CONTRIBUTION OF RADIONUCLIDES TO HEAT RELEASE IN THE PROCESS OF SNF DRY STORAGE

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In the paper the radionuclide contribution to heat release at dry storage of spent nuclear fuel (SNF) of WWER-1000 was researched. β- and γ-radiation from 60 of the most active fission products, and α-radiation from 20 actinides for the fuel with the initial burnup of 41.5 (MW·d)/t U and the cooling time of 5 years was considered. Contribution of radionuclides and actinides to the SNF heat release was determined for the storage time up to 50 years. It is shown that in this time range β- and γ-emitters (fission products) make the main contribution to the heat release, the contribution of β-radiation exceeding that of γ-radiation beginning with 1.3 times initially to 1.8 times at the end of the SNF storage. If the storage time is more than 50 years, the main contribution is made by α-emitters (actinides). Within the storage time of 0…10 years the heat release decreases quicker than the SNF activity, and after 10 years, the heat release decreases slower than the SNF activity.

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INTRODUCTION

One of the unsolved problems in nuclear power engineering is the storage of spent nuclear fuel (SNF). Currently, for intermediate storage (up to 50 years) the technology of the SNF dry storage is widely used. In Ukraine the SNF of WWER-1000 reactors is stored using dry method at Zaporozhye NPP [1], and it is planned to build a dry storage for spent nuclear fuel (DSSNF) for other nuclear power plants in the Chernobyl exclusion zone.

The technology of SNF dry storage that is employed at Zaporozhye NPP includes the following stages:

a) loading SNF into hermetic containers filled with an inert gas;

b) placing baskets with spent nuclear fuel in ferroconcrete containers that would provide radiation shielding and heat removal (due to natural air convection);

c) placing containers at the storage site.

One of the conditions to ensure safety of SNF storage is limitation of heat release of the stored fuel. The amount of fission products and actinides in SNF and, consequently, heat release of the spent fuel is determined by the level of fuel burnup and the time of its cooling in spent fuel pools at the reactor.

In papers [2–4] the contribution of certain nuclides and actinides to SNF heat release at long-term storage (up to 1000 years) in geological formations was analyzed. Contribution of different radionuclides to the heat release caused by β- and γ-radiation at SNF intermediate storage by the dry method has not been investigated.

The aim of this work is to determine the contribution of SNF radionuclides of WWER-1000 reactors to heat release due to α-, β-, and γ-radiation at fuel storage by the dry method.

CALCULATION METHOD AND SOURCE DATA

The SNF heat release is determined by the energy discharged by β- and γ-radiation of fission products and by α-particles emitted by the actinides. Gamma-rays and α-particles have discrete, and β-particles – continuous energy spectrum. Therefore, the energy release of the l-th radionuclide caused by α- and γ-radiation is determined by the relations [5]:

$$\mathcal{E}_l^i(t) = N_l(t)\lambda_l \sum_k E_{lk}^i n_{lk}^i , \qquad (1)$$

where $i = \alpha$, and γ $N_l(t)$ is the number of the nuclide nuclei l; λ is the constant of l-th nuclide decay; E^{γ}_{lk} is k – line energy of l-th nuclide; E^{α}_{l} is energy of k-th α-transition of l-th nucleus; n^{γ}_{lk} is k-th line quantum yield of l-th nuclide; n^{α}_{lk} is intensity of k-th α-transition of l-th nucleus.

The sum in relation (1) is the total energy E^{γ}_{l} generated by γ-rays or α-particles (E^{α}_{l}) by l-th radionuclide per 1 decay. Then relation (1) will be written in the form:

$$\mathcal{E}_l^i(t) = N_l(t)\lambda_l E_l^i . \qquad (2)$$

The energy release of l-th nuclide caused by β-radiation is determined by the relation:

$$\mathcal{E}_l^{\beta}(t) = N_l(t)\lambda_l \int_0^{E_l^{\beta}\max} \frac{dI_l^{\beta}(E)}{dE} E dE , \qquad (3)$$

where $E^{\beta}_{l}\,max$ is maximum boundary energy of β-radiation and $\frac{dI_l^{\beta}(E)}{dE}$ is the total β-spectrum for all the transitions of l-th nuclide. The integral in relation (3) determines the energy of β-radiation of l-th nuclide, so (3) should be rewritten as follows:

$$\mathcal{E}_l^{\beta}(t) = N_l(t)\cdot\lambda_l \cdot E_l^{\beta} . \qquad (4)$$

Then the energy release from all the radionuclides is determined by the relation:

$$\mathcal{E}_{sum}^{\alpha+\beta+\gamma}(t) = \sum_{l_\alpha}\mathcal{E}_{l\alpha}^{\alpha}(t) + \sum_{l_\beta}\mathcal{E}_{l_\beta}^{\beta}(t) + \sum_{l_\gamma}\mathcal{E}_{l_\gamma}^{\gamma}(t) , \qquad (5)$$

where l_α is the number of α-nuclides; l_β (3) is the number of β-nuclides; l_γ is the number of γ-nuclides.

In this paper the radionuclide contribution to the heat release at dry storage of SNF from WWER-1000 reactor at Zaporozhye. NPP has been researched. Here we used the SNF basic characteristics for standard conditions: burnup of 41.5 (MW·d)/t U and cooling time

of 5 years. To calculate the SNF heat release β- and γ-radiation from 60 of the most active fission products and α-radiation from 20 actinides were considered. To determine the radiation characteristics (spectral composition and yields) we used estimated nuclear data based on ENDF/B-VI.8 employing universal shell for processing JANIS database [6].

Tabl. 1 presents characteristics of the highlighted (highest activity level) nuclides of the fission products, whose contribution to the SNF energy release exceeds 0.1%. The nuclides activity, in accordance with the data in [5], is given in Becquerel's per 1 ton of uranium. In accordance with relations (2) and (4) $\varepsilon_l^{\beta+\gamma} = \varepsilon_l^{\beta} + \varepsilon_l^{\gamma}$. The contribution of β- and γ-radiation of l-th nuclide to the energy release is determined by relation $\eta_l^{\beta+\gamma} = \varepsilon_l^{\beta+\gamma}/\varepsilon_{sum}^{\beta+\gamma}$, where $\varepsilon_{sum}^{\beta+\gamma}$ is the total energy release of β- and γ-radiation of the basic nuclides. The portion of energy release caused by β- and γ-radiation of l-th nuclide in the energy release together with the actinides $\eta_l^{\alpha+\beta+\gamma} = \varepsilon_l^{\alpha+\beta+\gamma} / \varepsilon_{sum}^{\alpha+\beta+\gamma}$, where $\varepsilon_{sum}^{\alpha+\beta+\gamma}$ is the total energy release of α-, β-, and γ-radiation of the highlighted nuclides and actinides.

Table 1

Characteristics of the highlighted nuclides of the fission products

Nuclides	E^{β}_l, MeV	E^{γ}_l, MeV	$T_{1/2}$, year	Activity, Bq/t U	ε_l^{β}, MeV/(s·t U)	$\varepsilon_l^{\beta+\gamma}$, MeV/(s·t U)	$\eta_l^{\beta+\gamma}$, %	$\eta_l^{\alpha+\beta+\gamma}$, %
^{85}Kr	0.228	$2.23\cdot10^{-3}$	10.72	4.19E+14	9.56E+13	9.65E+13	0.80	0.68
^{90}Sr	0.196	0	28.50	3.11E+15	6.08E+14	6.08E+14	5.05	4.31
^{90}Y	0.926	$3.06\cdot10^{-8}$	28.50	3.11E+15	2.88E+15	2.88E+15	23.88	20.37
^{106}Rh	1.407	0.206	1.01	7.64E+14	1.08E+15	1.23E+15	10.24	8.74
^{125}Sb	0.084	0.421	2.73	8.19E+13	6.88E+12	4.13E+13	0.34	0.29
^{134}Cs	0.158	1.555	2.06	1.39E+15	2.20E+14	2.39E+15	19.82	16.91
^{137}Cs	0.172	0.000	30.00	4.18E+15	7.18E+14	7.18E+14	5.96	5.09
^{137m}Ba	0.064	0.596	30.00	3.96E+15	2.53E+14	2.62E+15	21.72	18.53
^{144}Ce	0.082	0.016	0.78	6.35E+14	5.23E+13	6.25E+13	0.52	0.44
^{144}Pr	1.206	0.029	0.78	6.35E+14	7.65E+14	7.84E+14	6.51	5.55
^{147}Pm	0.062	$3.46\cdot10^{-6}$	2.69	2.00E+15	1.24E+14	1.24E+14	1.03	0.88
^{154}Eu	0.227	1.242	8.60	3.29E+14	7.45E+13	4.83E+14	4.01	3.42
^{155}Eu	0.046	0.053	4.96	1.57E+14	7.25E+12	1.56E+13	0.13	0.11
				Sum activity	$\varepsilon_{sum}^{\beta}$	$\varepsilon_{sum}^{\beta+\gamma}$		
highlighted				2.077E+16	6.875E+15	1.204E+16	–	–
all 60 nuclides				2.156E+16	6.888E+15	1.206E+16	–	–
highlighted / all, %				3.68	0.18	0.13	–	–

The nuclides, shown in Tabl. 1 (13 highlighted parent and daughter nuclides), determine SNF energy release with accuracy higher than 0.2%. Tabl. 2 lists characteristics of the highlighted actinides. The contribution to the energy of α-radiation of l-th actinide is determined by ratio $\eta\alpha_l^{\alpha} = \varepsilon_l^{\alpha}/\varepsilon_{sum}^{\alpha}$, where $\varepsilon_{sum}^{\alpha}$ is the total energy of α-radiation of the basic actinides. The portion of the energy release due to α-radiation of l-th actinide in the full energy release with β- and γ-radiation taken into account makes: $\eta\alpha_l^{\alpha+\beta+\gamma} = \varepsilon_l^{\alpha+\beta+\gamma} / \varepsilon_{sum}^{\alpha+\beta+\gamma}$.

Table 2

Characteristics of the highlighted actinides

Acti-nides	E_{α}, MeV	$T_{1/2}$, year	Activity, Bq/t U	ε_l^{α}, MeV/(s·t U)	$\eta\alpha_l^{\alpha}$, %	$\eta\alpha_l^{\alpha+\beta+\gamma}$, %
^{238}Pu	5.486	87.7	7.75E+13	4.250E+14	20.04	3.00
^{239}Pu	5.150	2410	1.28E+14	6.588E+14	31.06	4.65
^{240}Pu	5.156	6560	2.04E+13	1.052E+14	4.96	0.74
^{241}Am	5.478	432	4.54E+13	2.487E+14	11.72	1.75
^{242}Cm	6.043	0.446	6.93E+11	4.188E+12	0.22	0.03
^{243}Am	5.264	7360	8.92E+11	4.695E+12	0.25	0.04
^{243}Cm	5.835	30	4.03E+11	2.351E+12	0.11	0.02
^{244}Cm	5.795	18.1	1.16E+14	6.710E+14	31.64	4.73
			Sum activity	$\varepsilon_{sum}^{\alpha}$		
highlighted			3.52E+14	2.071E+15	100.0	14.96
20 Actinides			4.65E+15	2.072E+15	–	–
highlighted / all, %			–	0.06	–	–

From these data it follows that the contribution of the actinides to the energy release of SNF is approximately 15% at the moment of loading fuel to dry storage. Here the energy release is determined by eight basic actinides. It should be noted, that a considerable difference between the activity of eight basic actinides and the total activity is due to high activity of ^{241}Pu ($A_{Pu\text{-}241}$ = 4.295E+15 Bq/t U), but its share in the energy release is insignificant (it makes about 0.026%).

Energy conversion of *α*-, *β*-, and *γ*-radiation into heat occurs at their interaction with the elements of SNF (uranium dioxide – fuel, zirconium – shell of fuel rods, and stainless steel in structural units of fuel assemblies), as well as of metal structural units of hermetic basket for the SNF storage (fuel assembly guides and walls of the basket – steel). The energy release of *α*- and *β*-particles is determined by ionization losses *dE/dx* and depends on the energy of particles, elemental composition of a matter, and, to a large extent, is determined by the type of particles. It is known that the depth of penetration into the matter is maximum for *γ*-rays and minimum for heavy particles. To calculate the range of *α*-particle path the widely used SRIMM package [8] was applied. To estimate the range of *β*-particles path the extrapolated path of electrons [7, 9] was used. The range of *γ*-ray path was estimated from the condition of photon flux attenuation in a hundred times. Fig. 1 shows the dependences of *α*-, *β*-, and *γ*-particle path in uranium dioxide, zirconium and iron on their energy. The energy range 0…7 MeV was chosen, as long as the maximum energy of *α*-particles was E_α = 6.11 MeV (^{242}Cm), of *β*-particles E_β = 3.52 MeV, and of *γ*-rays – 3.4 MeV (^{106}Rh).

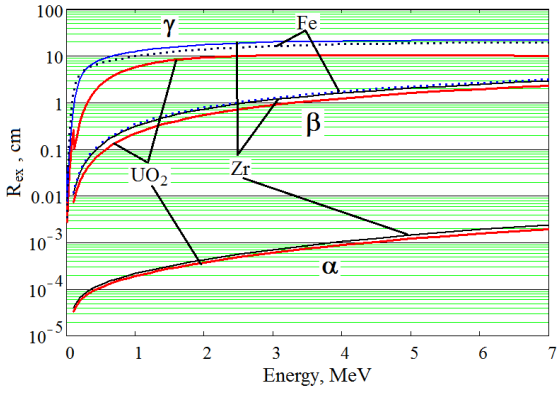

Fig. 1. Dependence of α-, β-, γ-particles on the energy in uranium dioxide, zirconium and iron

From the data shown in Fig. 1, it follows that *α*-particles are absorbed mainly directly in uranium dioxide, and *α*-particles emitting from the surface of "pellets" made of UO_2, are absorbed in the fuel rod shell made of zirconium. Beta particles are mostly absorbed in the fuel rods (uranium dioxide and zirconium shell), as well as in the steel hexagonal guides for spent fuel assemblies (SFA) (0.6 cm thick Fe) that are placed in a hermetic basket of the storage container. Those *β*-particles, whose path in Fe exceeds 0.6 cm, are absorbed in the walls of the hermetic basket of the storage container (2.5 cm thick Fe). Gama-rays emitting outside the hermetic basket with SFA, were calculated by MCNP package. For this purpose the geometrical module commonly used for calculations of *γ*-ray passage to the surface of the SFA [1] was modified, the biological shield (7.5 cm steel and 70 cm concrete in the radial direction) was removed.

CALCULATION RESULTS

Using relation (5) and the nuclides characteristics from Tables 1 and 2 we defined the SNF energy release for certain nuclides with α-, *β*-, and γ-radiation for an arbitrary point of time taken into account. The portion of energy release of γ-rays leaving the storage container "basket" and not causing SNF heating was taken into account. The calculation of the energy release from the actinides showed that the electrons with discrete energies produced at their decay, as well as gamma-rays, and x-rays made an insignificant contribution as compared to that of α-particles. Considerable contribution is made by generation of ^{241}Am (daughter nuclide) from ^{241}Pu (parent nuclide) due to *β*-decay. The relation for the number of atoms of americium produced by plutonium at the moment of time *t* is presented in the form [10]:

$$N_{\mathrm{Am}}(t) = N_{\mathrm{Pu}}(0)\cdot\frac{\lambda_{\mathrm{Pu}}}{\lambda_{\mathrm{Pu}}-\lambda_{\mathrm{Am}}}\cdot\left[\exp(-\lambda_{\mathrm{Pu}}t)-\exp(-\lambda_{\mathrm{Am}}t)\right], \quad (6)$$

where $N_{\mathrm{Pu}}(0)$ is the number of atoms of ^{241}Pu at the initial moment, λ_{Pu} and λ_{Am} are constants of plutonium and americium decay.

An important feature of the radionuclide mixture is the variation of their average energy in time that is defined by the relation:

$$E^i_{\mathrm{av}}(t) = \sum_l N_l(t)\lambda_l E^i_l \Big/ \sum_l N_l(t)\lambda_l, \quad (7)$$

where i = α, β, γ.

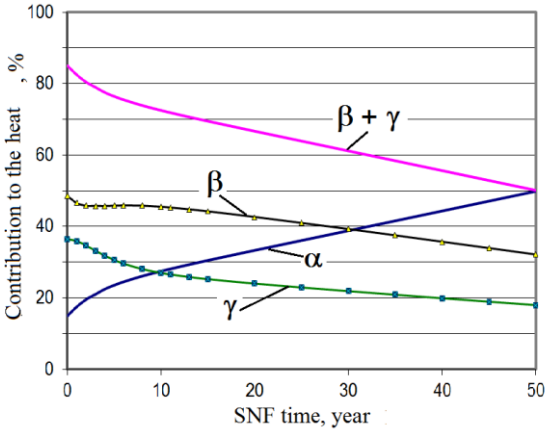

Fig. 2. Dependence of α-, β-, and γ-emitters contribution on the SNF cooling time

The calculations made in MCNP package showed that the portion of *γ*-rays that left the "basket" was about 2% of their total energy release at the initial moment of time.

With the increase of the cooling time the portion of such gamma-rays decreases. Fig. 2 presents the dependencies of the contribution of SNF from *α*-, *β*-, and *γ*-emitters to the heat release on the SNF cooling time.

For the cooling time up to ~ 30 years maximum contribution to the heat release is made by *β*-emitters. The total heat release of the fission products (*β*- and

γ-emitters) is greater than that of the actinides for the cooling time up to ~ 50 years, after 50 years the dominant factor of contribution to the energy release is α-radiation of actinides.

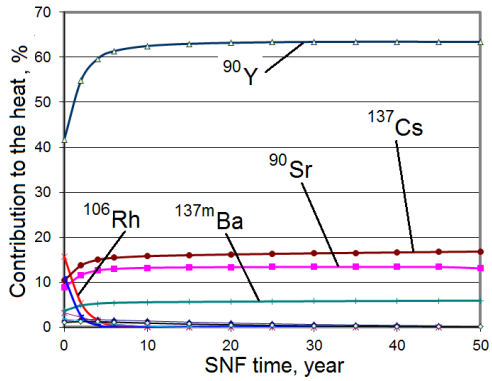

Fig. 3a. Contributions of certain nuclides to the energy release caused by β-radiation

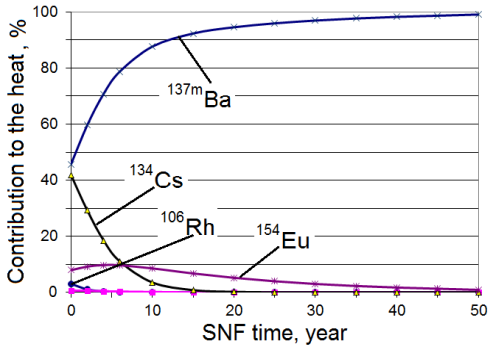

Fig. 3b. Contributions of certain nuclides to the energy release caused by γ-radiation

From these data it follows that the main contribution due to β-radiation to SNF heat release is made by two radionuclides: ^{90}Sr and its daughter nuclide ^{90}Y, and ^{137}Cs and its daughter nuclide ^{137m}Ba. Heat release due to γ-radiation at short cooling time (up to ~ 20 years) is determined by three radionuclides: ^{137m}Ba, ^{134}Cs, and ^{154}Eu and at longer time – by ^{137m}Ba (Figs. 3a, 3b).

Fig. 4 presents the contributions of highlighted actinides to the energy release caused by α-radiation depending on the SNF cooling time. The heat release caused by α-radiation for the cooling time up to ~ 30 years is determined by four actinides: ^{238}Pu, ^{239}Pu, ^{241}Am, ^{244}Cm, and for longer cooling time – by three nuclides: ^{238}Pu, ^{239}Pu, ^{241}Am, with ^{241}Am as a dominant.

In general, due to rather quick decrease in the activity of β- and γ-emitters (fission products), which make the major contribution to the SNF heat release, for the first 10 years of SNF storage by dry method, the SNF heat release decreases in ~ 2 times (0.56 from the initial one), the same occurs with the activity of the major heat-generating nuclides. The dependence of heat release and radionuclide activity on the cooling time is shown in Fig. 5. Within 0…10 years of SNF storage the heat release decreases quicker than the SNF activity. This is due to the fact that at the cooling time interval of 0…5 years the average energy of β- and γ-radiation decreases significantly (Fig. 6), the contribution of β- and γ-emitters in this time interval is maximum (see Fig. 2). At the cooling time over 10 years, the SNF heat release decreases more slowly than its activity. This is due to the growth of contribution of the actinides, whose half-time is longer than that of the fission products, to the SNF heat release.

Note, the good agreement of the data on the SNF heat release depending on the cooling time in our paper with the data described in [11], and with the data using the package modules SCALE-SAS2H and TRITON at Oak Ridge National Lab, USA [12]. The accuracy of these data agreement is up to 1%.

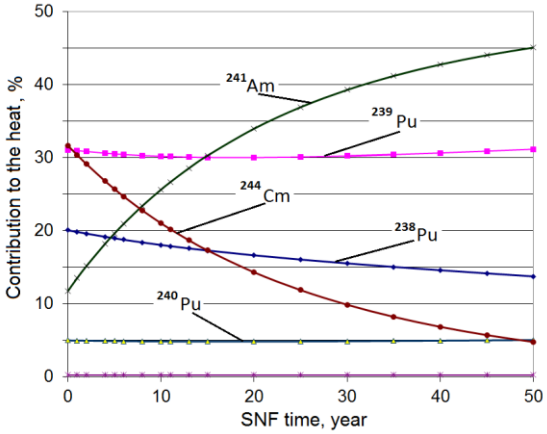

Fig. 4. Contributions of certain actinides to the energy release due to the α-radiation

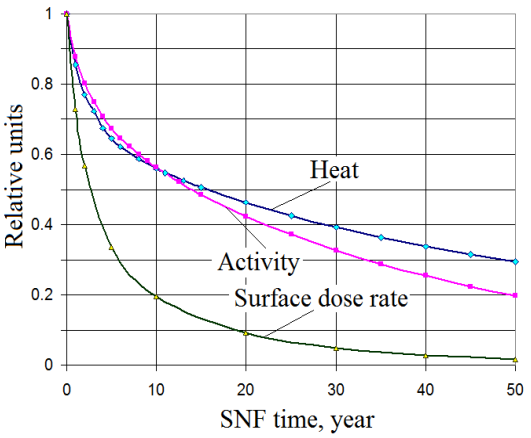

Fig. 5. Dependence of the heat release and of SNF radionuclide activity on the cooling time

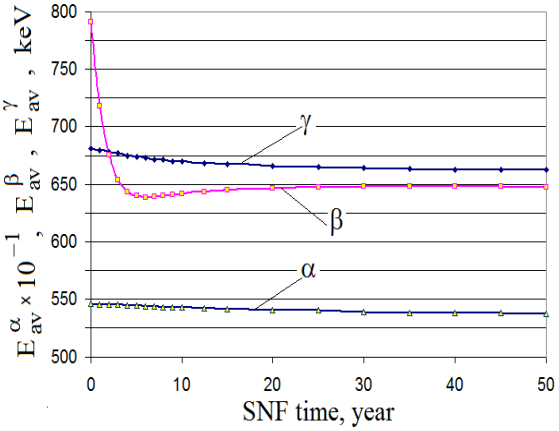

Fig. 6. Dependence of the average energies of α-, β-, and γ-radiation of the highlighted actinides and nuclides on the SNF storage time

We should also note a significantly more rapid drop in the dose rate on the surface of the container with SNF depending on the storage time, as compared to the change in heat release, described in [13]. This is due to the fact that the dose rate on the container surface is produced by high-energy γ-rays emitted by radionuclides, whose half-life is short.

CONCLUSIONS

The radionuclide contribution to the heat release at dry storage of SNF from WWER-1000 was studied. β- and γ-radiation from 60 of the most active fission products, and α-radiation from 20 actinides for the fuel with an initial burnup of 41.5 (MW·d)/t U and cooling time of 5 years was considered. The contribution of radionuclides and actinides to SNF heat release for the storage time up to 50 years was determined. It is shown that in this time range the main contribution to the heat release is made by β- and γ-emitters (fission products). The contribution of β-radiation is determined by ^{90}Sr (^{90}Y) and ^{137}Cs (^{137m}Ba). Heat release caused by γ-radiation for short cooling time (up to ~ 20 years) is determined by three radionuclides: ^{137m}Ba, ^{134}Cs, and ^{154}Eu and for longer time – by ^{137m}Ba. The contribution of β-radiation is 1.3 times higher initially and 1.8 times higher at the end of the SNF storage than that of γ-radiation. At the storage time of more than 50 years the major contribution is made by α-emitters (actinides) ^{238}Pu, ^{239}Pu, ^{241}Am. Within the storage time of 0…10 years the heat release decreases quicker than the SNF activity, and after 10 years the heat release decreases slower than the SNF activity.

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ВКЛАД РАДИОНУКЛИДОВ В ТЕПЛОВЫДЕЛЕНИЕ В ПРОЦЕССЕ СУХОГО ХРАНЕНИЯ ОЯТ

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Исследован вклад радионуклидов в тепловыделение при сухом хранении ОЯТ ВВЭР-1000. Учитывались β- , γ-излучения от 60 наиболее активных продуктов деления и α-излучение от 20 актиноидов для топлива с начальным выгоранием 41,5 (МВт·сут)/т U и временем выдержки 5 лет. Для времен хранения до 50 лет определены вклады радионуклидов и актиноидов в тепловыделение ОЯТ. Показано, что в этом временном диапазоне основной вклад в тепловыделение вносят β-, γ-излучатели (продукты деления), вклад β-излучения превышает вклад γ-излучения от 1,3 в начале до 1,8 в конце хранения ОЯТ. При времени хранения свыше 50 лет основной вклад вносят α-излучатели (актиноиды). В диапазоне времен хранения 0…10 лет уменьшение тепловыделения происходит быстрее, чем изменение активности ОЯТ, а в дальнейшем тепловыделение спадает медленнее, чем активность ОЯТ.

ВНЕСОК РАДІОНУКЛІДІВ У ТЕПЛОВИДІЛЕННЯ В ПРОЦЕСІ СУХОГО ЗБЕРІГАННЯ ВЯП

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Досліджено внесок радіонуклідів у тепловиділення при сухому зберіганні ВЯП ВВЕР-1000. Враховувалися β- , γ-випромінювання від 60 найбільш активних продуктів поділу та α-випромінювання від 20 актиноїдів для палива з початковим вигоранням 41,5 (МВт·сут)/т U і часом витримки 5 років. Для часів зберігання до 50 років визначені вклади радіонуклідів і актиноїдів у тепловиділення ВЯП. Показано, що у цьому часовому діапазоні основний внесок у тепловиділення вносять β-, γ-випромінювачі (продукти розподілу), вклад β-випромінювання перевищує внесок γ-випромінювання від 1,3 спочатку до 1,8 у кінці зберігання ВЯП. У разі часів зберігання понад 50 років основний внесок вносять α-випромінювачі (актиноїди). У діапазоні часів зберігання 0…10 років зменшення тепловиділення відбувається швидше, ніж зміна активності ВЯП, а в подальшому тепловиділення спадає повільніше, ніж активність ВЯП.