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DETERMINATION OF EXTERNAL IRRADIATION FROM SOURCES WITH COMPLEX GEOMETRY CONTAINING RADIOACTIVE WASTE

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A combined technique using both Monte-Carlo and quasi-analytic method has been developed for calculation of irradiation from complex objects containing radioactive waste. Monte Carlo method is used to calculate spectral, angular and spatial distributions of γ-quanta close to one of the surfaces of the concrete cask loaded with containers filled with radioactive waste (RW). Radiation characteristics at the prescribed distance are determined by integrating the point sources with calculated distributions over this surface. Summation of photons of external radiation from individual RW isotopes allows finding the photon flux for any storage time. Application of the technique allows to calculate asymmetric loading of casks with containers filled with RW of different activity and to optimize the arrangement of the casks in the RW storage area. This allows reducing the effect of radiation-absorbed dose on the service personnel and on the areas surrounding RW storage.
KEY WODS: radiation shielding; radioactive waste; radionuclide; dose rate; Monte Carlo method

ОПРЕДЕЛЕНИЕ ВНЕШНЕГО ИЗЛУЧЕНИЯ ОТ ИСТОЧНИКОВ СО СЛОЖНОЙ ГЕОМЕТРИЕЙ СОДЕРЖАЩИХ РАДИОАКТИВНЫЕ ОТХОДЫ

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Разработана комбинированная методика, использующая метод Монте-Карло и квазианалитический метод для расчета излучения от сложных объектов содержащих радиоактивные отходы (РАО). Методом Монте-Карло, рассчитываются спектральные, угловые и пространственные распределения γ – квантов вблизи одной из поверхностей бетонного контейнера, котором размещены бочки с РАО. Интегрированием по этой поверхности точечных источников с рассчитанными распределениями определяется характеристики излучения на заданном расстоянии. Суммирование фотонов внешнего излучения от отдельных изотопов РАО позволяет определить поток фотонов на любой момент времени. Применение методики дает возможность рассчитать несимметричную загрузку контейнеров бочками с РАО разной активности и оптимизировать размещение контейнеров в зоне хранения РАО. Это позволяет уменьшить дозовые нагрузки на обслуживающий персонал и окружающей хранилище территории.
КЛЮЧЕВЫЕ СЛОВА: радиационная защита; радиоактивные отходы; радионуклиды; мощность дозы; метод Монте-Карло

ВИЗНАЧЕННЯ ЗОВНІШНЬОГО ВИПРОМІНЮВАННЯ ВІД ДЖЕРЕЛ ІЗ СКЛАДНОЮ ГЕОМЕТРІЄЮ, ЯКІ МІСТЯТЬ РАДІОАКТИВНІ ВІДХОДИ

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Розроблена комбінована методика, що використовує метод Монте-Карло і квазіаналітичний метод для розрахунку випромінювання від складних об'єктів, які містять радіоактивні відходи (РАВ). Методом Монте-Карло розраховуються спектральні, кутові і просторові розподіли γ-квантів поблизу однієї з поверхонь бетонного контейнера в якому розміщені бочки з РАВ. Характеристики випромінювання на заданій відстані визначаються інтегруванням по цій поверхні точкових джерел з розрахованими розподілами. Підсумовування фотонів зовнішнього випромінювання від окремих ізотопів РАВ дозволяє визначити потік фотонів на будь-який момент часу. Застосування методики дає можливість розрахувати несиметричне завантаження контейнерів бочками з РАВ різної активності й оптимізувати розміщення контейнерів в зоні зберігання РАВ. Це дозволяє зменшити дозові навантаження на обслуговуючий персонал, а також територію, що оточує сховище.
КЛЮЧОВІ СЛОВА: радіаційний захист; радіоактивні відходи; радіонукліди; потужність дози; метод Монте-Карло

In the process of the reactor units operation at all nuclear power plants (NPP) in Ukraine there exists a problem with storage of radioactive waste (RW). While operating normally the nuclear power plants produce significant amount of radioactive waste (gaseous, liquid and solid). The gaseous waste is filtered by various filters, including liquid ones.

As a result both liquid and solid RW is produced. To reduce the amount of this RW at NPPs it is preliminarily processed. The liquid radioactive waste (LRW) as “distillation residue” is evaporated resulting in a variety of solid waste – «salt melt». The solid radioactive waste (SRW) is burned and pressed. Despite the RW compaction the storage facilities for all types of RW are almost full up. At Zaporozhye, Rovno and Khmelnitsky NPP melt salt storages are filled to 85-95%, SRW storages are filled to about the same value.

To solve this problem in the world practice there is a safe and cost-effective method of conditioned RW temporary storage (up to 50 years) at the areas of nuclear power plants. Cylindrical containers up to 200 litres filled with radioactive waste are placed in reinforced concrete rectangular casks, which are arranged in the open air or in the light-duty hangars. Outside the safeguard zone of the storage the dose rate (DR) should not exceed the permissible limits of radiation safety standards for the NPP personnel working at the premises adjacent to the storage area. In a number of papers some methods to reduce the dose rate from radiation sources located on the perimeter of the storage areas have been proposed [1,2,3].

The aim of the present work is developing an effective calculation technique for external radiation from reinforced concrete rectangular casks loaded with containers filled with RW of different activity and isotope composition. Due to arrangement of the sources with different dose rate along on the storage area perimeter such techniques would allow reducing the effect of radiation-absorbed dose on the NPP personnel.

INITIAL DATA AND METHODS OF DOSE RATE CALCULATIONS

From the data given in [1,3,4] and from the analysis of the isotope composition of RW produced by Ukrainian NPPs it follows that the main contribution to the γ- quanta radiation is made by 3 isotopes whose half-life is over 2 years: ^{60}Co, ^{134}Cs and ^{137}Cs, and with shorter half-life ($T_{1/2}$ <1 year): ^{54}Mn and ^{110m}Ag. The averaged content of these isotopes for different types of RW is presented in Table. From these data it follows that at increase of the storage time the RW isotope composition changes considerably due to decrease of the content of isotopes with short half-life.

Table.

Nuclide content in the salt melt, pressed SRW, and ash produced after burning of SRW and radioactive oil

# #	Isotope	Average energy of γ– quanta, MeV	$T_{1/2}$ Year	Content of isotopes χ_j, %		
				Ash	SRW	Salt melt
1	^{54}Mn	0.83	0.858	24	4.0	
2	^{60}Co	1.25	5.27	59	21.1	20
3	^{110m}Ag	0.86	0.685		25.2	
4	^{134}Cs	0.70	2.06	12	13.1	15
5	^{137}Cs	0.66	30	5	36.6	65

Pressed SRW are produced from waste with density of ≈1 g/cm^3 in the form of construction debris (~ 70% of concrete and ~ 30% of iron). After pressing SRW its density reaches 4 g/cm3, the density of compressed ash is 2.2 g/cm^3, and the density of the salt melt is in the range of 1.7 – 2.1 g/cm^3. The elemental composition of the salt melt and ash is similar to that of the concrete.

At the site of Zaporozhye NPP it is planned to construct light-duty hangar storage (without biological shielding) where reinforced concrete rectangular casks will be arranged in four tiers. The air-conditioned RW in containers of 200 litres are placed in rectangular casks (4 containers in each) with overall dimensions of 1.65 m × 1.65 m × 1.375 m and wall thickness of 0.15 m [1,8]. The greatest contribution to the dose rate on the storage perimeter is evidently made by the radiation sources arranged along the borders of storage facilities or storage areas. Hence, loading the cask with containers filled with RW of different activity and spectral composition of radiation, as well as the casks arrangement in the RW storage facility determine the DR along the perimeter of the safeguard zone.

External radiation of the cask is determined by change in the spectral composition of isotopes γ-quanta contained in RW when it is absorbed and attenuated both in the containers with RW and in the casks walls. Passage, absorption and scattering of photons depend on the geometry of the entire object as well as on the elemental composition both of RW and the shielding walls of the cask. Only Monte Carlo simulation of geometric characteristics and elemental composition of such objects allows determining correctly the external radiation characteristics. When calculating shields in nuclear engineering the method of volume integration of radiation sources, implemented in MicroShield [5] and VOLUME [6] packages is often applied. However, applying these packages in calculation of sufficiently thick biological shields (in our case the casks walls) results in a fairly large error.

For calculation of the external radiation characteristics the widely applied in nuclear engineering MCNP package based on Monte-Carlo method [7] is used. In this package a geometrical model of the protective reinforced-concrete cask with four cylindrical sources, was developed (Fig.1).

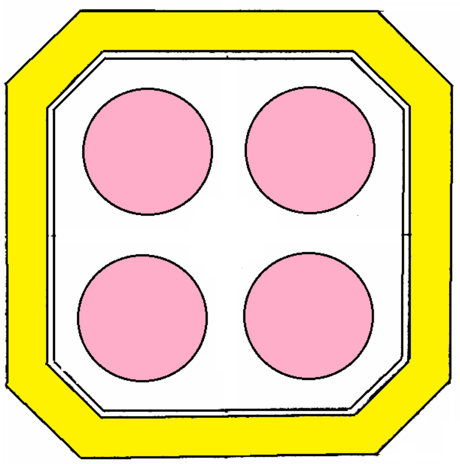

Fig.1. Model of the cask with RW

MCNP package allows calculating radiation characteristics (spectral composition, angular distributions, dose rate, etc.) at any distance and with any elemental and isotope composition of radioactive waste in the containers loaded into casks. A disadvantage of such method (as well as of the Monte-Carlo-based packages as a whole) is a long computation time, especially for the objects with rather complex geometry. As a rule, this fact does not allow optimizing considered processes. In our case possible loading of containers with RW of different isotope composition is an additional problem.

In the paper a combined technique using both Monte-Carlo and quasi-analytic methods in Serf_MC package is applied:

1. Monte-Carlo method was used to calculate spectral and angular distributions, as well as gamma flux density close to one surface of the cask loaded with containers filled with radioactive waste.

2. Further these distributions are used for calculation of γ–quanta transport at any distance by integrating the point sources over the cask surface.

It is assumed that each element of the source surface *dS* = dxdy emits γ–quanta with spectral – $I_E(E)$ and angular – $I_{AN}(\varphi)$ distribution, and density $n_\gamma(x,y)$ depending on (x,y) co-ordinates of *dS* element, where *E* is energy, φ is the angle of gamma quanta emission relative to the normal to the surface. Gamma flux with energy E_i in point P with co-ordinates X, Y, (Fig.2) is determined by the following equation:

$$N_i(z,X,Y,E_i) = I_E(E_i)\cdot\int_0^{Hy}\int_0^{Hx}\frac{n_\gamma(x,y)\cdot I_{AN}(\varphi)B(\mu(E_i)\cdot R)e^{-\mu(E_i)\cdot R}}{R^2}dxdy\,, \qquad , \qquad (1)$$

where *Hx* and *Hy* are width and height of a lateral surface of the cask, φ = φarcos (z/R) is the angle of gamma quantum emission to point P relative to the normal to the plane, $R=\sqrt{(x-X)^2+(y-Y)^2+z^2}$, *B, μ* are the build-up factor and the linear coefficient of γ–quanta absorption in the air, correspondingly.

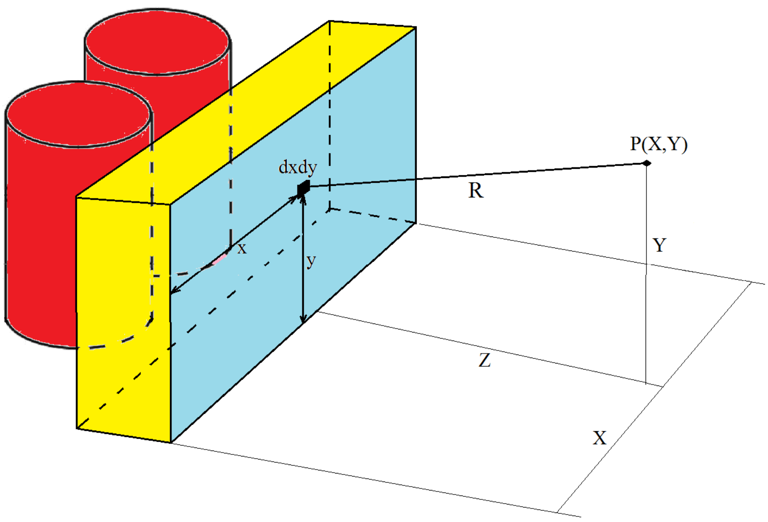

Fig.2. Calculation pattern of radiation from a lateral surface of the cask

It is known that at passage of γ-quanta with energy E_1 through objects they are absorbed and re-scattered into photons with lower energy. These processes cause decrease of the amount of γ–quanta with energy E_1 (at large thickness of the objects γ-quanta with the initial energy may not leave the object) and generation of a large number of low-energy photons ($E < E_1$). It is obvious, that change in the RW isotope composition causes change in the radiation characteristics. The following isotopes: ^{60}Co, ^{134}Cs, ^{137}Cs, ^{54}Mn and ^{110m}Ag make the main contribution to the radiation from RW. For the main types of concentrated RW (salt melt, pressed ash and SRW) the characteristics of radiation generated by these isotopes outside the cask are calculated.

In that case spectral composition of the external radiation $I_E(E)$ is determined by combined distribution of photons by energy produced by all the isotopes in the radioactive waste. For the salt melt (see Table 1) the spectral composition is determined by the following equation:

$$I_E(E) = I_{Co60}(E) + I_{Cs134}(E) + I_{Cs137}(E). \quad (2)$$

RESULTS OF CALCULATION

Fig.3 represents γ-quanta yields on the surface of the cask filled with salt melt calculated in package MCNP, a number of photons per 1 Bk activity of ^{60}Co, ^{134}Cs and ^{137}Cs isotopes.

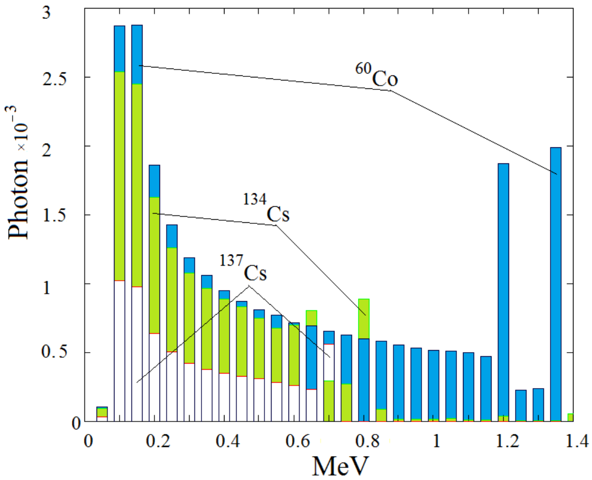

Fig.3. γ-quanta yields on the cask surface per 1 Bk activity of ^{60}Co, ^{134}Cs and ^{137}Cs isotopes

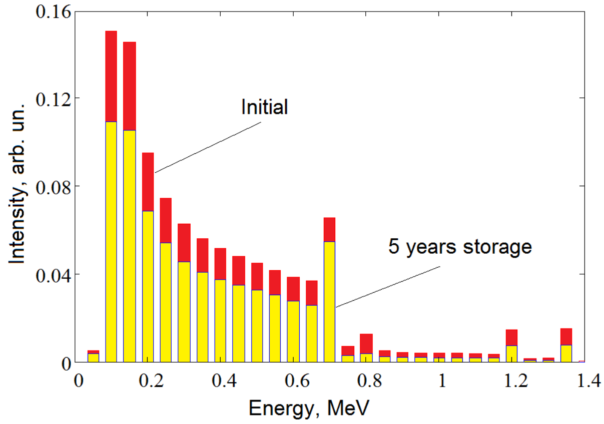

Fig.4. γ-quanta spectra produced by salt melt on the cask surface

The proposed method of summation by individual isotopes allows obtaining radiation spectral distribution by energy for any isotope composition for any storage time. Fig.4 shows spectral distributions caused by salt melt with initial isotopic composition of ^{60}Co – 15%, ^{134}Cs – 20%, ^{137}Cs – 65%, and after 5 year of their storage.

The data represented in Fig.3 show that γ-quanta spectral composition on the container surface differs considerably for each isotope. The calculations showed that as opposite to the energy spectra the external radiation angular distributions for different isotopes differ slightly. For example, when the cask is filled with salt melt γ-quanta angular distribution for all given isotopes is described with high accuracy by the model function:

$$I_{AN}(\varphi) \approx \cos^{Pa}(\varphi), \quad (3)$$

where Pa = 4.75. The uncertainty of the calculated data description by the model function is of a few percent within the angle range of 0 – 60°, with more than 99% of photons emitted by RW.

The dose rate values depending on the distance from the cask surface for different isotopes were obtained by equation (2) applying the Monte Carlo-calculated characteristics of radiation on the cask surface. To compare the results obtained when applying the combined method, the dose rate variations from the distance were calculated by Monte Carlo method (Fig.5).

In the calculations of photons flux in the air at distances of tens of meters from the source, direct registration of particles in the point is ineffective, and it is difficult to obtain statistically reliable results in a reasonable time. Therefore, special detection methods are applied. The "point detector" method is one of them [7]. When using the MCNP package the "point detector" method lies in the following: detection of particles is determined by the sphere, which leans on a cone with solid angle $d\Omega_P$, and whose radius is shrinking to zero. In this case the particles flux will be registered, if a particle scatters inside $d\Omega_P$ corner. A particle is not scattering when it is moving inside $d\Omega_P$ angle to the direction of sphere. This resulted in statistical uncertainty of the calculations in the range of 0.5 ÷ 2%.

Comparison of the obtained results shows that when using Serf_MC method the dose rate is overestimated as compared to the results obtained with Monte Carlo method. This is due to the fact that Serf_MC method is based on the "ray approximation" method where the scattering photons are not included. It is advisable to use Serf_MC method, since the calculations made for given task by this method are conservative, i.e. the dose rate overestimation is relatively small (from 20 to 70%). For comparison, calculation of similar dependences by the method of "volumetric integration of point sources" gives an overestimation of 150 - 600%. Fig.6 represents spatial distribution of dose rate produced by a lateral surface of the cask filled with salt melt.

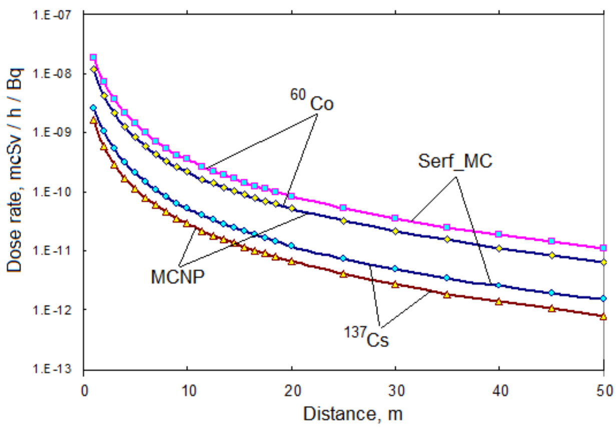

Fig.5. Dose rates produced by ^{60}Co and ^{137}Cs at different distances from the surface of the cask filled with salt melt

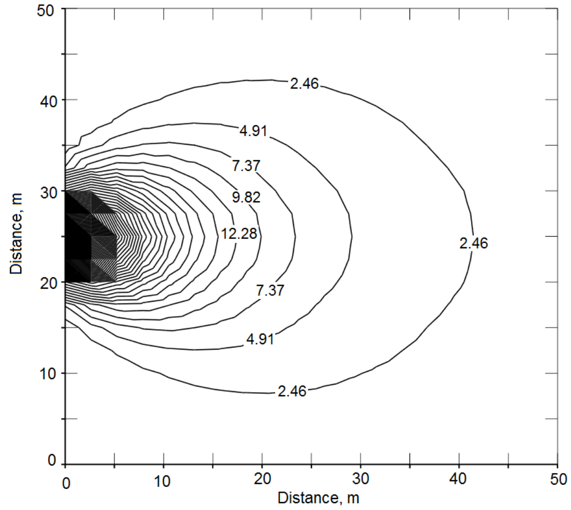

Fig.6. Isolines of dose rates produced by the surface of the cask filled with salt melt, (in arbitrary units)

The developed technique allows effective calculation of the dose rate distribution from the container filled with RW of different isotope composition and storage time, at any spatial point on the lateral side of the cask.

ASYMMETRIC CASKS LOADING

To minimize radiation from one of the cask sides in [8] the containers with radioactive waste were shifted to the cask walls and the space between the containers was filled, for example, with sand, Fig7. But such an arrangement for many reasons would complicate the technological process of radioactive waste handling.

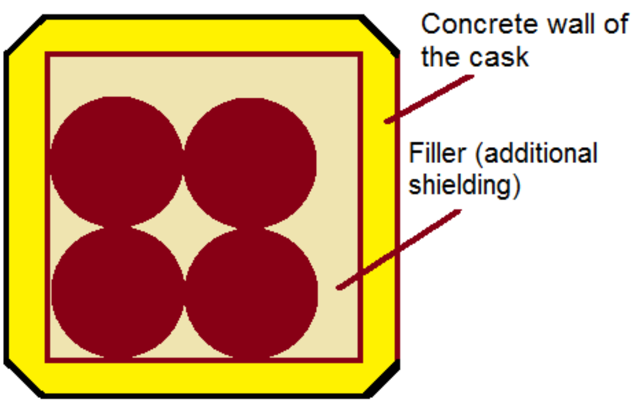

Fig.7. Cask loaded with shifted containers and filled by sand as additional biological shielding

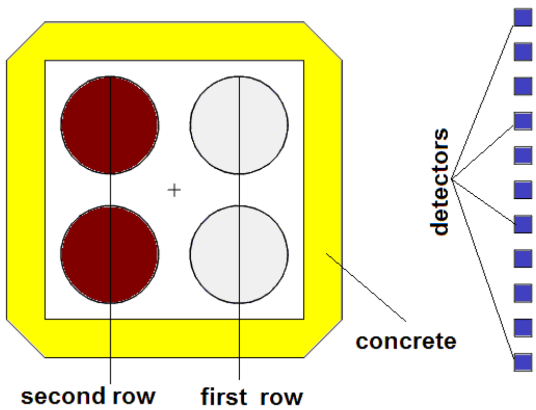

Fig.8. Cask loaded with containers filled with RW of different activity (low-active RW serving as biological shield)

Radioactive wastes stored at the Ukrainian NPPs are of rather different activity. Salt melt and pressed ash from the burned SRW and oil at the newly build radioactive waste reprocessing complex (RWRC) at Zaporozhye NPP are most active. As long as some RW are stored for 3,5 or more years, it is possible to load the casks with containers filled with RW of different activity so, that the containers with low-active RW from the 1st row serve as a shield for more active RW from the 2nd row, Fig.8 [3]

We have studied shielding properties of containers from the 1st row for different types of RW. When loading RW in the form of salt melt and pressed ash (RW density ~ 2 g/cm^3), the contribution of the 2nd row to the external radiation for ^{60}Co is about 10%, and for ^{137}Cs is about 6%. When the 1st row is loaded with pressed RW whose density runs up to ~ 4 g/cm^3, the contribution of the 2nd row is smaller. Fig.9 shows the dose rate distribution inside and around the cask for the containers with melt salt of different activity (the calculations performed with MCNP package).

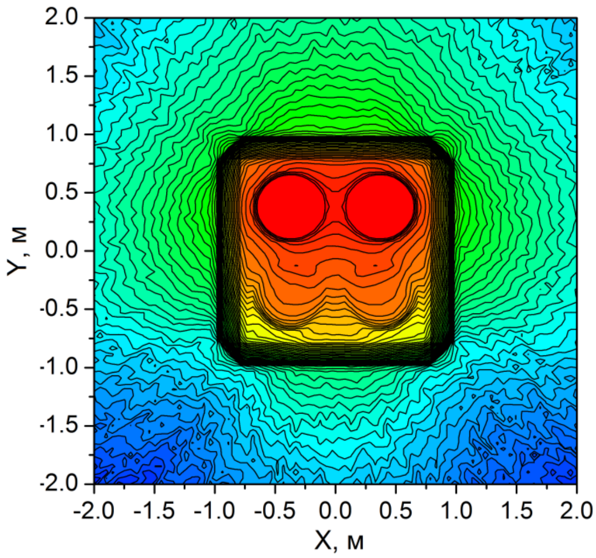

Fig.9. Dose rate distribution inside and around the cask for containers with melt salt of different activity

CONCLUSION

To calculate characteristics of radiation from complex objects containing radioactive waste a combined method using both Monte Carlo and quasi-analytic methods is proposed.

It is shown that summation of external radiation photons from all the isotopes in considered RW allows obtaining radiation characteristics (spectra, dose rate, etc.) for any isotope composition at any storage time.

It is shown that asymmetrical loading of casks with containers filled with RW of different activity allows reducing considerably the dose rate at one of the container sides.

The proposed techniques would allow reducing the effect of radiation-absorbed dose on the service personnel handling with radioactive waste as well as on the environment in the process of RW storage.

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