THE EFFICIENCY OF RADIATION SHIELDING MADE FROM MATERIALS WITH HIGH ATOMIC NUMBER AND LOW MASS DENSITY

V.G. Rudychev¹ , M.O. Azarenkov¹ , I.O. Girka¹ , Y.V. Rudychev2,1 ¹V.N. Karazin Kharkiv National University, Kharkiv, Ukraine; ²National Science Center "Kharkov Institute of Physics and Technology", Kharkiv, Ukraine E-mail: vrudychev@karazin.ua

The radiation shielding from γ -quanta of the existing transport containers (TC) for transportation of spent nuclear fuel (SNF) is made of steel or steel plus Pb $25...30$ cm thick and weighting $\sim 60...80$ t. The application of materials with high atomic number, dispersed (solids grinded to a powdery state) to the densities in the range $4 < \rho < 8$ g/cm³, is investigated. Simulations based on the Monte Carlo method show that at the densities of dispersed depleted U larger than 5 g/cm³ and shielding thicknesses of more than 30 cm, the absorption of γ -quanta of SNF is greater than that of the shielding made of steel of the same thickness. The application of such materials, while the weight characteristics of the shields are not exceeded, provides radiation shielding for SNF with the high burnup rate and the smaller cooling time or larger amount of the transported SNF.

INTRODUCTION

Nowadays, a transportation of spent nuclear fuel (SNF) to the temporary storage facilities for SNF is an integral part of the nuclear fuel cycle. A significant part of SNF transportation is carried out by railway in transport casks (TC) [1]. The TCs, in which SNF is transported by railway, are complex technical objects which must ensure the radiation and nuclear safety. Spent fuel assemblies (SFAs) are placed in a sealed cylindrical steel tank (basket) filled with an inert gas (helium) to transfer heat from the SFA to the tank walls. Nuclear safety is provided by maintaining the distances between the SFAs and introducing the neutron absorbers (boron, etc.) to the spacer elements. The radiation shielding which surrounds the SNF basket is a massive thick-walled layered internal structure of the container. The shielding is made from a metal, which provides the protection against gamma quanta, and materials in the form of rubber, plastic, etc., which provide the protection against neutrons. This design provides radiation protection and prevents mechanical damage of the sealed basket under possible external influences. The radiation protection of the TC determines the possible amount of SFAs transported in it, the level of burnup and the time spent by SNF in the NPP cooling pools. It should be noted that there are restrictions on weight (it must be within the range 100…140 t) and dimensions of shipping containers (outer diameter must be smaller than 325 cm in for the USA, usually – smaller than 280 cm).

Restrictions on the weight and dimensions have led to the following characteristics [2, 3] of the existing TCs (Table 1).

The efficiency of SNF transportation depends not only on the number of spent fuel assemblies in the fuel cell, but also on the level of SNF burnup – it should be maximal, and the cooling time (time of storage in the NPP cooling pools) should be minimal. Therefore, the efficiency of transportation is practically determined by the radiation protection from SNF gamma quanta, which flow depends on the amount of SFAs, the level of burnup, and the storage time of SNF.

The objective of the present paper is to study options for the gamma shields of SNF. On the one hand, the shields should not exceed the permissible limits in terms of the weight and overall dimensions characteristics. On the other hand, the shields should prevail the existing shields made of steel, cast iron and combined shields made of steel, lead, and depleted uranium in terms of efficiency.

Table 1

Characteristics of existing Transport Containers

1. DESCRIPTION OF THE METHODOLOGY

Analysis of the efficiency of applying the various materials for the SNF gamma shielding was carried out in [4] with taking into account the weight characteristics. To compare the efficiency of the shields in terms of both attenuation of the SNF gamma-ray flux and weight characteristics, the following physical observable was applied in [4]. This observable was mass thicknesses t_M . Its essence can be explained as follows. For different densities of shielding materials ρ_1 but for the same mass thickness t_M , the weight of the shields is the same, while the linear thickness of the shields for different materials is $t = t_M/\rho_1$ cm. Here t_M is in g/cm², and ρ_1 is in g/cm³. The radiation source was a cylinder with the diameter of 1.8 m and the length of 4.5 m. The source was filled with SNF, surrounded by a cylindrical shield with the thickness t and density ρ . The changes in the dose rate (DR) caused by passing of the γ -quanta from SNF through the shields made from Fe, Pb, and U of the same mass thickness are shown in Fig. 1 [4]. The spectrum of γ -quanta corresponds to the spectrum of SNF from the WWER-1000 reactor with a burnup of 55 MW day/kg U and a cooling time (Tstor) of 7 years. The linear thickness (in cm) of the shields made from Pb and U is equivalent to the thickness of the shielding made from Fe.

Fig. 1. Dependences of the dose rate on the thickness of the shields made from Fe, Pb, and U

It is well-known that for materials with a large atomic number Z ($Z = 82$ for Pb, and $Z = 92$ for U), the mass absorption coefficients of γ -quanta $\mu(E\gamma)$ in the entire range of photon energies are greater than that for Fe $(Z = 26)$. That is, for the same mass thickness, the absorption of γ -quanta in shielding made from Pb and U should be greater than in the shielding made from Fe. However, as it is shown in Fig. 1, at large shield thicknesses with real dimensions of the TC (cylindrical geometry), this is true up to a certain thickness of \sim 21 cm; and at larger thicknesses, the shield made from Fe is more effective. This is due to the longer mean free path of γ -quanta in the shield made from Fe, since the geometrical dimensions of the Fe shields are larger than the dimensions of the shields made from Pb and U with the same mass thickness. The mean free path of the -quanta is defined by the relation:

where ρ is the density of the shielding material.

In gamma-ray radiography the *mean free path* of a pencil beam of mono-energetic photons is the average distance a photon travels between collisions with atoms of the target material. It depends on the material and the energy of the photons: $\ell = \mu^{-1} = ((\mu/\rho)\rho)^{-1}$, where μ is the linear attenuation coefficient; μ/ρ is the mass attenuation coefficient and ρ is the density of the material.

Table 2 shows the mean free paths l_{FP} in Fe, Pb, and U (columns 1, 2, and 3) at tabular densities of 7.86, 11.35, 18.9, respectively, for different energies of -quanta. For all photon energies, the mean free paths in Pb and U are shorter as compared with that in Fe, and much shorter for photons with energies < 0.8 MeV. As it follows from eq. (1), the mean free path increases with the decrease in the density of the shielding material. Let assume, for example, that the density of uranium is smaller than the original one 18.9 g/cm³. In Table 2, the mean free paths for uranium with densities in the range from 4 to 7 g/cm^3 , which are smaller than that of Fe, are presented (columns 4–7).

A new design of the transport cask for transporting and/or storing SNF was proposed in [5]. Therein, the gamma radiation shield consisted of coaxially placed steel inner, intermediate and outer construction baskets. The cavities between the cylindrical surfaces of the baskets were filled with radiation-shielding material. The material with the average atomic number larger than 80, which was assumed to be dispersed (solids grinded to a powdery state) to the densities in the range $4 < \rho < 8$ g/cm³, was applied as the filler. The annular cavity between the cylindrical surfaces of the intermediate and outer baskets was filled with the material used for neutron radiation-shielding of SNF, consisting of the elements with a low atomic number (H, C, Al, B). For example, rubber, graphite, etc. could be used.

Fig. 2. The transport cask schematic

Shielding material	Fe	U Ph								
No.	-	2	3	4	5	6	7			
Density ρ , g/cm ³	7.86	11.35	18.9	4.0	5.0	6.0	7.0			
Energy E_{γ} , MeV	mean free paths l_{FP} , cm									
0.3	1.16	0.22	0.10	0.49	0.39	0.32	0.28			
0.5	1.51	0.55	0.27	1.28	1.02	0.85	0.73			
0.8	1.90	1.00	0.52	2.48	1.99	1.66	1.42			
1	2.12	1.25	0.67	3.19	2.55	2.13	1.82			
1.25	2.38	1.51	0.83	3.95	3.16	2.63	2.26			

The mean free paths for Fe, Pb, and U with different densities

2. NUMERICAL RESULTS

The transport of the SNF γ -quanta from the radiation source similar to [4] through cylindrical shields (Fig. 2) is analyzed numerically using the MCNP package [6]. Different shields with different densities and thicknesses made from dispersed Pb, depleted uranium U, depleted uranium dioxide $UO₂$, as well as the shields with initial density of the Fe and U are considered. The long-lived isotopes: ^{134}Cs , ^{137}Cs , and ^{154}Eu are chosen as sources of SNF gamma-rays [7]. In Fig. 3 the spectral distributions produced by SNF isotopes 137 Cs and 154 Eu at the TC surface are shown. Gamma-quanta are considered to pass through the shields made from depleted uranium (marked as U) with a density of 18.9 g/cm^3 , dispersed depleted uranium with a density of 6.0 $g/cm³$ (marked as U(6)), and iron (marked as Fe) with a density of 7.86 $g/cm³$. The thicknesses of the shields for U and U(6) are equivalent to 35 cm of Fe.

Fig. 3. Spectral gamma distributions produced by SNF isotopes ¹³⁷Cs and ¹⁵⁴Eu after passing through the radiation-shield made from U, dispersed U(6) and iron Fe with $\rho = 18.9$ *; 6.0, and 7.86 g/cm³, respectively*

Fig. 4. Spectral gamma distributions on the TC surface produced by SNF isotopes ¹³⁷Cs, ¹³⁴Cs, and ¹⁵⁴Eu after passing through the radiation-shield made from U(6) at different cooling times of SNF (cooling time T in given in years)

As it follows from the data presented, the photon spectra produced by the ^{137}Cs and ^{154}Eu isotopes after passing through the radiation-shield made from Fe and U(6) are practically the same. The photon flux passed through U shield (equivalent to 35 cm of Fe) is less absorbed within the uranium with the density of 18.9 $g/cm³$ than within shields of the same equivalent thickness (35 cm Fe) but with the lower density. It should be noted that the spectra behind the shields made from U, U(6), and Fe are practically the same for each of the 137 Cs and 154 Eu isotopes if the spectra are calculated per unit area.

The spectral composition of the SNF gammaradiation changes with the SNF cooling time due to different half-lives of long-lived isotopes. Isotopes 134 Cs, 137 Cs, and 154 Eu were shown in [7] to make the main contribution to the spectral composition. Fig. 4 shows the change in the spectral compositions at the surface of the fuel cells for photons produced by these isotopes after passing through the shielding of dispersed depleted uranium U(6) at different cooling times of SNF. SNF from WWER-1000 reactors with a burnup of 41.5 MW day/kg U is considered. As it follows from the data presented, the contribution to the spectral composition of high-energy photons from 154 Eu and $134Cs$ is the only one to change in the range of SNF cooling times of 3…15 years. However, their contribution to the total DR is fractions of a percent only.

The shielding materials, made from Pb, U, and uranium dioxide $UO₂$ with densities lower than the tabular density of Fe are used in practice. For these materials, the Monte Carlo method is used to calculate the dependences of the DR produced by SNF for the model of the radiation source and shielding from the flux of quanta presented in Fig. 2. The dependences of the DR on the thickness of the shielding made from U of different densities is presented in Fig. 5. The SNF from a WWER-1000 reactor with a burnup of 41.5 MW day/kg U and a cooling time of 3 years is considered. Dashed curves marked as Fe show DRs for iron shielding. Solid curves marked as $U(4) - U(7)$ correspond to the shielding made from depleted uranium with densities from 4 to 7 $g/cm³$. Note that the thickness of the shielding for uranium with different densities is taken in cm, as well as for Fe. The dependences of the DRs in the range of the shielding thicknesses 25…40 cm is shown in Fig. 5 in zoomed scale. Just this range is observed in existing TCs.

Fig. 5. Dependences of DRs on the thickness of U shielding with different densities. WWER-1000 SNF with a burnup of 41.5 MW day/kg U and a cooling time of 3 years is considered

The obtained data on the dependences of the DRs on the thickness of shielding made from Pb, U, and uranium dioxide $UO₂$ with different densities makes it possible to draw the following conclusion. The radiation shielding made of Fe with standard density is more effective than shielding made from the materials with high atomic number with the density of ≤ 4 g/cm³ in the range of thicknesses from 25 to 40 cm. This is clearly seen from the data shown in Fig. 5, curve U(4), for depleted uranium. For uranium with the densities of 4.5 and 5 g/cm³, DR_U is smaller than DR_{Fe} starting from thicknesses of \sim 35 and \sim 30 cm, respectively. For uranium with densities of 6 and 7 g/cm^3 , DR_U is smaller than DR_{Fe} over all the studied range of shielding thicknesses.

In the following, the protection characteristics of the radiation shields and their weight subject to the thickness of the shielding made from iron and depleted U of different densities are evaluated. The masses of the shields are determined and compared with those for the shields of different thickness made of Fe, which are presented in Table 3, column 1. The inner diameter of the space filled with SNF is 1.8 m, the height of the cylinder with SNF is 4.5 m, and the height of the shielding is 5 m in the calculations. The DR at the surface of the 25 cm thick shield made of Fe is assumed to be 100 μSv/h. The dependences of the DRs subject to the thickness of the shielding are shown in Fig. 5. Table 2 shows the DR values in μSv/h behind the shields with the thickness of t, as well as the masses of the corresponding shields in tons. The data is calculated for the shielding made of Fe and for that made from depleted uranium of different densities. The DR values for U of different densities are marked with the symbols ">" and "<". This means that the DR behind the U shield with the thickness t is greater (">") or less ("<") than the DR behind the Fe shield with the thickness t.

For example, it follows from the data given in Table 2, that in the case of the shield thickness of 35 cm if it is made from depleted U with the density of 4.5 g/cm^3 , the mass of the shield is 53.2 t (column 7), and the DR is 84.2 μSv/h.

Table 3

t,	Fe		U(4)		U(4.5)		U(5)		U(6)		U(7)	
cm	DR	mass	DR	mass	DR	mass	DR	mass	DR	mass	DR	mass
	↑	κ			6	⇁		9	10		12	13
25	100.0	63.3	>138.5	32.2	>121.4	36.2	>104.4	40.3	<97.0	48.3	< 95.3	56.4
30	91.0	77.8	>101.3	39.6	>95.1	44.5	<90.6	49.5	< 88.8	59.4	<88.3	69.3
35	84.5	92.9	>86.6	47.3	<84.2	53.2	<82.8	59.1	<82.2	70.9	<81.9	82.7
40	77.2	108.6	>78.5	55.3	< 75.8	62.2	< 75.4	69.1	< 75.0	82.9	< 74.8	96.8

The DR values behind the shields with thickness t made from Fe and depleted U of different densities, as well as the masses of the corresponding shields

It is interesting to underline for comparison, that for the shield made of Fe with a thickness of 35 cm, the DR has the close value of 84.5 μSv/h, however it is almost nine tons heavier: its mass is of 92.9 t. This result is of a high significance. It demonstrates that the application of the shield made from a depleted uranium instead of the iron with the same thickness makes it possible to significantly reduce the mass of the shield. Moreover, such replacement provides the reduction of the DR from SNF behind such shield. The efficiency of SNF transportation is also determined by the amount of SNF in the TC, which depends on the volume of the container. The DR at the surface of the TC radiationshield is well-known to be determined mostly by that

SNF (those SFAs), which is positioned at the perimeter of the cask basket. This is explained by the efficient self-absorption of the radiation from the SNF inside the basket. It is shown in the present paper that the DR from a cylindrical radiation source (the basket with SNF, Fig. 6) behind the shielding is proportional to the surface area of the source, $\sim 2\pi R_1 \times h$, while the volume of the source is $\pi R_1^2 \times h$.

Fig. 6. Schematic of the SNF radiation source and the gamma-radiation shielding

The DR increases linearly, $DR_2 \approx DR_1 \times (R_2/R_1)$, with the increase of the TC radius $R_1 \rightarrow R_2$ (see Fig. 6). And the SNF volume (i.e., the amount of SNF) increases quadratically, i.e., stronger, $V_2 = V_1 \times (R_2/R_1)^2$. In the present model, $R_1 = 0.9$ m and $R_2 = 1$ m are taken. In this case, the DR increases by $\sim 11\%$: $DR_2 \approx DR_1 \times 1.11$, while the amount of SNF increases by 23%: $V_2 = V_1 \times 1.23$. Our calculations with the source radius of $R_2 = 1$ m show that DR₂ with shielding thicknesses from 25 to 40 cm made from various materials (both Fe and Pb, U, uranium dioxide $UO₂$ with different densities) differ from those given in Table 3 by less than 11%. Therefore, the following options are considered.

1) For the TC with the SNF basket with the radius of 0.9 m and the shield made of Fe with the thickness of 30 cm, the DR₁ is 91 μ Sv/h, and the mass of the shield is 77.8 t.

2) For the TC with the SNF basket with the radius of 1.0 m and the shield made of Fe with the thickness of 30 cm, the DR₂ is 101 μ Sv/h, and the mass of the shield is 85.2 t.

3) For the TC with the SNF basket with the radius of 1.0 m and the shield made from depleted uranium with the density of 4.5 $g/cm³$ and the thickness of 40 cm, the DR₂ is 84.8 μ Sv/h, and the mass of the shield is 67.9 t.

Thus, application of the thicker radiation shields made from the materials with higher atomic number and lower density than those for Fe in designing the TCs is shown to provide three simultaneous advantages. First, the mass of the shields can be reduced as compared with the shields made of Fe. Second, the amount of the SNF transported in the TC (the volume of the SNF basket)

can be increased. Third, DR outside the TC can be reduced.

CONCLUSIONS

Using a cask for transporting the spent nuclear fuel as an example, the efficiency of gamma-radiation shielding, which forms the main part of the cask mass, is studied. The gamma-radiation shielding is represented by coaxially placed steel baskets, the cavities between the cylindrical surfaces of which are filled with a radiation-shield material with high atomic number, dispersed to densities in the range $4 < \rho < 8$ g/cm³. Simulations are carried out by the Monte Carlo method. The simulations show that at thick shielding (30…40 cm), made of steel (Fe) and depleted uranium $(U(6))$ with density of 6 g/cm³, as well as depleted uranium (U) with density of 18.9 $g/cm³$ (in this case, the mass thickness is the same as for Fe), the spectra of the photons which passed through the radiation-shield are the same. The attenuation capacity of the shielding, when γ -radiation of long-lived SNF isotopes (^{134}Cs , 137 Cs, 154 Eu – they produce the main contribution to the dose rate behind the shielding) passes through the shielding, is minimal for U (18.9 $g/cm³$) and is maximal for U(6) (6 g/cm³). The attenuation capacity of γ -shields with a large thickness is practically independent on the level of burnup and cooling time of spent nuclear fuel. The application of the gamma radiation shielding made of depleted uranium dispersed up to densities in the range of $4.5 < \rho < 7$ g/cm³ in designing the transport container (TC) is shown to reduce the mass of the TC as compared with the steel shield and reduce the dose rate (DR) on the container surface. Reducing the DR also make it possible to reduce the cooling time of spent nuclear fuel in the near-reactor spent fuel cooling pool. The smaller mass of the shielding with better absorption of γ -quanta from spent nuclear fuel as compared with the shielding made of steel make it possible to increase the size of the basket with spent nuclear fuel and the thickness of the shielding. Such design satisfies the requirements to the weight characteristics of the TC used for rail transportation, and increases the efficiency of the transportation (increases the amount of spent nuclear fuel in one TC).

REFERENCES

1. A. Historical. Review of the Safe Transport of Spent Nuclear Fuel, FCRD-NFST-2016-000474 (Rev. 1), ORNL, Oak Ridge, 2016, 88 p.

2. Managing Aging Effects on Dry Cask Storage Systems for Extended Long-Term Storage and Transportation of Used Fuel – Revision 1, September 30, 2013, Argonne National Laboratory, FCRD-UFD-2013-000294б, ANL-13/15.

3. О.В. Григораш, О.М. Дибач, С.М. Кондратьєв др. Питання ядерної та радіаційної безпеки централізованого сховища відпрацьованого ядерного палива АЕС України *// Ядерна та радіаційна безпека.* 2017, т. 3(75), с. 3-10.

4. V.G. Rudychev, N.A. Azarenkov, I.O. Girka, Y.V. Rudychev. Efficiency of various materials application for radiation shielding at transportation and storage of spent nuclear fuel by dry method // *Problems of Atomic Science and Technology. Series "Physics of Radiation Effect and Radiation Materials Science".* 2020, N 2(126), p. 64-70.

5. В.Г. Рудичев, М.О. Азарєнков, І.О. Гірка, Є.В. Рудичев. *Контейнер для транспортування та/або зберігання відпрацьованого ядерного палива.* Патент України №145814 від 06.01.2021, Бюл. №1.

6. X-5 Monte-Carlo Team. MCNP-A General Monte-Carlo N-Particle Transport Code. Version 5. Volume I: Overview and Theory. USA: Los Alamos National Laboratory, 2003, LA-UR-03-1987

7. I.I. Zalubovsky, S.A. Pismenetskiy, V.G. Rudychev, S.P. Klimov, Y.V. Rudychev, et al. Protective structures for storing spent nuclear fuel from the Zaporozhye NPP // *Atomic Energy*. 2012, v. 112, p. 261.

Статья поступила в редакцию 04.03.2021 г.

КОМБИНИРОВАННЫЙ РАСЧЕТ ИЗЛУЧЕНИЙ ОТ ПОВЕРХНОСТНЫХ ХРАНИЛИЩ РАО БОЛЬШИХ РАЗМЕРОВ НА ОСНОВЕ МЕТОДА МОНТЕ-КАРЛО

В.Г. Рудычев, Н.А. Азаренков, И.А. Гирка, Е.В. Рудычев

Радиационная защита от гамма-квантов существующих транспортных контейнеров (ТК) для перевозки отработавшего ядерного топлива (ОЯТ) изготавливается из стали или стали и Pb толщиной 25…30 см и массой $\sim 60...80$ т. Исследовано применение материалов с высоким атомным номером, диспергированных (твердые частицы, измельченные до порошкообразного состояния) до плотностей в диапазоне $4 < \rho < 8$ г/см³. Моделирование методом Монте-Карло показывает, что при плотностях диспергированного обедненного U больше, чем 5 г/см³ и толщине защиты более 30 см поглощение гамма-квантов ОЯТ больше, чем у защиты из стали такой же толщины. Применение таких материалов при непревышении весовых характеристик обеспечивает радиационную защиту для ОЯТ с высокой степенью выгорания и меньшим временем выдержки или большим количеством транспортируемого ОЯТ.

КОМБІНОВАНИЙ РОЗРАХУНОК ВИПРОМІНЮВАНЬ ВІД ПОВЕРХНЕВИХ СХОВИЩ РАВ ВЕЛИКИХ РОЗМІРІВ НА ОСНОВІ МЕТОДУ МОНТЕ-КАРЛО

В.Г. Рудичев, М.О. Азаренков, І.О. Гірка, Є.В. Рудичев

Радіаційний захист від гамма-квантів існуючих транспортних контейнерів (ТК) для перевезення відпрацьованого ядерного палива (ВЯП) виготовляється зі сталі або сталі і Pb товщиною 25…30 см і масою ~ 60…80 т. Досліджено застосування матеріалів з високим атомним номером, диспергованих (тверді частинки, подрібнені до порошкоподібного стану) до щільності в діапазоні $4 < \rho < 8$ г/см³. Моделювання методом Монте-Карло показує, що при щільності диспергованого збідненого U більше, ніж 5 г/см³ і товщині захисту більше 30 см поглинання гамма-квантів ВЯП більше, ніж у захисту зі сталі такої ж товщини. Застосування таких матеріалів при неперевищенні вагових характеристик забезпечує радіаційний захист для ВЯП з високим ступенем вигоряння і меншим часом витримки або великою кількістю ВЯП, що транспортується.