SECTION 2

PROBLEMS OF MODERN NUCLEAR POWER ENGINEERING

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EFFICIENCY OF VARIOUS MATERIALS APPLICATION FOR RADIATION SHIELDING AT TRANSPORTATION AND STORAGE OF SPENT NUCLEAR FUEL BY DRY METHOD

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A model of the transport container intended for transportation of spent nuclear fuel (SFN) is studied. The passage of γ -quanta from the major long-lived isotopes is examined. The radiation shields made of iron, lead and depleted uranium, which are equivalent in mass to the thickness of iron of 15 to 35 cm is considered. The calculations are carried out using the Monte Carlo method (in MCNP and PHITS packages). The change in the characteristics of γ -radiation beyond the shields, made of different materials and with different thicknesses, is determined. For SNF from WWER-1000 with the thicknesses up to ~ 21 cm, the shield made of lead and uranium is shown to be more effective. If the thickness of the shield exceeds ~ 21 cm, then the shield made of iron is more effective. Increasing the thickness of the shields above 25 cm is shown to be inefficient, since the shields mass increases but the dose rate decreases slightly in this case.

INTRODUCTION

The spent nuclear fuel (SNF) is transported from nuclear power plants (NPPs) to the storage facilities (including those with dry method of storage) as well as to the waste processing facilities in airproof containers by rail. The transport containers (TC) must satisfy the conditions of radiation and nuclear safety, and ensure non-exceeding thermal loads and physical shielding in the case of emergency. Currently, there are a lot of TC designs, some of the TC modifications being used as the storage containers. The major TC characteristics which provide the given dose rate outside the container were presented in the review [1]. These characteristics were as follows: dimensions of the radiation shield, the number of fuel assemblies placed in the container, their type, burnup, and storage time.

All the TCs, in which more than ten spent fuel assemblies (SFAs) are located, as a rule, have the mass of above one hundred tons and thicknesses of steel shields from 200 to 300 mm. In some cases, combined steel and lead shields are used. This is true, for example, for TCs produced by the following companies: NAC International S/T Storage Casks, NAC-STC [1], and HOLTEK International HI-STAR 190 UA [2]. In particular, depleted uranium (DU) was investigated as a shielding material. Namely, comparative characteristics of the shielding properties of two options of TCs (with combined shielding of steel plus DU and steel plus lead) were presented in [3] for the case, when the container was loaded with ten or twelve SFAs. The thicknesses of the container shields made of different materials, which attenuate radiation from 24 SFAs on the container surface to the level of 10 mR/h, were given in [4] (Fig. 1).

The DUCRETE material, mentioned in Fig. 1, is concrete filled with DU. The heat, generated by individual SNF isotopes at dry storage from the moment

of the spent fuel transportation from the NPP up to the storage time of fifty years, was investigated in [5].

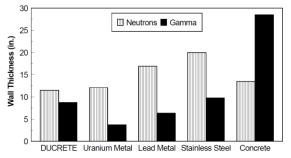


Fig. 1. Thicknesses of container shields made of different materials, which attenuate radiation from 24 SFAs on the container surface to the level of 10 mR/h

The heat transfer from SFA with SNF inside the container was described in [6]. There are practically no data on the contribution of various SNF radionuclides to the dose rate outside the container, and no comparison of the efficiency of the shield materials, subject to their mass characteristics, is given in scientific literature.

The purpose of the present paper are to analyze γ -quanta transport through the shields of different thicknesses and elemental composition, compare the efficiency of the shields made of various materials, subject to the mass characteristics, and determine the individual SNF isotopes contribution to the dose rate.

1. MODEL OF THE RADIATION SHIELD, CALCULATION TECHNIQUE AND INITIAL DATA

The influence of the shield material and thickness on the radiation characteristics of the SNF external radiation is studied in the present paper. The geometry of the model is chosen as close as possible to the

geometry of the TC designed by HOLTEK International HI-STAR 190 UA [2]. It is planned to transport SNF from the Rivne, Khmelnitsky and South Ukrainian NPPs to the central storage facility for spent nuclear fuel (CSFSNF) in the exclusion zone of the Chernobyl NPP in these containers. The radiation source is a cylinder with the diameter of 1.8 m and the height of 4.5 m, filled with homogeneous SNF, surrounded by a cylindrical shield (Fig. 2). The thickness and material of the shield vary in the calculations. One meter high cylinder, placed in the middle of the radiation source, is used as a detecting system of the radiation passed through the shield. The study of the efficiency of radiation shield materials is based on simulation of γ-radiation transport using the Monte Carlo method in MCNP [7] and PHITS [8] packages. The schematic of the TC, used to simulate the SNF radiation transport through the radiation shield in the PHITS package, is presented in Fig. 2.

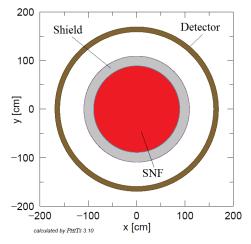


Fig. 2. Schematic of the TC used to simulate SNF radiation transport through the radiation shield in the PHITS package

The developed model allows to vary the thickness and material of the shield for the specified volume of the radiation source (SNF zone). The characteristics of the radiation passed through the shield are determined in the detector. The density of homogenized SNF is assumed to be 3.5 g/cm^3 . The partial contribution of SNF elements, when taking into account the presence of a spacing fence made of steel, is considered to be equal to the following: 0.146 for zirconium, 0.523 for uranium, 0.073 for oxygen, and 0.258 for iron.

To correctly compare the efficiency of the shields in the respect of the attenuation of the γ -quanta flux from SNF as well as to the mass characteristics, the same mass thicknesses are used. Table 1 presents the mass thicknesses of the shields in g/cm² and the corresponding thicknesses of the shields for iron, lead, and uranium in cm. Respectively, the masses of the shields made of different materials and having different thicknesses, but with the same mass thickness, are the same. Further, all the thicknesses of lead and uranium shields are given in cm of the iron thickness for simplicity. In other words, the equivalent thickness of 20 cm for iron, lead and uranium corresponds to their mass thickness of 157.2 g/cm².

Table 1 Mass thicknesses of shields in cm²/g and corresponding thicknesses for iron, lead, and uranium in cm

Material	Fe	Pb	U	Mass thickness, g/cm ²				
Thickness, cm	15	10.39	6.22	117.9				
	20	13.85	8.30	157.2				
	25	17.31	10.37	196.5				
	30	20.78	12.44	235.8				
	35	24.24	14.52	275.1				
Density, g/cm ³	7.86	11.35	18.95	_				

The attenuation of the γ -quanta flux passed through the shield with thickness t_{MAT} is proportional in the ray approximation to the following relation:

$$N_{\gamma} \approx \exp[-\mu_{MAT}(E_{\gamma}) \cdot \rho_{MAT} \cdot t_{MAT}] \times \\ \times F[\mu_{MAT}(E_{\gamma}) \cdot \rho_{MAT} \cdot t_{MAT}].$$
(1)

In (1), ρ_{MAT} , g/cm³ is the product of the material density and thickness; t_{MAT} , cm is the mass thickness which is measured in g/cm²; $\mu(E_{\gamma})$ is the mass attenuation coefficient of the flux of the γ -quanta with the energy E_{γ} in the substance (Compton and photo effects, pair production are taken into account), its dimensionality is cm²/g. The mass thickness of the shield materials in the exponential factor and in the factor, allowing for the accumulation factor, determines the attenuation of the γ -quanta flux. Note, that Monte Carlo method is the only method allowing the correct calculation of shields at thicknesses larger than ~ 10 cm for iron, and at the corresponding mass thicknesses for other elements.

The external radiation, generated by SNF on the shield surface, is determined by initial γ -quanta, emitted by all γ -sources. Usually, in the calculations one considers the spectral composition, which depends on the enrichment, burnup in the reactor (PWR or BWR), and storage time.

The highest γ -activity of SNF with the storage time of more than 3 years is known to be produced by four nuclides, that undergo beta decay followed by γ -quanta emission from daughter nuclides: ${}^{90}\text{Sr} \rightarrow {}^{90}\text{Y}$, ${}^{106}\text{Ru} \rightarrow$ ${}^{106}\text{Rh}$, ${}^{137}\text{Cs} \rightarrow {}^{137}\text{Ba}$, ${}^{144}\text{Ce} \rightarrow {}^{144}\text{Pr}$, and also by 2 isotopes: ${}^{134}\text{Cs}$ and ${}^{154}\text{Eu}$. These six γ -emitters of sufficiently long-lived SNF isotopes determine the characteristics of photon radiation on the outer surface of radiation shield. Table 2 presents the half-lives of the major isotopes, γ -quanta energy E_{γ} , whose quantum yield exceeds 5% of the total one, and the total quantum yields (Yield/1 Bq) for the energies $E_{\gamma} > 0.2$ MeV.

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Isotope	<i>T</i> , year	E_{γ} (yield > 5%), MeV	Yield/1 Bq, ($E_{\gamma} > 0.2 \text{ MeV}$)
⁹⁰ Y (⁹⁰ Sr)	28.5	1.76	0.00016
¹⁰⁶ Rh (¹⁰⁶ Ru)	1.009	0.51; 0.62	0.33
¹³⁴ Cs	2.06	0.57; 0.60; 0.8	2.224
¹³⁷ Cs (¹³⁷ Ba)	30	0.662	0.9
¹⁴⁴ Pr (¹⁴⁴ Ce)	0.778	0.697; 1.49; 2.19	0.0235
¹⁵⁴ Eu	8.6	0.25; 0.72; 0.87; 1.0; 1.27	1.22

Table 2 Characteristics of the major isotopes and quantum yields per 1 Bq

The calculation of characteristics of the radiation, generated by an individual nuclide, allows to determine the external radiation characteristics, including dose loads for an arbitrary concentration of isotopes [9]. Monte Carlo simulation is as follows. The emission of γ -quanta is isotropically sampled over the entire volume of SNF in accordance with the energies and quantum yields for each of the six isotopes. The flux of photons, passing through the shield, is determined on the detecting surface with the photons scattering in the shield and in the SNF zone taken into account. This technique allows to determine the radiation characteristics on the shield surface for the given type of SNF, burnup, and storage time.

2. CALCULATION RESULTS

Note, that the spectrum of the γ -quanta in the flux incident on the inner surface of the shield differs from the initial spectrum of SNF isotopes due to attenuation and transformation (Compton scattering) in the SNF zone. The spectral distributions produced on the inner surface of the shield by photons, which are irradiated by isotopes ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu from the entire SNF region (cylinder diameter of 1.8 m, height of 4.5 m), are shown in Fig. 3. The data, given in Fig. 3, show a significant transformation of the initial discrete spectrum of the SNF isotopes into a continuous spectrum with a significant attenuation of the initial lines intensity.

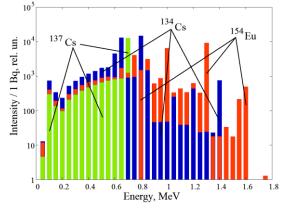


Fig. 3. Spectral distributions produced by photons, irradiated by ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu isotopes from the entire SNF region, on the inner surface of the shield

In the present paper, transport of γ -quanta from six long-lived SNF isotopes, which are the major γ -quanta sources, through cylindrical shields made of iron, lead, and uranium with thicknesses, that are equivalent to the thickness of iron from 15 to 35 cm, is calculated. The spectral distributions of photons irradiated by ¹³⁷Cs isotope after the radiation passes through the shields made of iron and uranium are presented in Figs. 4a, 4b. The spectra for the shield made of iron (15 and 25 cm) are shown in Fig. 4a, and those for uranium (equivalent to the thickness of iron of 15 and 25 cm) – in Fig. 4b. The same figures show the dose rates corresponding to the spectra of γ -quanta passed through the shield. Here, the dose conversion coefficient, given in [10], is used.

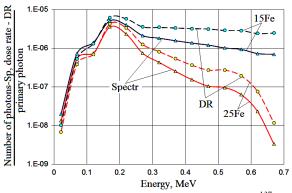


Fig. 4a. Spectra and dose rates, produced by ¹³⁷Cs isotope beyond the shield of iron 15 and 25 cm thick

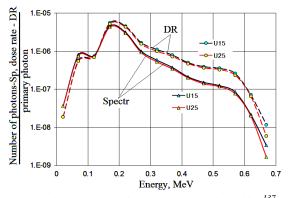


Fig. 4b. Spectra and dose rates, produced by ¹³⁷Cs isotope beyond the shield of uranium, equivalent to iron 15 and 25 cm thick

It follows from the data, given in Fig. 4a, that the increase in the thickness of the shield made of iron from 15 to 25 cm significantly reduces the number of photons with energies above 0.3 MeV. The passage of γ -quanta irradiated by ¹³⁷Cs through the shield made of uranium with the equivalent thickness of 15 to 35 cm practically does not change the spectral distribution, which is close to the spectrum for the shield made of iron 25 cm thick. The calculations have shown, that the spectral composition of photons, which have passed through the shield made of iron more than 25 cm thick, remains practically unchanged as compared to the spectrum for the thickness of 25 cm, and is similar to the spectra of photons, which have passed through the shield made of uranium. The average photon energy is one of the characteristics of photons spectral distributions. The average energies of photons, passed through the shields

made of uranium with equivalent thickness in the range of 15...35 cm and of iron with thickness in the range of 22.5...35 cm, are almost the same and make ~ 0.24 MeV. For Fe with thickness in the range of 15...22.5 cm, the average photon energy varies from 0.33...0.24 MeV.

The data similar to those in Figs. 4a, 4b but calculated for the passage of photons irradiated by ¹⁵⁴Eu isotope through the shields made of iron with thicknesses of 15 and 25 cm are given in Fig. 5a, and in the case of the shields made of uranium with equivalent thicknesses – in Fig. 5b. In contrast to ¹³⁷Cs isotope, whose energy of the initial γ -quanta is 0.662 MeV, the discrete spectrum of the initial ¹⁵⁴Eu photons contains a significant number of γ -quanta with high energies: of 1.27 MeV and higher. The spectra of photons, which passed the shields made of iron with thicknesses from 15 to 25 cm, and equivalent shields made of uranium, are similar. The average energies of photons, which passed through the shields made of iron with thicknesses in the range of 15...35 cm vary from 0.6 to 0.27 MeV, and in the case of uranium (with equivalent thicknesses) - from 0.65 to 0.27 MeV. The average energies become practically equal for the shields made of iron and uranium at thicknesses of 27.5...35 cm.

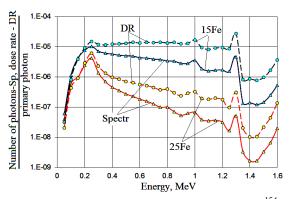


Fig. 5a. Spectra and dose rates, produced by ^{154}Eu isotope beyond the shield of iron 15 and 25 cm thick

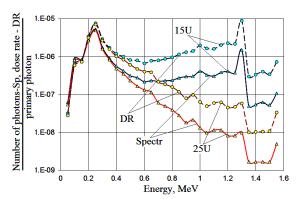


Fig. 5b. Spectra and dose rates, produced by ¹⁵⁴Eu isotope beyond the shield of uranium with equivalent thickness of 15 and 25 cm

It follows from the data, given in Fig. 5b, that an increase in the thickness of the shield made of uranium results in a significant decrease in the number of photons with energies above 0.6 MeV. However, the number of photons and their spectral distributions at energies $E_{\gamma} < 0.4$ MeV remain the same in this case.

The main characteristic of the shield efficiency is the dose rate, generated by SNF on the shield surface. The dependences of the dose rates (DR), generated by γ -quanta irradiated by ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu isotopes and passed through the shields made of iron, lead, and uranium with equivalent thicknesses from 15 to 35 cm are shown in Fig. 6. The DR is given in relative units per 1 Bq with quantum yields for each isotope taken into account. Besides, the DR dependences on the thickness of iron, lead, and uranium shields are calculated for ⁹⁰Y, ¹⁰⁶Rh, and ¹⁴⁴Pr isotopes. The shape of the dependences of DR, generated by ⁹⁰Y and ¹⁴⁴Pr isotopes is similar to that for ¹⁵⁴Eu. However, for ¹⁰⁶Rh isotope it is similar to the DR dependences for ¹³⁷Cs (see Fig. 6). Note, that the DR values for ⁹⁰Y, ¹⁰⁶Rh, and ¹⁴⁴Pr isotopes are less than unity as compared to those for ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu.

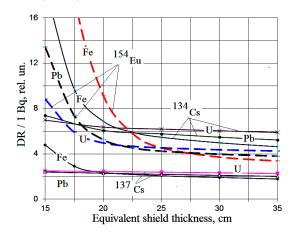


Fig. 6. Dependences of the DRs, generated by γ-quanta irradiated by ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu isotopes, and passed through the shields of iron, lead, and uranium

It follows from the data, given in Fig. 6, that the DRs on the surface of the shields weakly change with thicknesses of the shields made of iron of more than 25 cm as well as lead and uranium shields with equivalent thicknesses. This is explained by the fact that, due to the Compton scattering and absorption of high-energy γ -quanta, the spectrum of photons passed through the shield becomes a low-energy one, with the average energy close to 0.27 MeV. The dependences of the ratios of the contribution of γ -quanta with energies $E_{\gamma} < 0.5 \text{ MeV}$ (DR_{γ}) to the DR produced by photons with all the energies (DR_{Σ}) versus the equivalent thickness of the shield for ^{134}Cs , ^{137}Cs , and ^{154}Eu isotopes are shown on Fig. 7. The smaller contribution of low-energy photons, which passed through the shield made of iron, to DR_{Σ} is due to the large values of the mass attenuation coefficients for γ -quanta in the shield made of lead and uranium. Note, that the radiation characteristics for the case, in which γ -quanta, irradiated by all the six isotopes under study and passed through the shield made of lead, are very close to those for the shields made of uranium after the shield thickness exceeds a certain magnitude.

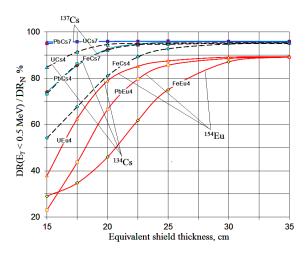


Fig. 7. The ratios of the contribution of γ -quanta with energies $E_{\gamma} < 0.5 \text{ MeV} (DR_{\gamma})$ to the DR produced by photons with all the energies (DR_{Σ}) vs. the equivalent thickness of the shield

The DR on the shield surface is determined by the total DR_{Σ} from all SNF isotopes. The total spectrum of photons for SNF with the given burnup level and exposure time is used in almost all the publications to calculate the characteristics of the external radiation. In the present paper, the technique, developed in [10], is used. This technique allows to calculate the DR_N for the individual nuclides and to determine the total dose rate DR_{Σ} for SNF with an arbitrary burnup level and cooling time for the specified shield characteristics. The ratios of the dose rates subject to the storage time of SNF, generated by individual isotopes, to the total dose rate, DR_N/DR_{Σ} , on the surface of the shield 25 cm thick, made of iron are shown in Fig. 8. The characteristics of the WWER-1000 reactor fuel with the burnup of 55 (MW day)/kg U are used in the calculations. Note, that $^{90}\mathrm{Y}$ isotope contribution to the dose rate DR_{Σ} is less than 0.02%.

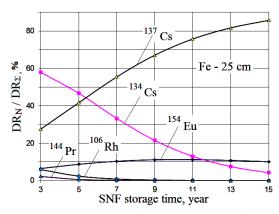


Fig. 8. The ratios of the dose rates, generated by the individual isotopes, to the total DR vs. the SNF storage time

It is obvious, that the DR on the shield surface depends on the isotopes contained in the spent fuel. Their concentration, in turn, depends on the burnup and storage time. Using the calculation results similar to the data in Figs. 6 and 8, the values of DR_{Σ} from the six main isotopes for WWER-1000 SNF are calculated. The

dependences of the total dose rate DR_{Σ} on the equivalent thickness of the shields made of iron, lead, and uranium for WWER-1000 spent fuel with the burnup of 55 (MW·day)/kg U and cooling for 7 years are shown in Fig. 9. The SNF with such characteristics is supposed to be transported to the CSFSNF in the exclusion zone of the Chernobyl NPP [2].

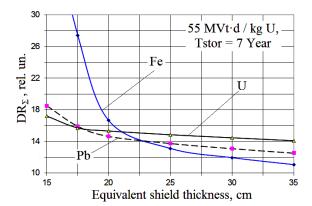


Fig. 9. The total dose rate DR_{Σ} vs. the equivalent thickness of the shields made of iron, lead, and uranium

It follows from the data given in Fig. 9, that the dose rate DR_{Σ} weakly differs for the shields made of iron, lead, and uranium at the shield equivalent thickness of ~ 21 cm. Further increase in the shield thickness results in slight decrease of the dose rate DR_{Σ} . The shape of the DR_{Σ} dependences for other terms of SNF cooling differs little from the dependences shown in Fig. 9. Note, that the shields made of lead and uranium are more effective for small thicknesses (≤ 21 cm), while the shields made of iron are more effective for larger thicknesses. Apparently, this is due to the longer mean free path of γ -quanta in the shield made of iron are larger than those of lead and uranium.

Table 3

Dose rates DR_{Σ} for the equivalent thicknesses of shields made of lead and uranium of 25, 30, and 35 cm, and for the iron shield of 25 cm, as well as the extra

mass of the shield

Thickness,]	$DR_{\Sigma}, \mu Sv$	Extra mass,	
cm	Fe	Pb	U	t
25	100	104.7	113.2	_
30	91.1	99.7	110.3	14.42
35	84.1	95.4	107.5	29.46

Since the dose rate only weakly decreases and the container mass significantly increases with further increase in the shield thickness above 25 cm then such increase of the shield thickness should be considered as inefficient from the point of view of the radiation shielding. The following example demonstrates this below. Let assume the dose rate DR_{Σ} on the surface of the shield made of iron 25 cm thick to be 100 μ Sv/h. The DR_{Σ} values are calculated for the equivalent thicknesses of the shields made of lead and uranium of 25, 30, and 35 cm, and for the iron shield with the

thickness of 25 cm using the data from Fig. 9 (Table 3). The extra mass of the shield, as compared to the thickness of 25 cm, is given in the rightmost column.

CONCLUSIONS

The characteristics of the external radiation beyond the shields of cylindrical geometry, made of different materials but with the same mass characteristics, are compared.

For the initially low-energy γ -quanta with the average energy of ~ 0.24 MeV, irradiated by ¹³⁴Cs and ¹³⁷Cs isotopes, the spectra of the external radiation, produced beyond the shield made of iron 25 cm thick, are shown to be close to the spectra beyond the shields made of lead and uranium with equivalent thickness of 15...35 cm.

For the high-energy photons, irradiated by ¹⁴⁴Pr and ¹⁵⁴Eu isotopes, an increase in the thickness of the shields results in a significant decrease in the number of photons with the energies above 0.6 MeV. In this case, the number of photons and the parts of the spectra are almost the same at the energies $E_{\gamma} < 0.4$ MeV if the shields are made of lead and uranium. The major (> 80%) contribution to the dose rate at the thicknesses above 22.5 cm is shown to be made by the reradiated γ -quanta with energies $E_{\gamma} < 0.5$ MeV.

The contributions of the individual isotopes to the total dose rate and the efficiency of the cylindrical radiation shield, made of different materials with different thicknesses, are determined for WWER-1000 SNF.

The shield made of a lead and uranium is shown to be more effective up to the thicknesses of ~ 21 cm for WWER-1000 SNF. And the shield made of iron appears to be more effective for larger thicknesses. An increase in the equivalent thickness of the shields above 25 cm is ineffective, since it results in a significant increase in the shield mass, but the dose rate decreases weakly.

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ЭФФЕКТИВНОСТЬ МАТЕРИАЛОВ РАДИАЦИОННОЙ ЗАЩИТЫ ПРИ ТРАНСПОРТИРОВКЕ И ХРАНЕНИИ ОЯТ СУХИМ МЕТОДОМ

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В рамках модели транспортного контейнера для перевозки ОЯТ исследовано прохождение γ-квантов от основных долгоживущих изотопов через радиационные защиты из Fe, Pb и обедненного U, эквивалентных толщине Fe от 15 до 35 см. Выполнены расчеты методом Монте-Карло (в пакетах MCNP и PHITS) прошедшего через защиты излучения. Определено изменение характеристик γ-излучения за защитами из разных материалов и толщин. Показано, что для ОЯТ ВВЭР-1000 до толщин ~ 21 см более эффективна защита из Pb и U, а при больших толщинах – защита из Fe. Показано, что не эффективно увеличивать толщины защит свыше 25 см, так как растет вес защит, а мощность дозы уменьшается незначительно.

ЕФЕКТИВНІСТЬ МАТЕРІАЛІВ РАДІАЦІЙНОГО ЗАХИСТУ ПРИ ТРАНСПОРТУВАННІ ТА ЗБЕРІГАННІ ВЯП СУХИМ МЕТОДОМ

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У рамках моделі транспортного контейнера для перевезення ВЯП досліджено проходження γ-квантів від основних довгоживучих ізотопів через радіаційні захисти з Fe, Pb і збідненого U, еквівалентних товщині Fe від 15 до 35 см. Виконано розрахунки методом Монте-Карло (у пакетах MCNP і PHITS) випромінювання, що пройшло крізь захисти. Визначено зміну характеристик γ-випромінювання поза захистами з різних матеріалів і товщин. Показано, що для ВЯП ВВЕР-1000 до товщин ~ 21 см ефективнішим є захист з Pb і U, а при великих товщинах – захист з Fe. Показано, що не ефективно збільшувати товщини захистів понад 25 см, бо зростає вага захистів, а потужність дози зменшується незначно.