 block complies with the standards accepted in the world practice for security systems.

The following was achieved in the named project. 1). The nuclear-physical properties of the AZ and the design of the WWER-1000 reactor components were improved. 2). New control and diagnostic systems were introduced. 3). The possibility of development of processes that can lead to severe accidents is completely excluded in the AZ of the reactor, international tendencies of increasing the safety of high-power NPPs are taken into account. 4). The fuel burn-out level of 55 MW·day/(kg U) has been reached. 5). The 4-5 year fuel campaign is now possible. 6). The transition to uranium-gadolinium fuel with the compensation of its initial reserve of reactivity by gadolinium is assumed. 7). The digital control and management systems were used. 8). The design of the reactor pressure vessel was changed. 9). Witness samples are placed on the inner wall of the pressure vessel. 10). The efficiency of ≈ 35.6% was reached [7]. 11). The technical resource of the pressure body steel is now 60 years [8].

The control and protection system (CPS) is also subject to reconstruction and reconfiguration [7]. As known, the CPS is used to compensate for the excessive reactivity of the fuel being loaded, as well as for controlling and regulating the energy release of the reactor during its operation. The in-reactor control system and the CPS form a centralized control system (CCS). The basis of the CCS is a computer that provides a control over SPNDs' signals (responsible for the registration of the neutron flux), technological parameters and their processing. The respective program provides calculations of following parameters: the current power and the maximum permissible level for each FA; the safety factor $K_{\text{del}}$ up to the maximum permissible capacity and the probability of the crisis-free operation of each fuel assembly and the entire reactor as a whole; the vapor content in the coolant; the burnout of SPNDs' emitter materials; the coefficients of uneven energy release in the AZ; the nuclear fuel reactivity reserve; the recommended movements of control rods, and others. This program also collects data for the analysis of the possibility of emergency situations.

Some mathematical calculations for the operation of a nuclear reactor by the computer programs are there only because of the difficulty (or inability) of direct monitoring of parameters related to the safe operation of the reactor. These parameters and their calculations are controlled by an external mainframe common to all nuclear power reactors. Information exchange between the computers of each NPP nuclear reactor with a common mainframe (for example, BESM-6 at Kursk NPP) is carried out automatically by means of a communication device (for example, AKKORD 1200M).

## THE APPLICATION OF HAFNIUM

Pure hafnium is used as the material of the control rods of the CPS. For the first time, the control rods of hafnium were used in the nuclear reactor of the submarine S.S. (N) 571 Nautilus in 1965 (USA). The good characteristics of hafnium allowed it to be used in the 1st block of the Yankee Rowe NPP in Shippingport, where they worked accident-free for 15 years. Further use of hafnium in nuclear power industry was limited by the high cost of obtaining it. Therefore, for the PWR reactors an alloy Ag-In-Cd was developed. In this alloy, the thermal neutrons were absorbed predominantly by Cd ($\sigma_\gamma = 2450$ b) and the resonant neutrons – by In ($I_\gamma = 3170$ b). However, since 1970, the cost of Ag, which accounts for 80% of the alloy, was increasing and peaked in 1980 due to the rapid pace of development of electronics. In connection with this, Westinghouse (USA) developed a program for the replacement of PWR control rods from the Ag-In-Cd alloy to (95.3%Hf + 4.5%Zn + 0.2%)-alloy. Such an exchange will allow to save an initial design of the control rod cluster in the FA. In 1997, hafnium was used for the first time for mobile control rods of the CPS at the WWER-1000 at Rivne NPP (Ukraine) under experimental operation.

The efficiency of using hafnium as a neutron absorber is characterized not only by the value of its thermal neutron absorption cross-section ($\sigma_\gamma$ = 105 b), but rather by its exceptional ability to absorb higher energy neutrons in the energy range of their deceleration (the values of the resonance integrals of the Hf isotopes are large – from 7137 b for $^{177}$Hf to 630 b for $^{179}$Hf).

The further evolution of WWER-1000 AZ designs confirms that, due to the properties of hafnium, all subsequently built blocks have very high operational parameters and extremely high level of safety ~ $10^{-7}$ (Fig. 1), for example, at the Tianwan NPP (China) and Kudankulam (India). The experience in hafnium exploitation in modern submarine reactors has shown that this material is almost ideal in elements of a CPS. This is due to the FA features of WWER, in which the uranium-water grid are characterized by the tight arrangement of the FE, i. e. insufficient distance for the complete slowing down of the fission neutrons, which don't have time to slow down to the state of thermal equilibrium with the moderator atoms [9]. As a result, a more hard energy spectrum of neutrons is established in WWER than in reactors on thermal neutrons of other types. In addition, a higher proportion of epithermal neutrons leads to an increase in neutron capture by $^{238}$U and forming of $^{239}$Pu and $^{241}$Pu, which have higher values of cross-sections in the (n, f) fission reaction and the number of neutrons $\nu_f$ per one fission act than $^{235}$U.

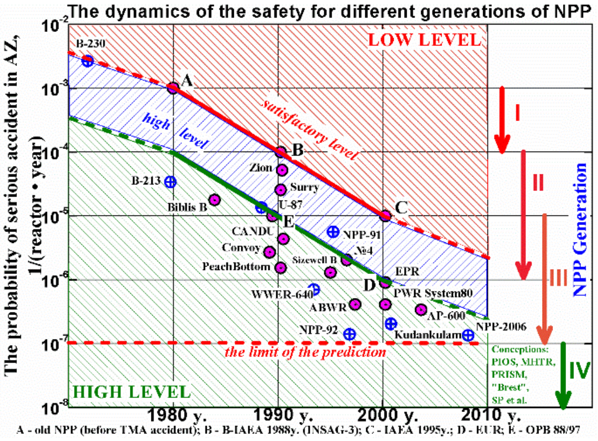


*Fig. 1. Safety of different NPP generations:*
⊕ *– technical documentation data,*
● *– literature data*

This fact complicates the regulation of the nuclear reactor by regulation rods from $B_4C$ and limits the loading of AZ by the U–Gd MOX fuel to the level of ~ 30%.

The cross sections for the capture of neutrons by hafnium in the region of higher energies (above the resonance region in the range 1...100 keV) are ~ 3.5–4 times higher than for $B_4C$ and the Ag-In-Cd alloy [10].

The accumulation of plutonium isotopes is of great importance for increasing the work of the reactor on a single fuel load (i. e., the duration of the fuel campaign). While the $^{235}$U content in nuclear fuel is gradually decreasing (due to its burnout), the content of plutonium isotopes increases and reaches a level, at which more than half of all fission events in nuclear fuel are of the plutonium isotopes nuclei [11].

## DEFORMATION AND RADIATION GROWTH FEATURES OF Hf

Hafnium belongs to the metals with a hexagonal close-packed (HCP) crystal lattice with a small symmetry and exhibits anisotropy of properties in various directions. Crystallographic anisotropy affects the behavior of the radiation deformation of a metal in two ways: 1) at the polycrystalline level – through interactions between the metal grains; 2) at the level of a single crystal – through diffusion anisotropy [12].

Hf consists of small crystals (grains) of various shapes and orientations and, as a whole, does not possess the properties of anisotropy.

Materials with a HCP crystal lattice structure (Zr, Ti, Be, Y, Hf, Cd) are widely used in key elements of AZ structures and are objects of irradiation with intense neutron fluxes. The main problems in the use of these materials are due to radiation growth and the associated phenomenon of radiation creep, the cause of which is the anisotropic nature of their crystal structure. The behavior of these processes largely depends on the parameters and nature of the mechanical/thermal treatment of structural elements in the manufacturing process [13]. Under the neutron irradiation, the products of these materials show a high rate of radiative growth. In the future, with increasing radiation dose, this process reaches saturation. The saturation mode corresponds to the steady parity between the migration of defects formed under irradiation – clusters of vacancies and intrinsic interstitial atoms on the radiation sinks and effluents formed in the material in the manufacture of structural elements in the technological process – grain boundaries and dislocations.

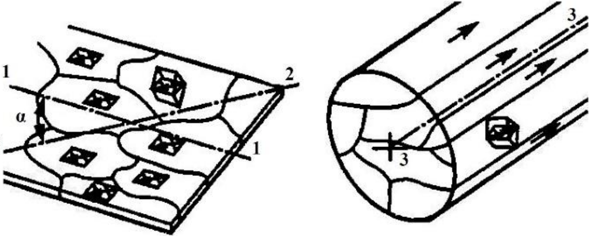
Until recently, the radiation growth was considered as a change in the shape of the irradiated material at its constant volume and absence of stresses and was explained by the orthogonality of the alignment of interstitial and vacancies loops (Buckley loop model [14]). However, the validity of such a definition, confirmed by electron microscopy, corresponds only to uranium with an orthorhombic crystal lattice at high doses of irradiation by fragments of fission reaction products. For some time, it was assumed that a similar mechanism would also occur under neutron irradiation. Subsequently, a few years later, post-radiation measurements of the irradiated materials density with a HCP crystal lattice revealed an increase of their volume (i.e., a decrease in density). Experiments were carried out with the determination of changes in the dimensions of irradiated samples in the longitudinal, transverse and normal directions with respect to the direction of the deformation process (deformation, extrusion, rolling, drawing, forging, etc.) [15].

During the thermo mechanical treatment of polycrystalline hafnium by pressure, the deformation of the grains of metal occurs. Such deformation is accompanied by a rotation of the slip planes (all HCP metals under the deformation are characterized by a slight slip along the directions lying in the basal plane {0001} and a difficult slip along the axis "$c$"). The class relative to $\gamma = c/a$ determines the primary (dominant) sliding system. The position of $\gamma$ with respect to $\sqrt{3} = 1.732$ has a decisive role in relation to the twinning systems involved in the classical deformation. The contribution to deformation of crystals in the $c$ direction is introduced only by pyramidal sliding with the Burgers vector $<c + a>$ and twinning [16]. Depending on the ratio $c/a$, the basic slip along the $a$ direction is usually primary and dominates under deformation.

For hafnium, the primary slip system is the prismatic plane {1010} <$11\bar{2}0$>, the basic slip system is secondary {0001} <$11\bar{2}0$>. The contribution to deformation is made only by pyramidal sliding with the Burgers vector <*c* + *a*> and twinning in the direction *c*: {1011} <1120> and {$11\bar{2}2$} <$11\bar{2}3$>.

The deformation texture is tested with a goniometer by measuring the diffraction of X-rays scattered by grain boundaries. With a small deformation of the grains of metal (~ 1%) because of different orientations, they are deformed to different degrees in relation to the load. With increasing deformation, differences between grains decrease, their microstructure changes. The grains of the metal are gradually stretched in the direction of the plastic flow, the density of the defects increases inside the grains. With considerable deformation, a crystallographic orientation of the grains appears in the metal (so-called, a deformation texture that is the result of simultaneous deformation of grains along several sliding systems depending on the method of deformation and the presence of impurities).

During extrusion, there appear so-called axial textures, when the crystallographic direction for most grains is parallel to the axis of the metal rod. In the case of rolling, the crystallographic plane forms an angle $\alpha$ with rolling plane (Fig. 2).



*a* *b*

*Fig. 2. Textures of rolling (a) and extrusion (b): 1–1 – rolling direction; 2–2 – the axis of the texture; 3–3 – Extrusion direction*

In the experiments, it was established that the Zr single crystal exhibits a radiation growth in the direction of the *a* axis and is compressed in the direction of the *c* axis of the HCP of the crystal lattice. Growth in the direction of the *a*-axis Buckley associated with the loops dislocations formation of intrinsic interstitial atoms, displaced from the nodes of the crystal lattice under the action of neutron irradiation on prismatic planes; compression in direction of the *c*-axis – the formation of dislocations loops on the basis planes [17]. This model was used to obtain the relation describing the radiation growth of polycrystalline materials with a HCP lattice. The magnitude of radiation deformation growth in any given direction $\vec{d}$ in polycrystalline material was associated with its crystallographic texture. It is proportional to the anisotropy parameter of radiation growth *G* and is described by the relationship:

$$G_{\mathrm{d}} = 1 - 3 \cdot f_{\mathrm{d}}^{\mathrm{c}}, \qquad (2)$$

$f_{\mathrm{d}}^{\mathrm{c}}$ is a texture parameter that is based on a numerical description of the pole figures representing a stereographic three-dimensional picture of the X-ray diffraction measurements of a goniometer compressed to a two-dimensional circle as a function of the orientation of the crystallographic planes – base poles $f^{\mathrm{c}}$ in direction *d*. The values $f_{\mathrm{d}}^{\mathrm{c}}$ are calculated from the relationship

$$f_{\mathrm{d}}^{\mathrm{c}} = \sum\nolimits_{\alpha} V_{\alpha} \cdot \cos^2 \alpha \qquad (3)$$

$V_\alpha$ is the volume fraction of grains with basic poles at an angle $\alpha$ to direction *d*.

In the calculations presented above, it is understood that each grain of the metal behaves like an independent single crystal, and that the change in its volume is zero. Equation for $G_{\mathrm{d}}$ it was modified by Holt and Ibrahim [18], to show the lack of vacancy loops in the basal planes, as Buckley believes, assuming the diffusion of vacancies to grain boundaries and representing the parameter $G_{\mathrm{d}}$ in the form:

$$G_{\mathrm{d}} = 1 - f_{\mathrm{d}}^{\mathrm{c}} - 2A_{\mathrm{d}}, \qquad (4)$$

$A_{\mathrm{d}}$ is the anisotropy parameter of grain boundaries with respect to direction $\vec{d}$. For a material with equiaxial grains, this equation reduces to the Buckley equation.

If the proportion of base poles $f_{\mathrm{d}}^{\mathrm{c}}$ equals 1/3, then with a chaotic distribution of the basis poles with respect to the direction $\vec{d}$ there will be no radiation growth. For example, most of *β*-hardened zircaloy specimens have an approximately random distribution of the base poles (i. e., $f_{\mathrm{d}}^{\mathrm{c}} \approx 0.33$) and their radiation growth is very small or zero [19].

The process of radiative growth takes place in three stages: 1) stable growth, depending on the crystallographic structure; 2) transient unsteady growth regime due to radiation-induced changes in the macrostructure – the formation of point defects clusters and dislocation loops under neutron irradiation; 3) the reach of saturation with fluence of neutrons is not higher $5 \cdot 10^{24}$ n/m$^2$.

The last stage is associated with three processes with prolonged irradiation – thermal creep, radiation creep and radiation growth. Due to the presence of residual after heat treatment and annealing of stresses between the grains of metal and the stresses arising from radiation growth, the processes of thermal and radiation creep are included, leading to the relaxation of these stresses. However, these processes cannot make big changes in the classical picture of radiation growth [20]. Nevertheless, when replacing materials of such critical structures as control rods and CPS protection, theoretical and experimental research is needed in the conditions of nuclear reactors.

## REACTOR TESTS OF Hf

The authors investigated the behavior and properties of hafnium, grade GPE-1, produced by GNP “Zirkoniy” (Dneprodzerzhinsk, Ukraine) [21, 22, 23] in the AZ of the SM reactor in an aqueous coolant at the temperature 300 ºC and pressure 15 MPa. Samples for the studies were obtained by hot extrusion at a temperature of 1100 ºC and pressure 14 t/cm$^2$ with subsequent vacuum recrystallization annealing at temperature 850 ºC (1 h) for stress relief between grains of metal (grain size ≈ 28 μm). During this processing, a deformation texture is formed, determined by prismatic sliding and twinning. Irradiation with neutrons up to the fluence value of $5.9\cdot10^{24}$ n/m$^2$ ($E_N > 0.1$ MeV) forms the microstructure of the initial radiation damage, which is essentially the statistical distribution of Frenkel pairs in the volume of metal grains. The pairs arise in the processes of radiation capture reactions (n, γ) of thermal and resonant neutrons by Hf isotopes' nuclei, and the processes of shifting of the Hf atoms at the end of the tracks of the primary knocked out atoms.

In the cascade zone of displacements of hafnium atoms, in the process of a-thermal clustering, and near this zone, vacancies and interstitials form clusters, – dislocations loops. Not all neutrons in the fission spectrum are capable of transferring the energy necessary for the formation of a displacement cascade to the initially knocked out hafnium atoms ($E_d$ = 180 eV) [24]. In the work of Heinisch and Singh [25], the features of the structure of the radiation damage of metals with an increase in their atomic weight are noted, – the increase in the compactness of the displacement zones and the density of defects in them.

The microstructure of the irradiated Hf samples was tested by means of an electron microscope JEM-2000 FXII (120 keV). The occurrence of dislocation loops in a prismatic $\{10\bar{1}0\}$ and the pyramidal planes $\{10\bar{1}1\}$. The formation of loops in the basal plane at this irradiation dose was not detected [22]. The formation of radiation defects stimulated a macroscopic tendency to radiation growth of the Hf samples ($\Delta l/l$ = 0.14…0.34%), the increase of micro-hardness and the reduction of plasticity. There were no signs of corrosion damage to the surface of the samples.

The following observations of damage/changes in the Hf samples, related to their irradiation, were made:

- no changes in the diameter of the samples over the entire irradiation period up to the maximum neutron fluence of $7.81\cdot10^{21}$ n/cm$^2$ ($E_N > 0.1$ MeV) were observed;
- the density of the sample material decreases monotonically with the increasing radiation dose; the decrease does not exceed 1%;
- the radiation growth of Hf samples doesn't exceed 0.4%, while maintaining the growth at a steady rate up to the fluence values of $5.9\cdot10^{21}$ n/cm$^2$;
- the weight gain of samples characterizing the corrosion of Hf under the water-water type nuclear reactor conditions (~ 300 °C, 15 MPa) does not exceed 450 μm/dm$^2$ for 7,000 hours of operation. In this case, all samples of hafnium retained a smooth shiny metal surface;
- when Hf samples are irradiated with neutrons, their micro-hardness increases from 2100 to 4000 MPa, which indicates the compaction of the metal and the reduction of its plasticity.

The results of the reactor tests and the post-reactor studies indicate the high dimensional and corrosion stability of Hf rods, made by the method of hot extrusion, under the irradiation with fission neutrons up to the fluence of $0.6\cdot10^{22}$ n/cm$^2$ ($E_N > 1$ MeV) in water at 300 ºC.

## THE PRESERVATION OF Hf PROPERTIES TO ABSORB NEUTRONS

It is well known that a change in the physical and mechanical properties in metals under irradiation with neutrons often determines the time of their use in the elements of the structures of the AZ reactors. To clarify the physical reasons for the prolonged operation of hafnium in the Yankee Rowe nuclear block in Shipping port (USA), a computational analysis of the retention by this metal of its ability to absorb thermal and resonant neutrons during long-term operation was carried out.

Stable hafnium isotopes, which have high values of the cross sections for the radiative capture of thermal and resonance neutrons, follow one after another in a sequence of mass numbers $A$ = 176…180. Therefore, in the nuclear reactions (n, γ) of radiation capture of neutrons, they are successively converted into heavier isotopes, which leads to a change in their concentrations upon irradiation of natural composition of hafnium. The isotope $^{180}$Hf has much smaller neutron absorption cross-sections ($\sigma_\gamma$ = 13 b; $I_\gamma$ = 35 b) and, consequently, it has a low rate of further radiation transformation. Therefore, the $^{180}$Hf isotope accumulation takes place, which determines the "burnout" of natural hafnium. This process is described mathematically by a system of coupled differential equations, the solution of which makes it possible to calculate the concentration of Hf isotopes at different times [23] (Fig. 3).

The burning-out of $^{177}$Hf isotope, which has the highest value of the resonance integral $I_\gamma$ = 7127 b, with a great rate turns it into $^{178}$Hf and $^{179}$Hf. Their concentration increases and reaches a saturation level at the time of almost complete disappearance of $^{177}$Hf due to small rates of their transformations to $^{180}$Hf, which is ~ 4 and 11 times less. The concentration of $^{180}$Hf monotonically increases, since its transformation rate in the steady neutron flux of the WWER-1000 AZ into the Ta and W isotopes is lower than the conversion rate of $^{178}$Hf (3 times) and $^{179}$Hf (18 times).

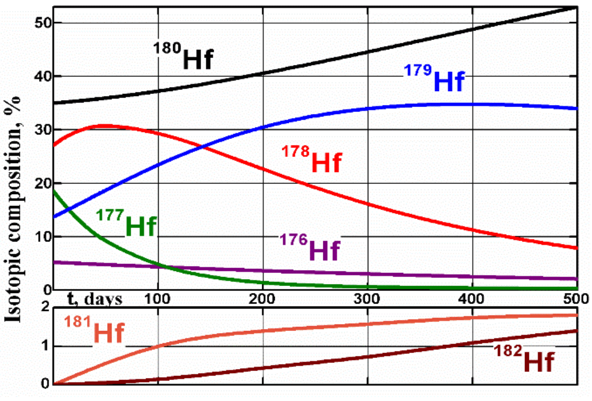


*Fig. 3. The change in the concentration of Hf isotopes under irradiation in a neutron flux of the WWER-1000 AZ*

The radiative redistribution of the natural hafnium isotopic composition leads to the last stable $^{180}$Hf isotope and its burning out at the rate of ~ 2% per year. This process doesn't change the number of atoms in the irradiated hafnium, these «burnt out» atoms in the neutron flux in the reactor AZ are transformed into Ta and W isotopes, in which the total cross-sections $\hat{\sigma}_i$ of neutron absorption is ~ 10 times higher than in $^{180}$Hf. Due to this, the decline in the neutron absorption level by CPS rods is compensated for, when the rods are continuously irradiated over 300 days. The absorptivity is determined by the macroscopic absorption cross-section $\sum_i \hat{\sigma}_i N_i(t)$, where $N_i(t)$ is the concentration of i-th isotope at the time $t$, and $\hat{\sigma}_i$ is the total macroscopic absorption cross-section of thermal and resonant neutrons (Fig. 4).

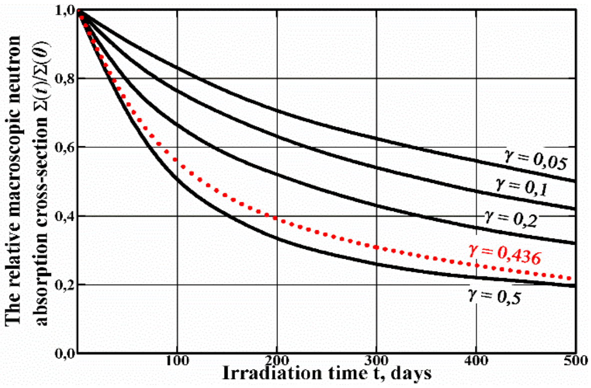


*Fig. 4. Change in the ability of Hf to absorb neutrons upon irradiation in the WWER-1000 AZ (γ = 0.436). The curves for γ = 0.05; 0.10; 0.20, and 0.5 – calculation from [27]*

The large dependence of the neutron absorption by hafnium on the value of the resonance integrals of its isotopes is related to the stiffness parameter $\gamma \sim \Phi_{res}/\Phi_{thermal}$. The stiffness parameters for the energy spectra of different pressurized-water reactors' AZ's are significantly different. The reasons for this are: 1) the use of different moderators ($H_2O$, $D_2O$, C); 2) the difference in temperatures and pressures in their AZ; 3) different levels of fuel enrichment. As a result, the rate of change in the natural hafnium isotope composition and its ability to absorb neutrons in the WWER, PWR, RBMK, CANDU, BWR depend in different ways on the irradiation time (see Fig. 4).

The regulation of the energy release by the CPS rods is carried out by the commands of the executive mechanisms that are generated by the computer based on the information from neutron SPNDs located in the volume of the reactor AZ and in each FA located in more than 500 locations [26]. Requirements for the material of the SPND emitter don't differ from the requirements for the material of the CPS rods. Hafnium as an emitter material of an instantaneous SPND makes it possible to increase the level of operation safety for nuclear reactors to the practical limit value of the probability of a severe accident in a nuclear reactor, equal to $10^{-7}$ in the units of [reactor · year] (see Fig. 1) [28].

## CONCLUSIONS

In the conditions of constant increase of the requirements to the reliability, safety and operation efficiency of the active and under-construction pressurized-water reactors, new applications for hafnium are opened. This is due to its unique nuclear-physical properties: the effective absorption of thermal and epithermal neutrons (which is important in the light of the development of fast neutron reactors); good corrosion resistance; high thermal conductivity; dimensional stability under radiation conditions; partial preservation of the ability to absorb neutrons at prolonged exposure.

The listed hafnium qualities make it possible to use Hf as a material of control rods for CPS and emitters of Compton type SPND.

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## ГАФНИЙ В АТОМНОЙ ЭНЕРГЕТИКЕ: ЭВОЛЮЦИЯ ПОВЫШЕНИЯ ЭКОНОМИЧЕСКИХ ПОКАЗАТЕЛЕЙ И БЕЗОПАСНОСТИ ЭКСПЛУАТАЦИИ ЯДЕРНЫХ РЕАКТОРОВ С ВОДОЙ ПОД ДАВЛЕНИЕМ

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Увеличение надёжности и безопасности, повышение эффективности ядерных реакторов, продление кампании топлива и контроль уровня его выгорания требуют поиска и использования новых материалов, в том числе и для элементов управления энерговыделением в активной зоне. В работе обсуждается, какое место может занять гафний при решении перечисленных задач. Высокие сечения поглощения тепловых и резонансных нейтронов, коррозионная стойкость, механические свойства и теплопроводность гафния позволяют использовать его без оболочек и покрытий в качестве поглощающего и одновременно конструкционного материала для регулирующих стержней ядерных реакторов. Другим применением чистого гафния в ядерной энергетике может быть использование его в качестве эмиттера в комптоновском детекторе прямого заряда (ДПЗ). Такой ДПЗ будет обладать мгновенной реакцией на изменения потока нейтронов, что во многом позволит достичь положительных эффектов (увеличения выгорания топлива; контроля координаты и времени кризисного кипения; исключения процессов, приводящих к тяжелым авариям).

## ГАФНІЙ В АТОМНІЙ ЕНЕРГЕТИЦІ: ЕВОЛЮЦІЯ ПІДВИЩЕННЯ ЕКОНОМІЧНИХ ПОКАЗНИКІВ ТА БЕЗПЕКИ ЕКСПЛУАТАЦІЇ ЯДЕРНИХ РЕАКТОРІВ ІЗ ВОДОЮ ПІД ТИСКОМ

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Збільшення надійності та безпеки, підвищення ефективності ядерних реакторів, подовження кампанії палива та контроль рівня його вигоряння вимагають пошуку і використання нових матеріалів, в тому числі, й для елементів управління енерговиділенням в активній зоні. У роботі розглядається, яке місце може зайняти гафній при вирішенні перерахованих завдань. Високі перетини поглинання теплових і резонансних нейтронів, корозійна стійкість, механічні властивості та теплопровідність гафнію дозволяють використовувати його без оболонок і покриттів в якості поглинаючого та одночасно конструкційного матеріалу для регулюючих стрижнів ядерних реакторів. Іншим застосуванням чистого гафнію в ядерній енергетиці може бути використання його в якості емітера в комптонівському детекторі прямого заряду (ДПЗ). Такий ДПЗ буде мати миттєву реакцію на зміни потоку нейтронів, що багато в чому дозволить досягти позитивних ефектів (збільшення вигоряння палива; контролю координати і часу кризового кипіння; виключення процесів, що призводять до тяжких аварій).