CHANGE OF RADIOACTIVE WASTE CHARACTERISTICS AT THEIR PROCESSING AND STORAGE AT NUCLEAR POWER PLANTS

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This paper offers some methods developed for calculation of radiation from the casks for storage of radioactive wastes (RW) at the nuclear power plant (NPP) area. Monte Carlo method was used to calculate spectral-angular characteristics of the cask external radiation generated by separate nuclides. The obtained data allowed to provide simple methods for calculating changes in the radiation characteristics with the waste storage time and determining the radiation spatial distribution away from the casks. We propose a technique of reducing the radiation dose due to arranging properly loading the casks with barrels containing wastes of different activity. It is shown that at the RW storage its radiotoxicity decay is slower than the dose rate decay due to decrease of the activity of short-lived radionuclides.

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1. INTRODUCTION

In the course of normal operation of nuclear power plants (NPP) radioactive wastes (RW) – gaseous, liquid and solid are produced. At different nuclear plants the yield of liquid wastes, on average, makes from 0.15 to 0.35 m^3/MW, and of solid wastes is 0.1...0.3 m^3/ MW per year.

The main task when processing RW is to prevent propagation of radioactive materials into the environment. For this purpose reprocessing of nuclear waste is arranged just directly at the nuclear power plants to reduce their volume and to recycle them into the form for safe interim storage. To reduce the volume of liquid RW they are evaporated, and firm RW are crushed, burned and pressed.

Currently all the radioactive waste storage facilities at the nuclear power plants in Ukraine are full up. One of the ways to reduce the maintenance costs is the technology of temporary storage of the conditioned RW in light hangar storages at the plant area. Reinforced concrete casks ("cubic") in the form of rectangular parallelepipeds are arranged in a multistage hangar (up to four tiers). These casks house up to four barrels with conditioned RW: salt melt, pressed solid RW and others.

The aim of this work is development methods for calculating radiation from the "cubic" casks designed for interim storage of radioactive materials to provide the possibility of reducing the radiation doses due to arranging properly loading the containers with waste of different activities. On the basis of the Monte Carlo methods a technique has been developed for calculating characteristics of radiation from the "cubic" casks loaded with cylindrical containers with air-conditioned RW.

2. CALCULATION METHODS

The object under research represents four cylindrical containers with RW (200 liters each), placed in the cask in the form of a parallelepiped with thick enough walls. In this case, the radiation source – RW can have different isotopic, elemental composition and density. External radiation from "the cubic" cask is determined by spectral composition of the isotope-emitted γ-rays and by transport of photons from containers with radioactive wastes through the walls of "cubic" cask. Passage, absorption and scattering of photons depend on geometry of the entire object and elemental composition.

Currently only Monte-Carlo simulation allows determining radiation from such objects correctly. The external radiation characteristics were calculated applying the MCNP package which is widely used in nuclear power engineering [1].In this package a geometrical model of a shielding reinforced concrete cask with four cylindrical sources, was developed (Fig.1). As long as in Ukraine, up to this day there is no final decision as to the characteristics of such a cask, the basic geometrical dimensions for it were taken as those of the cask used at the Novovoronezhskaya NPP [2].

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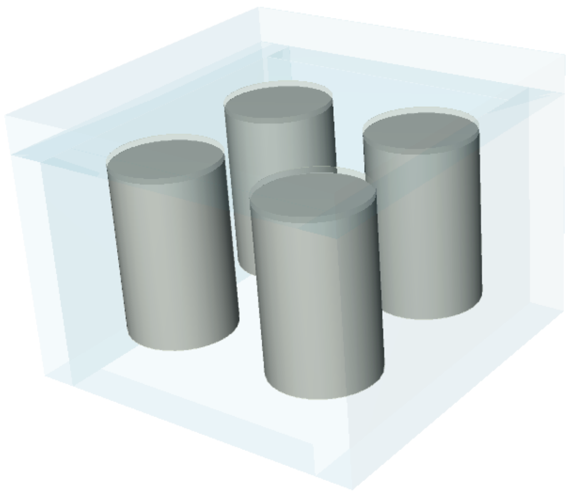

Fig.1. *Spatial configuration of the cask with 4 cylindrical sources*

Monte-Carlo simulation allows to determine comprehensive characteristics of an emitting system. However a significant disadvantage of this method is a long computation time. This limits application of this method for carrying out a large number of calculations.

Often it is necessary to know the characteristics of the radiation from the cask sidewall. Fig.1 shows that in this case the main contribution will be made by two barrels placed near this wall. In the calculation we used a volumetric method of integration of point sources. It is assumed that each element dV of the cylindrical source volume isotropically emits gamma rays with energy E and density $n_\gamma(E)$. Then the number of gamma rays $N_\gamma(E)$ at the point P [3], which is in the plane of the source base, will be determined by the following relation:

$$N_\gamma(b,h,E) = \frac{n_\gamma(E)}{2\pi}\int_0^h dz\int_0^R \rho d\rho\int_0^\pi \frac{B(\mu(E)x)e^{-\mu(E)x}d\phi}{\rho^2+b^2+z^2-2b\rho\cos\phi}\,, \tag{1}$$

where $x = x(z,\rho,\phi)$ – the distance between the volume element and the cylinder side surface, and this distance corresponds to the length of self-absorption in the material of which the source consists; b – the distance between the cylinder axis and the observation point P; $B(\mu(E)x)$ — the accumulation factor for the point source; $\mu(E)$ — the linear attenuation factor of gamma rays with energy E in the source matter; R — radius of the cylinder; h — height of the cylinder. Expression for $x = x(z,\rho,\phi)$ is as follows:

$$x = \frac{\rho^2 - b\rho\cos\phi + \sqrt{(\rho^2+b^2-2b\rho\cos\phi)R^2 - \rho^2b^2\sin^2\phi}}{\rho^2+b^2-2b\cos\phi}\sqrt{\rho^2+b^2+z^2-2b\rho\cos\phi}\,. \tag{2}$$

The relation for the gamma-ray flux from the cylindrical source beyond the flat shielding barrier at the point disposed away from the source in such a way that the radius-vector from the source center and the normal to the protective barrier form an angle α, has the form (Fig.2):

$$N_\gamma(b,h,E) = \frac{n_\gamma(E)}{2\pi}\int_0^h dz\int_0^R \rho d\rho\int_0^\pi \frac{B(\mu(E)x)B_w(\mu_w(E)y)e^{-(\mu(E)x+\mu_w(E)y)}d\phi}{\rho^2+b^2+z^2-2b\rho\cos\phi}\,, \tag{3}$$

where $y = \frac{t\sqrt{\rho^2+b^2+z^2-2b\rho\cos\phi}}{(b-\rho\cos\phi)\cos\alpha-\rho\sin\phi\sin\alpha}$; $\mu_w(E)$ – linear attenuation factor in the shield material; $B_w(\mu_w(E))$ – accumulation factor in the shield material.

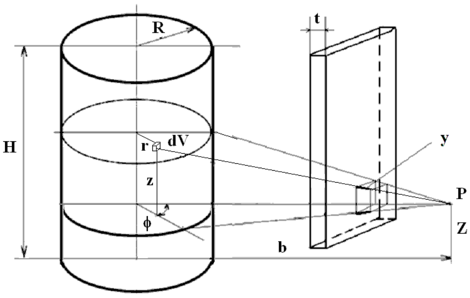

Fig.2. *Geometry for a self-absorbing cylindrical volume source with shielding*

Generally cylindrical sources are placed along the protective wall in such a way that radiation of separate sources propagates through the shield to the observation point under different angles. Radiation calculations using both Monte-Carlo method and method of volumetric integration of sources determine characteristics directly from the RW placed inside the shielding (protective) cask. One of the ways of saving the calculation time is to combine these two methods. The shielding cask surface is assumed to be the source of radiation. Each element of the source surface dS emits gamma rays with spectral – $I_e(E)$ and angular – $An(\phi)$ distributions, and gamma-ray density $n_\gamma(x,y)$ dependent on the element co-ordinate dS. E – energy, and ϕ – emission angle of γ-rays relative to the normal to the surface. Values: $An(\phi,E)$ and $n_\gamma(x,y,E)$ depend on the characteristics of the RW placed in the shielding cask and are determined by Monte-Carlo method. Photon irradiation both in the air and through the shields is determined by the method of surface integration of point sources. Transport scheme for γ-rays from the radiating rectangle to point P with coordinates X,Y behind the wall of concrete with t-thickness is presented in Fig.3. The distance between the point P and the area element $dS = dx \cdot dy$ with co-ordinates x, y is determined by the relation: $R = \sqrt{(x-X)^2 + (y-Y)^2 + z^2}$. Then the gamma-ray flux in the point P(X,Y) is determined by the following expression:

$$N_\gamma(z, X, Y, E_i) = I_e(E_i) \cdot \int_0^{H_y}\int_0^{H_x} \frac{n_\gamma(x, y, E_i) An(\phi, E_i) B_w(\mu_w(E_i)t')e^{-\mu_w(E_i)t'}}{R^2} dxdy\,, \tag{4}$$

where H_x and H_y — the rectangle dimensions, $\phi = arcos(z/R)$ – the angle under which a gamma ray emits to the point P relative to the normal to the plane (x,y), $t' = t \cdot R/z$.

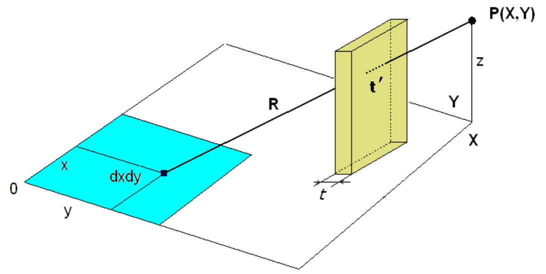

Fig.3. *Calculation scheme for gamma-ray irradiation from the surface source*

Such a technique provides quick and correct calculation of characteristics of radiation generated by several casks arranged at the storage sites or in the storage facilities. The γ -ray flux at the point of observation (and accordingly its dose rate too) depends on the energy of the initial photons from the RW. And energy of the initial photons is determined by isotope composition of radionuclides contained in the RW. Then γ-ray flux per one isotope decay j in the point P for all the calculation methods: Monte-Carlo method, volumetric integration of dot sources and the combined method can be written as:

$$N_j = \sum_{i=1}^{Ng} n_i^j \cdot N_\gamma(x, y, E_i)\,, \tag{5}$$

where n_i^j — quantum yields of i-line of j-th isotope, Ng^j– the number of lines of j-th isotope. Using expression (5) we'll obtain the relation for the γ-ray flux depending on the storage time for all isotopes in the form:

$$N_s(T) = \sum_{j=1}^{Niz} \left[\chi_i^j \cdot Q_j(T) \cdot \sum_{i=1}^{Ng^j} (n_i^j \cdot N_\gamma(x, y, E_i) \right]\,, \tag{6}$$

where $Qj(T)$ — activity of j-th isotope; χ — partial content of j-th isotope in the RW; Niz — the number of isotopes in the RW.

One of the widely used characteristics of radiation hazard for the RW or spent nuclear fuel (SNF) is radiotoxicity. Radiotoxicity of the j-th nuclide in the air or in the water is determined by relation:

$$RT_j = Q_j/PQ_j\,, \tag{7}$$

where Q_j – activity of the j-th nuclide under study; PQ_j — maximum permissible activity of this nuclide in the air or in the water. According to Radiation Standards in Ukraine [7] for the air $PQ_j = PC_{Aj}^{inhal}$, and for the water $PQ_j = PC_{Bj}^{ingest}$. For the mixture of radionuclides with known composition in the air the value of the $PC_{A\Sigma}^{inhal}$ admissible concentration is determined by the relation:

$$PC_{A\Sigma}^{inhal} = \frac{100}{\sum_J(\chi_j PC_{Aj}^{inhal})}, Bk/m^3\,, \tag{8}$$

where χ_j – specific concentration of each isotope. The formula for calculating of the permissible concentration of $PC_{B\Sigma}^{ingest}$is similar to the mentioned above.

3. INITIAL DATA

Concrete casks are rectangular parallelepipeds with overall dimensions of 1.65 m × 1.65 m × 1.375 m and the wall thickness of 0.15 m [2]. The air-conditioned RW loaded into the containers is pressed solid waste (including pressed ashes at Zaporozhye nuclear power plant) and salt melt. The solid radiation waste (SRW) density ranges from 2.2 g/cm^3 (pressed ashes) to 4 g/cm^3 (building waste, with metal debris); the salt melt density ranges from 1.7 to 2.1 g/cm^3. To determine the radiation characteristics we analyze two types of conditioned SRW: salt melt with density of 2 g/cm^3 and pressed SRW with density of 4 g/cm^3. The salt melt elemental composition is similar to the concrete elemental composition, and SRW building waste is concrete + metal (70 % of concrete + 30 % of iron). Pressed SRW is produced by compressing the waste having density of ≈1g/cm^3, this SRW can get to the pressing machine right after its extracting, without pre-cooling. Such SRW can contain a sufficient amount of short-lived nuclides whose half life is less than a year [4], see Table. The salt melt is produced by deep evaporation of liquid radioactive waste ("distillation residue") stored in containers at the NPP area for a long enough time with the nuclides composition [2], see Table. The data in Table on permissible concentration of isotopes in the air PC$_A^{inhal}$ and in the water PC$_B^{ingest}$ were derived from Radiation Standards in Ukraine [7].

Characteristics of nuclides contained in the salt melt and SRW

Serial number	Isotope	PC_A^{inhal}, Bk/m^3	PC_B^{ingest}, Bk/m^3	$T_{1/2}$, Year	Content of isotopes% Solid RW	Salt melt
1	^{54}Mn	1000	8.00E+05	0.85	4	
2	^{60}Co	70	8.00E+04	5.27	21.1	20
3	^{110m}Ag	200	2.00E+05	0.685	25.2	
4	^{134}Cs	100	7.00E+04	2.06	13.1	15
5	^{137}Cs	60	1.00E+05	30	36.6	65

Quanta yields of photons and energy spectra of γ-radiation for isotopic content of radionuclides in SRW and salt melt was determined from the database of package JEF PC [5].

4. CALCULATION RESULTS

One of the indicators of radiation safety at handling with RW is the dose rate value 1 m away from the source surface. Four cylindrical containers (barrels, 200 liters) are filled with SRW (density of 4 g/cm^3) or salt melt (density of 2 g/cm^3) and placed into the cask. Spectra of photons, that passed through the shielding wall of the cask, generated by separate nuclides contained in the SRW and salt melt (^{54}Mn, ^{60}Co, ^{110m}Ag, ^{134}Cs, ^{137}Cs) were calculated using Monte Carlo method. Total spectral distributions averaged over the container surface, that were generated by SRW whose radionuclide composition is given in Table 1, are shown in Fig.4. Spectra are calculated in accordance with relation (6) for the time of RW processing (T = 0),and the storage time for 20 years (T = 20). The same figure shows distribution of the dose rate (DR) from photons with such energies.In this case we used coefficients of photon (with energy E) conversion into the dose rate derived from ICRP-74 [6].

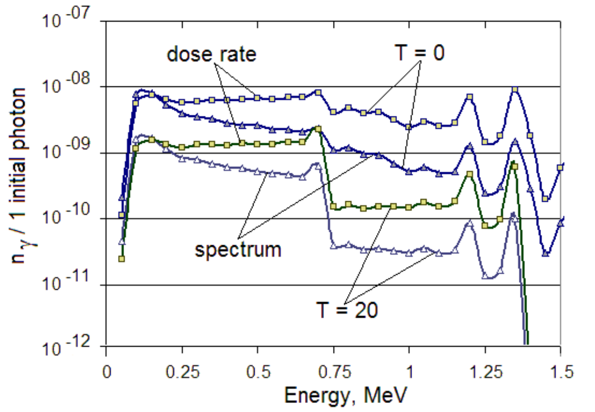

Fig.4. *Spectral distributions and dose rates depending on the photons energy*

The data given in Figure 4 show that in the RW itself and in the concrete walls of the cask the primary energy of γ-rays significantly decreases due to re-scattering of photons. The dose rate from photons, according to the conversion coefficients, grows with growth of the photons energy. High-energy gamma-quanta, whose energy is higher then 0.662 MeV, at T = 0 contribute 40 % to the DR. At increase in the RW storage time the content of isotopes with short half-life which emit high-energy γ-quanta abruptly decreases. Their contribution (E > 0.662 MeV) at storage time T = 20 years is 13 %. Fig.5 summarizes the contributions of various nuclides contained in SRW and salt melt to the dose rate depending on the storage time. Spectral distributions and RD depending on the energy of photons are similar to those shown in Fig.4. Spectral distribution and RD depending on the energy of photons are similar. Due to the presence of a short-lived isotope ^{110m}Ag ($T_{1/2}$ = 0.685 years) in SRW, which at T = 0 the 25% contributes to the dose rates, with increase of the storage time also increases contribution of high-energy photons from ^{60}Co, up to 57% at T = 3.1 years. Further growth of the storage time leads to some decrease of ^{60}Co contribution to the rate dose. For the salt melt, which has no short-lived isotopes, ^{60}Co contribution to the RD is maximal at T = 0, and with increase of the storage time it decreases. The contribution of ^{134}Cs to the RD makes about 10 % for the salt melt at T = 0 (somewhat lower for SRW) and with increase of the storage time it quickly falls down. The contribution of ^{137}Cs to the RD becomes determinative (over 90 %) at T > 22 years for the salt melt, and at T > 28 for the SRW.

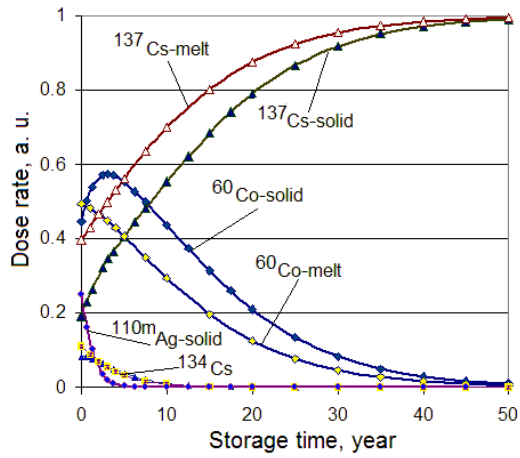

Fig.5. *Contributions of various nuclides contained in SRW and in salt melt to the RD depending on the storage time*

Decay of the isotopes activity in time to a great extent determines the change in the radiation characteristics due to the change in the isotope composition. Fig.6 shows changes in time in the permissible concentration of PC_A^{inhal} mixture of radionuclides in the air for the salt melt and SRW.

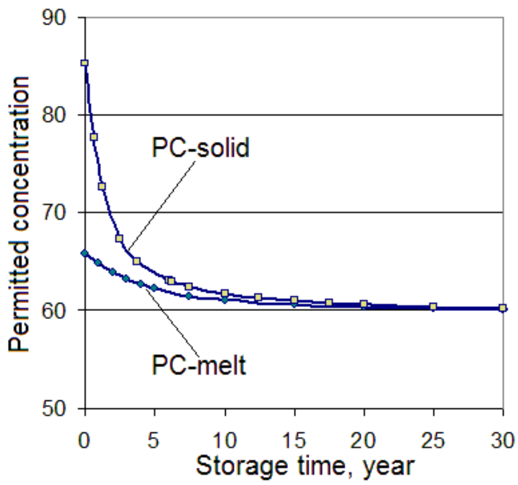

Fig.6. *Dependences of change in the course of time of the concentration of PC_A^{inhal} mixture of radionuclides in the air for the salt melt and SRW*

It is clear, that with increase of storage time the requirements to the content of RW aerosols in the air become stricter as long as PC_A^{inhal} decreases to 60 Bk/m^3 of the defined ^{137}Cs. The change in time of PC_A^{inhal} also influences the change in radiotoxicity. Dependences of activity, radiotoxicity and dose rate on the SRW storage time are shown in Fig.7.

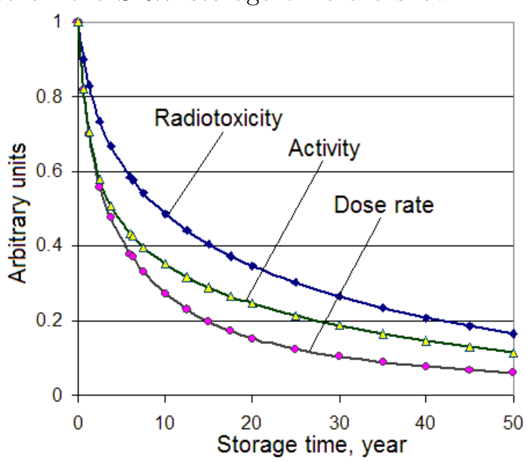

Fig.7. *Dependences of activity, radiotoxicity and dose rate on the SRW storage time*

It is seen, that because of PC_A^{inhal} change the radiotoxicity falls slower than the activity and the dose rate. When arranging the casks containing RW at the storage sites or in the storage facilities of hangar type a requirement often occurs to minimize the radiation dose at any part of it. And at the same time, the construction of an additional biological shield is undesirable. Paper [2] offers a method allowing to reduce the radiation doses at one side of the cask. The idea is to remove the cylindrical containers (barrels) to the other wall of the cask, and to fill the free space with bulk absorbent of γ-rays, for example, sand. This method is effective enough, but in this case the cask weight considerably increases and the process of RW treatment becomes complicated. Since the RW storage facilities house wastes with different activity and storage time, it is possible to place 2 barrels with waste of lower activity along one wall, and 2 barrels with waste of higher activity along the other wall. In this paper we consider the option of arranging sources with different activity. For this purpose transport of radiation from two sources (barrels) with RW were calculated using the MCNP package: a) the barrels are placed near the wall of the cask, 1st row; b) the barrels are placed in the second row, so that the barrels with RW in the first row serve as shields (Fig.8).

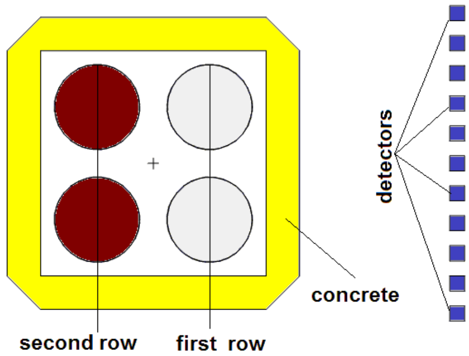

Fig.8. *Layout of containers with RW of different activity in a cask*

The calculations for different isotope composition of RW and densities of 2 and 4 g/cm^3 showed that the containers with RW from the 1st raw serve as a good shield. On average, the RD produced by the sources in the 2nd row makes 7...10% of the total RD from all the casks with RW. That is, placing of more active radiation sources in the 2nd row might be an effective way to reduce the RD in a predetermined direction, for example, at the boundary of the storage facilities. Fig.9 shows the detector-averaged photon spectra produced by the sources with the identical activity in the 1st and 2nd rows. Spectral distributions are given for ^{60}Co and ^{137}Cs, the RW density is assumed to be 4 g/cm^3.

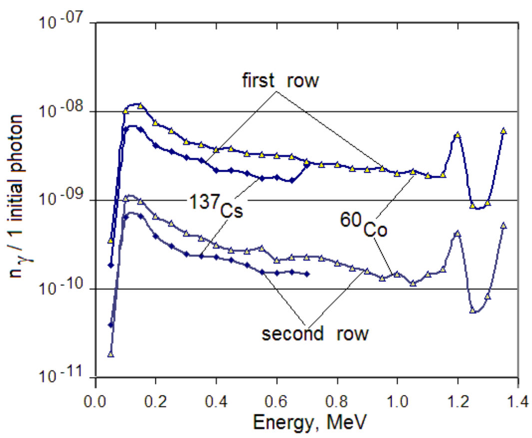

Fig.9. *Detector-averaged photon spectra produced by the sources with the identical activity in the 1st and 2nd rows*

It is clear that the amount of initial and scattered photons (isotopes ^{60}Co and ^{137}Cs) from the second row is ten times less than that from the first raw. Hence, by analogy with relation (6) the flux from the cask may be written in the form:

$$N_s(T) = N_{s1}(T) + N_{s2}(T)\,. \qquad (9)$$

Indices: 1 - characteristics of isotopes and radiation transport from the first row of the casks, 2 - from the second row of the casks.

5. CONCLUSIONS

Spectral-angular characteristics of the cask external radiation obtained by the Monte Carlo method for separate nuclides are used as input data for the common calculation techniques:

a) change in the radiation characteristics with waste storage time;

b) determination of the radiation spatial distribution away from the casks.

It is shown that at the waste storage its radiotoxicity decay is slower than the dose rate decay due to decrease of the activity of short-lived radionuclides. To reduce the radiation dose rate at one side of the cask a method of radiation calculation for arrangement of the cask with RW of different activity is developed.

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ИЗМЕНЕНИЕ ХАРАКТЕРИСТИК РАДИОАКТИВНЫХ ОТХОДОВ ПРИ ОБРАЩЕНИИ И ХРАНЕНИИ НА АЭС

В. Г. Рудычев, И. А. Гирка, Е. В. Рудычев, А. А. Каплий, А. Ф. Щусь, Т. А. Сокольцова

Разработаны методики расчета излучения от контейнеров хранения РАО на территории АЭС. Методом Монте-Карло рассчитаны спектрально-угловые характеристики внешнего излучения контейнера, создаваемые отдельными нуклидами. Полученные данные позволили создать простые методики расчета изменения характеристик излучения от времени хранения и определения пространственного распределения излучения на удалении от контейнеров. Предложена методика уменьшения дозовых нагрузок за счет надлежащего размещения в контейнеры емкостей с отходами разной активности. Показано, что при хранении РАО радиотоксичность спадает медленнее по сравнению со спадом мощности дозы за счет снижения активности короткоживущих радионуклидов.

ЗМІНИ ХАРАКТЕРИСТИК РАДІОАКТИВНИХ ВІДХОДІВ ПРИ ПОВОДЖЕННІ ТА ЗБЕРІГАННІ НА АЕС

В. Г. Рудичев, І. О. Гірка, Є. В. Рудичев, А. А. Каплій, А. П. Щусь, Т. А. Сокольцова

Розроблені методики розрахунку випромінювання від контейнерів зберігання РАВ на території АЕС. Методом Монте-Карло розраховані спектрально-кутові характеристики зовнішнього випромінювання контейнера, створювані окремими нуклідами. Отримані дані дозволили створити прості методики розрахунку зміни характеристик випромінювання від часу зберігання і визначення просторового розподілу випромінювання на віддалі від контейнерів. Запропоновано методику зменшення дозових навантажень за рахунок належного розміщення в контейнери ємностей з відходами різної активності. Показано, що при зберіганні РАВ радіотоксичність спадає повільніше в порівнянні із спадом потужності дози за рахунок зниження активності короткоживучих радіонуклідів.