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V. I. Borysenko, V. V. Goranchuk, N. M. Sidoruk

Institute for Safety Problems of Nuclear Power Plants NAS of Ukraine, Lysogirska str., 12, Kyiv, 03028, Ukraine

vborysenko@ispnpp.kiev.ua

FORECASTING CHANGES IN THE NEUTRON-PHYSICAL CHARACTERISTICS OF FUEL - CONTAINING MATERIALS

The paper presents the results of calculations for determining neutron-physical characteristics (NFC) of fuel-containing materials (FCM), including the possibility of formation of critical masses. The lack of reliable information at present about the material composition of some places of FCM requires additional studies to substantiate the nuclear safety of such clusters of FCM. An important and, in some cases, the only, method of investigating nuclear safety of places of FCM accumulation is the calculation method for determining the criticality parameters of FCM and predicting the change of neutron-physical characteristics important for nuclear safety during the planned time of FCM location in the «Ukryttya» object. Ensuring a reliable control of the intensity of fissions in FCM, as well as clarifying the supplementary cementitious material composition, requires the introduction of additional methods and channels for monitoring FCM, for example, on the activity of air and water environments determined by short-lived products of fission of nuclear fuel in FCM. The paper presents information on the predicted change in the neutron-physical characteristics of FCM for 120 years from the time of formation of FCM. In particular, the following characteristics are considered: neutron activity; alpha-activity; power of residual energy releases; change in the ratio of main alpha and neutron emitters; the effect of changes in other characteristics of FCM on subcriticality, as well as the conditions and configuration of possible criticality in FCM. For a long period of "planned" storage of FCM in the «Ukryttya» object facility, some NFC FCM will undergo significant changes. The main decline in residual energy releases, and, accordingly, FCM activity, including neutron activity, occurred in the first years after the accident. After more than 30 years, the rate of change of these characteristics of FCM significantly decreased.

Keywords: neutron-physical characteristics, fuel-containing materials, criticality parameters, nuclear safety, «Ukryttya» object.

The code system

To calculate the NFC FCM, the SCALE code [1] was applied, as well as the analytical dependencies between the amount of nuclide and its activity.

The SCALE software system was developed at the Oak Ridge National Laboratory of the United States, commissioned by the US Nuclear Regulatory Commission. SCALE is a software tool for analyzing nuclear safety and designing fuel-based systems. The first version of the SCALE was released in 1980, and since then it has been widely used both in the US and abroad to perform analyzes of criticality, radiation safety, heat transfer, burnup [2].

The SCALE software complex was used to justify the nuclear safety of the Spent Fuel Storage Facility (SFSF) of the ZNPP, SFSF-2 of the ChNPP and the Central Spent Fuel Storage Facility (CSFSF) [3]. In accordance with [2] SCALE is used to substantiate the nuclear safety of spent nuclear fuel (SNF) storage systems in many countries, including: Bulgaria, Germany, Hungary, Slovakia, Sweden, USA, Japan.

Validation of the radionuclide concentration determination model for fuel burnup.

The estimation of the number of radionuclides in RBMK nuclear fuel at the time of the accident was carried out by simulating the operation of the RBMK fuel channel for 700 effective days at a capacity of 2 MW, which corresponds to fuel burn-out of ~ 11 MW·day/kg U, defined as the average fuel burnup at the time Accident [4, 5]. Validation of the calculation model was performed by comparing the calculated and experimental values [6] of the concentration of the determining radionuclides for the fuel burn-out depth RBMK-1000 ~ 11 MW·day/kg U.

Residual energy releases of FCM

In Fig. 1 shows the calculated values of the change in the contribution of β-, γ- and α-emitters to the power of residual energy releases. The contribution to the power of residual energy releases of FCM from α-emitters is now less than 10 %, and the contribution from stimulated fission caused by spontaneous neutrons and neutrons (α, n) reactions is insignificant and can be ignored. Analytical assessment of the contribution to the residual energy release of FCM from the forced fuel division into FCM is presented below.

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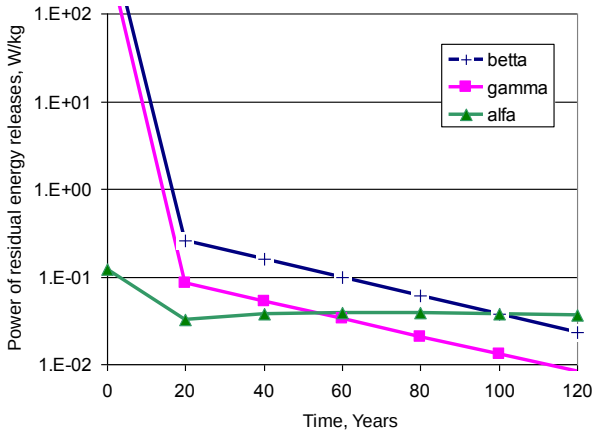

Fig. 1. Changes in the contribution of β-, γ- and α-emitters to the power of residual energy releases.

Change in the activity of α-emitters

In contrast to the intensity of β- and γ-emitters, which are continuously decreasing after their accumulation in the spent fuel, the intensity of the α-emitters after the decay, in the first years after "unloading" the fuel from the reactor, grows up to a 60-year storage life, and then begins continuous decline. In Fig. 2 graphs of the activity of the main α-emitters are presented.

Despite the high activity of the α-emitters, their contribution to the power of the residual energy releases is currently less than 0.04 W/kg, which is equivalent to ~ 10 % of the total residual energy release, but over time, the contribution of α-emitters to the total power of the residual energy releases increases, and 110 years later will exceed the relative contribution from the other radiators. At the same time, the total power of residual energy releases throughout the entire period is constantly decreasing (see Fig. 1).

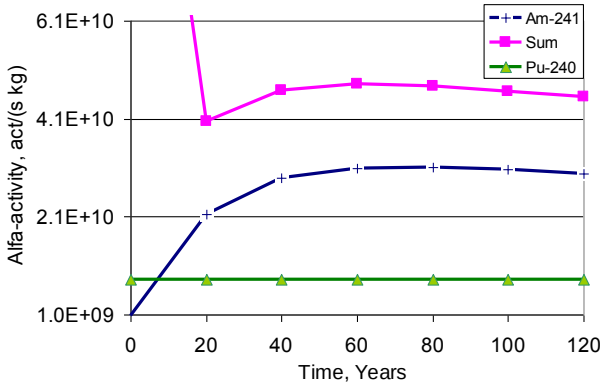

Fig. 2. Change in the α-activity of FCM

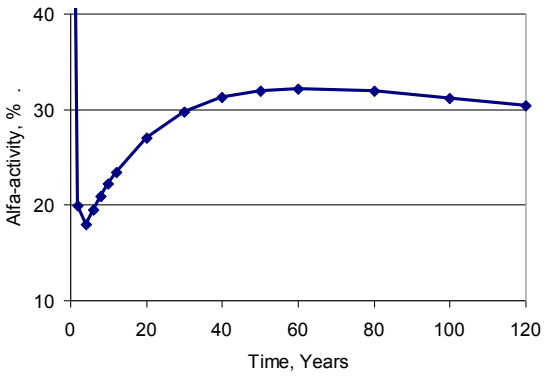

Fig. 3. The change in the total α-activity of FCM.

The change in the total α-activity is shown in Fig. 3. After reaching an intermediate minimum of ~ 20 % of the initial α-activity of FCM, the relative increase in α-activity after 60 years from accident will reach its maximum ~ 32 % of the initial α-activity of FCM, after which there will be a continuous decrease.

The contribution of spontaneous fission neutrons to the residual energy release of FCM

Consider the algorithm for determining the upper estimate of energy release from fission of fuel nuclei in FCM, based on the following provisions.

For today (~ 30 years after the accident), the estimated neutron activity of fuel due to spontaneous fission neutrons and neutrons from the (α-n) reaction on light nuclei is ~ $2.5 \cdot 10^6$ n/t (see Fig. 4). At K_{ef} = 0.999, ignoring the neutron loss, we get that the maximum number of neutrons produced in the subcritical state is $2.5 \cdot 10^9$ n/t. If even ALL neutrons cause the fission of nuclear fuel - this will correspond to – $2.5 \cdot 10^9$ acts of fissions per ton of fuel. Considering that 200 MeV of energy is released in the fission event, $3.1 \cdot 10^{10}$ fissions/s are needed for 1 W power. Thus, the contribution from fission to energy release of FCM is: 0.082 W/t.

Variations in the power of the neutron source to $1.0 \cdot 10^7$ n/t, as well as other "assumptions", can give an upper estimate of the power output from fission to 1 W/t, which is less than 0.5 % of the residual energy release on "today".

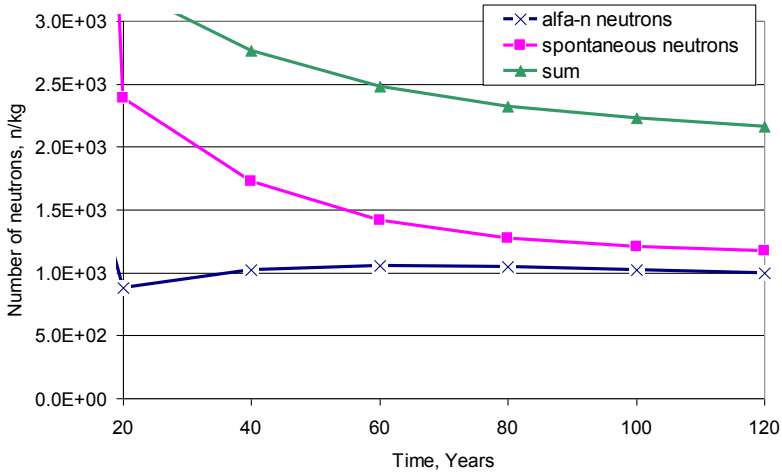

Fig. 4. Contribution to neutron activity of FCM from spontaneous fission neutrons and neutrons (α, n) reactions.

Some authors [7] use another algorithm for determining the forced fission power in a subcritical reactor, which requires additional "justifications". For example, it is necessary to point out the incorrect application in formula (2) [8] (analogue of formula (1) of the present paper) instead of the lifetime of the instantaneous neutrons l_{inst}, the effective lifetime of the neutrons l_{ef}.

$$\varphi = n \cdot v = \frac{Q \cdot \rho}{1 - K_{ef}} \cdot \ell_{inst} \cdot v \quad (1)$$

Let us consider why such a replacement is erroneous. From the definition:
FLUX OF NEUTRONS - $\varphi = n \cdot v$, is the product of the neutron density (n/cm^3) of a given energy and the neutron velocity (cm/s) of a given energy.

Despite the deceptive dimension of n/(cm^2·s), these are not neutrons in cm^2 per 1 s, but, in accordance with the definition - the neutron flux - the total path traveled by neutrons of a given energy, in 1 cm^3 per 1 s.

Thus, a neutron can contribute to the neutron flux only when it "lives" and has the corresponding energy. To determine the thermal neutron flux at an energy of 0.0253 eV, and, correspondingly, the velocity v = 2200 m/s, it is necessary to know how long this thermal neutron lives at an "average" velocity v = 200 m/s. This time corresponds to the neutron diffusion time. Therefore, in formula (1) there should not be $l_{inst} = l_{mod} + l_{dif}$, where l_{dif} is the diffusion time of a thermal neutron, and l_{mod}, is the moderation time of a neutron, which can be neglected in this analysis. Usually, the condition that $l_{mod} < l_{dif}$ is fulfilled, therefore the formula (1) is acceptable.

If we consider the effect of delayed neutrons on the formation of the thermal neutron flux, then they practically do not differ in any way from the instantaneous neutrons. Namely, they have the same diffusion time as the instantaneous neutrons - therefore, there is no difference in the contribution to the neutron flux between instant neutrons and delayed neutrons.

The delayed neutron contributes to the neutron flux only when it "lives", i.e. already emitted from the precursor nucleus and slowed down to thermal energies, and by formula (2) [8] it turns out that the delayed neutron already "forms" the neutron flux, while still in the precursor nucleus - which does not correspond to the neutron flux definition.

It should also be noted that as the degree of subcriticality increases, the neutron lifetime will decrease due to an increase in neutron absorption macro cross-section.

A correct application of formula (1) gives:

$$\varphi = n \cdot v = \frac{Q \cdot \rho}{1 - K_{eff}} \cdot \ell_{inst} \cdot v = \frac{1}{1 - 0.999} \cdot 10^{-4} \cdot 2.2 \cdot 10^5 = 22000 \frac{n}{\text{cm}^2 \cdot \text{s}}.$$

The macro-section of the ^{235}U fission can be obtained based on the conditions that the density of the FCM is 3 g/cm^3, the uranium content is 10 %, the enrichment is 1.15 %, respectively: Σ_f = 0.00509 cm^{-1}.

Accordingly, the number of fission reactions (the upper bound)

$$R = \varphi \cdot \Sigma_f = 112 \frac{fis}{\text{cm}^3} \quad .$$

And the number of fission reactions in 1 m^3 = $1.12 \cdot 10^8$, or the number of fission reactions per 1 ton of fuel = $3.7 \cdot 10^8$. Thus, the contribution to the energy release from stimulated fission in FCM caused by the spontaneous fission neutron flux does not exceed 0.1 W/t of fuel, which correlates well with the maximum estimation of the forced fission power given above.

Nuclear Safety of FCM

A large number of publications [4, 5] is devoted to the nuclear safety of places of FCM clusters.

Let us consider the conservative conditions under which criticality can be attained in the various locations of FCM clusters in the chemical composition.

In accordance with the accepted classification, the main FCM can be divided by chemical composition into brown and black ceramics [4, 5, 9].

In Fig. 5 shows the dependence of the neutron multiplication factor K_{ef} on the water volume content for homogeneous FCM with the following parameters: density 3 g/cm^3, mass content UO_2 – 60 %, enrichment according to ^{235}U – 1.15 %.

Figures 6 and 7 show the dependence of the neutron multiplication factor on the water volume content for heterogeneous FCM with the following parameters: the volume filled with cylindrical fuel rods located in 2.0 cm steps is a cylinder of radius 2 m and height 1 m surrounded by a concrete reflector thickness 0.5 m, enrichment at ^{235}U – 1.15 %. The density and size of the fuel pellet are chosen on the basis that the mass fraction of fuel in the simulated volume remains constant and is 60 %.

In Fig. 6 simulation data are presented for the case when only fuel and water are present in FCM. Fig. 7 shows the modeling data for the case when there are some other materials in FCM, in addition to fuel and water, according to [9].

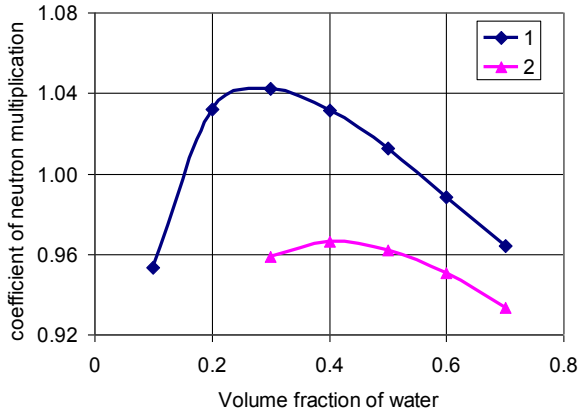

Fig. 5. Change in the coefficient of neutron multiplication in homogeneous FCM: *1* - for an infinite FCM medium; *2* - for a cylinder with a radius of 2 m, and a height of 1 m, surrounded by a concrete reflector 0.5 m thick.

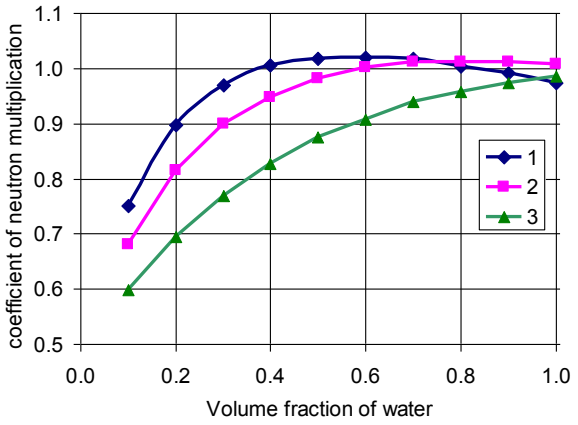

Fig. 6. The change in the multiplication factor of neutrons in heterogeneous FCM, consisting of fuel (UO_2) and water. (Tablet radius and density UO_2, respectively: 1 - 5.75 mm, 10.0 g/cm^3, 2 – 7.5 mm, 5.88 g/cm^3, 3 – 9.0 mm, 4.084 g/cm^3.)

Thus, it can be concluded that for the material composition of black and brown ceramics, under the assumption of a homogeneous model of the multiplying medium, it is impossible to obtain conditions for achieving criticality. Criticality in a homogeneous model of the multiplying medium at a ceramic density of 3 g/cm^3 can be achieved starting with a mass content of 60 % in fuel, which is not yet found in any of the fuel core samples.

For the case of a heterogeneous model of the multiplying medium, the criticality estimates are sufficiently identical.

In Table the simulation results for determining the minimum cylinder size consisting of fuel rods with a diameter of 11.5 mm and surrounding water are presented. Fuel rods are presented in the form of tablets UO_2 enrichment of 1.15 %, density of 10 g/cm^3.

The dependence of K_{ef} on the radius of the multiplying medium 1.0 m high, consisting of fuel cells arranged along a triangular lattice

Cylinder radius, m	0.5	0.55	0.60	0/65	0.75	1.00	1.50
K_{ef}	0.9937	1.0015	1.0089	1.0149	1.0227	1.0356	1.0437

Fig. 8 shows the results of modeling to determine the most optimal step in the location of RBMK fuel to achieve the greatest K_{ef}.

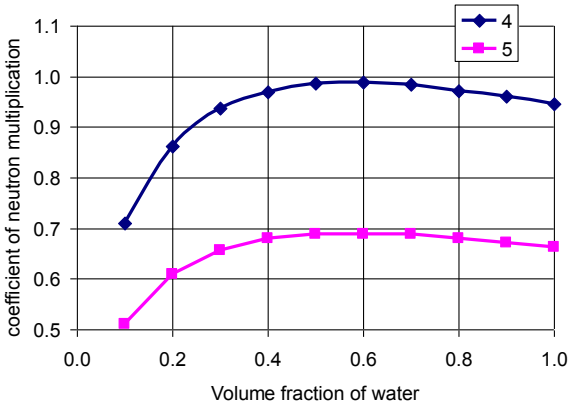

Fig.7. The change in the multiplication factor of neutrons in heterogeneous FCM, depending on the density of water in the interstitial space. The radius of the fuel pellet is 5.75 mm. In the figure, numbers indicate the simulation results for conditions with different fuel tablet compositions: 4 - 60 % UO_2, 31 % SiO_2, 5 - 45.7 % UO_2, 31 % SiO_2, 4 % Ca, 4 % Zr, 4 % Na, 3 % Mn, 2.3 % Mg.
The density of the tablet material is 10 g/cm^3.

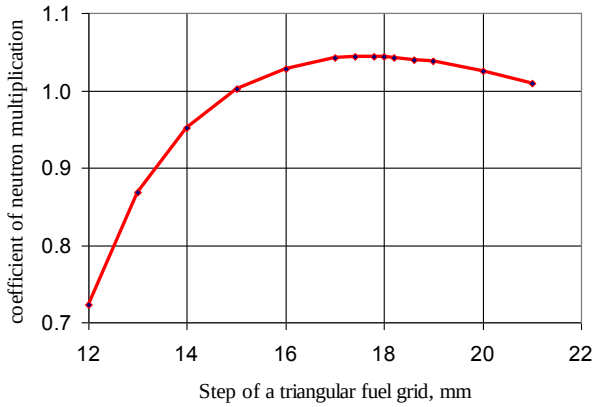

Fig. 8. The dependence of K_{ef} on the pitch of the fuel grid.

Conclusions

The results of the predictive modeling of the change in important NFC FCM show that during the period of forced storage of FCM in the «Ukryttya» object, the continuous decline is observed in terms of residual energy release, β-, γ-activity, and neutron activity. As for α - activity, after a significant decline in the first 4 years after the accident α - activity will increase up to 60 years, after which its continuous decline will begin. The growth of α-activity is determined by the accumulation of ^{241}Am, due to β-decay of ^{241}Pu with a short half-life of ~ 14 years.

The modeling results show that for a multiplying medium with the most conservative parameters, chosen from the experimental determination of the material composition of brown and black ceramics, subcritical conditions are ensured, including for possible flooding with water in an amount optimal for obtaining the maximum multiplying properties of FCM.

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В. І. Борисенко, В. В. Горанчук, М. М. Сидорук

Інститут проблем безпеки АЕС НАН України, вул. Лисогірська, 12, Київ, 03028, Україна

ПРОГНОЗУВАННЯ ЗМІНИ НЕЙТРОННО-ФІЗИЧНИХ ХАРАКТЕРИСТИК ПАЛИВОВМІСНИХ МАТЕРІАЛІВ

Представлено результати розрахунків по визначенню важливих параметрів паливовмісних матеріалів (ПВМ), що визначають їхні нейтронно-фізичні характеристики (НФХ), у тому числі й можливість утворення критичних мас. Відсутність у даний час достовірної інформації про матеріальний склад деяких місць скупчень ПВМ вимагає проведення додаткових досліджень для обґрунтування ядерної безпеки таких скупчень ПВМ. Важливим, у деяких випадках і єдиним, методом дослідження ядерної безпеки місць скупчення ПВМ є розрахунковий метод визначення параметрів критичності ПВМ і прогнозування зміни важливих для ядерної безпеки нейтронно-фізичних характеристик протягом планованого часу знаходження ПВМ в об'єкті «Укриття». Забезпечення надійного контролю за інтенсивністю поділу у ПВМ, а також уточнення матеріального складу ПВМ вимагає впровадження додаткових методів контролю ПВМ, наприклад по активності повітряного й водного середовищ, визначених за короткоживучими продуктами поділу ядерного палива в ПВМ. У роботі представлено інформацію про прогнозовану зміну нейтронно-фізичних характеристик ПВМ протягом 120 років із часу створення ПВМ, зокрема таких як: нейтронна активність, альфа-активність, зміна співвідношення основних нейтронних і альфа-випромінювачів, вплив зміни інших характеристик ПВМ на підкритичність, а також умови й конфігурація можливої критичності ПВМ. За тривалий період «планованого» зберігання ПВМ в об'єкті «Укриття» деякі НФХ ПВМ зазнають істотних змін. Основний спад залишкових енерговиділень, а відповідно й активності ПВМ, включаючи й нейтронну активність, припав на перші роки після аварії. Після більше 30 років швидкість зміни зазначених характеристик ПВМ істотно зменшилася.

Ключові слова: нейтронно-фізичні характеристики, паливовмісні матеріали, параметри критичності, ядерна безпека, об'єкт «Укриття».

В. И. Борисенко, В. В. Горанчук, Н. М. Сидорук

Институт проблем безопасности АЭС НАН Украины, ул. Лысогорская, 12, Киев, 03028, Украина

ПРОГНОЗИРОВАНИЕ ИЗМЕНЕНИЯ НЕЙТРОННО-ФИЗИЧЕСКИХ ХАРАКТЕРИСТИК ТОПЛИВОСОДЕРЖАЩИХ МАТЕРИАЛОВ

Представлены результаты расчетов по определению важных параметров топливосодержащих материалов (ТСМ), определяющих их нейтронно-физические характеристики (НФХ), в том числе и возможность образования критических масс. Отсутствие в настоящее время достоверной информации о материальном составе некоторых мест скоплений ТСМ требует проведения дополнительных исследований для обоснования ядерной безопасности таких скоплений ТСМ. Важным, в некоторых случаях и единственным, методом исследования ядерной безопасности мест скопления ТСМ является расчетный метод определения параметров критичности ТСМ и прогнозирования изменения важных для ядерной безопасности нейтронно-физических характеристик в течение планируемого времени нахождения ТСМ в объекте «Укрытие». Обеспечение надежного контроля интенсивности делений в ТСМ, а также уточнение материального состава ТСМ, требует внедрения дополнительных методов контроля ТСМ, например по активности воздушной и водной сред, определяемых короткоживущими продуктами деления ядерного топлива в ТСМ. В работе представлена информация о прогнозном изменении нейтронно-физических характеристик ТСМ в течение 120 лет с момента образования ТСМ, в частности таких как: нейтронная активность, альфа-активность, изменение соотношения основных нейтронных и альфа-излучателей, влияние изменения других характеристик ТСМ на подкритичность, а также условия и конфигурация возможной критичности в ТСМ. За продолжительный период «планированного» хранения ТСМ в объекте «Укрытие» некоторые НФХ ТСМ претерпят существенные изменения. Основной спад остаточных энерговыделений, а соответственно и активности ТСМ, включая и нейтронную активность, пришелся на первые годы после аварии. По прошествии более 30 лет скорость изменения указанных характеристик ТСМ существенно уменьшилась.

Ключевые слова: нейтронно-физические характеристики, топливосодержащие материалы, параметры критичности, ядерная безопасность, объект «Укрытие».

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