Investigation of Neutron Spectra Formation Behind the Radiation Shielding of a Shipping Cask With WWER-1000 Spent Nuclear Fuel

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ABSTRACT

The paper deals with the results of investigation of gamma-neutron radiation formation behind the radiation shielding of a shipping cask with WWER-1000 spent fuel. The cask short description is given. The radiation shielding structure, spent fuel characteristics, devices used are described. The way of neutron spectra reproduction and the estimate of different energy neutron dose commitment are presented which have been used to find the correction factors for standard radiometer indications.

Radiation safety provision is one of the main problems concerning the development and operation of shipping casks for spent nuclear fuel (SNF). In this case, as a rule, the radiation shielding efficiency control is based on the neutron- and gamma-radiation dose rate measurement results. At the same time it is known that indications of dose rate radiometers and dose meters used depend upon the energy radiation spectrum. It is known in the practice of radiation monitoring that energy dependences of neutron dose rate radiometer sensitivity (for example national РУС-У8, КДН-2, ДНА and foreign RUST instruments, etc.) are different from that of the maximum specific neutron dose equivalent hm (SV/cm^2/n) recommended by the National Radiation Safety Standards RSS-76/87, /I/ which is based on the analytical data of Sneider /2/ and Publication 21 of ICRP /3/.

These differences stipulate the need to introduce respective corrections for the instrument indications which reach some hundred percent for some types of radiometers. Knowlege of neutron and energy gamma-radiation spectra from shipping casks containing SNF is required for their design optimization.

A TK-10 shipping cask charged with 6 spent FAs of WWER-1000 reactor was used to investigate the neutron and gamma-spectra formation.

Spent nuclear fuel loaded into the cask was characterized by the following parameters:

- the campaign time - 825.5 eff.days;
- burn-up rate 37.2 GWD/t (U);
- cooling time 1265 days;
- initial enrichment in U-235 is 2.4% (18FE); 3% (48FE), 3.3% (251 FE).

Constructionally the TK-10 cask is a cylinder (∅2000×5880 mm), with 360 mm thick steel inner shielding screen and 20 mm thick steel outer shielding enclosure with the gap between them filled with antifreeze (water) forming the 120 mm thick neutron shield.

The radiation field of the cask charged with long-cooled spent nuclear fuel is formed by gamma-radiation of nuclear fission products, radionuclides of corrosion and activation origin, high-energy gamma-quanta of capture radiation on hydrogen nuclei (E_γ= 2.23 MeV), iron (E_γ= 7.64; 8.89; 9.3 MeV), gamma-quanta arising in neutron inelastic scattering on oxygen (E_γ= 6.13 MeV), etc.; as well as by neutrons mainly originated from Cm-244 spontaneous fission.

In experiments with the TK-10 cask the influence of neutron shield on neutron spectrum and gamma-radiation formation was also studied. Measurements in this case were performed both directly on the cask surface and 2m apart from it.

For neutron spectra investigation multisphere spectrometer was used which contains 6 spherical polyethylene moderators, ⌀ 50, 100, 150, 200, 250, and 300mm, a thermal neutron ДНМ-29Б and a ФЭУ-II4 type scintillation detector, a LP-4700 (Nokia) analyzer. The thermal neutron detector is a plastic light guide (⌀10×20mm) with dispersed in a thin layer over the end face and the side surface phosphor granules based on ZnSAg with boron. For investigation with the TK-10 cask this type of detector was preferred to the LiI/Eu/ crystal due to its low sensitivity to gamma-radiation which came to about 8.5 mR/h. The shape of this detector line unlike that of the LiI/Eu/ crystal allows to discriminate reliably by amplitude the pulses from neutrons and gamma-quanta /4/.

The task to reproduce the neutron spectra-shape , f/u/, based on the results of the counting rates, Ni, measured with a set of moderators (i=1.2,......n) comes to solving of the following integral equation system:

$$N_i = \int_{U_1}^{U_2} K_i(U) f(u)\, du \qquad (1)$$

where: U_1 and U_2 - boundaries of lethargy range (u=lgE) in which the spectrum is reproduced;

E - neutron energy;

$K_i(U)$ - detector sensitivity function in the moderator "i".

By breaking the lethargy range into m=46 equal intervals the system of equations (1) is transformed into the following matrix equation:

$$\bar{N} = \hat{K} \bar{f} \qquad (2)$$

where: $\bar{N}$ - vector with N_i components;

$\hat{K}$ - (N×m) matrix of sensitivity functions with matrix elements Kij;

$\bar{f}$ - vector of the spectrum to be reproduced with components fj.

Matrix elements Kij and components fj equal respectively:

$$K_{ij} = \frac{\int_{u_j}^{u_{j+1}} K_i(u)\, f(u)\, du}{\int_{u_j}^{u_{j+1}} f(u)\, du} (u_{j+1} - u_j)$$

$$f_j = \frac{\int_{u_j}^{u_{j+1}} f(u)\, du}{u_{j+1} - u_j} \qquad (3)$$

As the spectrum in its nature is subject to statistical fluctiations, there is no sense to look for a more exact equation (2) solution, and more relible results are expected with the statistical approach to its solution. In this case the vector used for solution should minimize the following functional:

$$F(\bar{f}) = \sum_{i=1}^{n} \left(N_i - \sum_{j=1}^{m} K_{ij} \cdot f_j\right)^2 \cdot D_i^{-1} \leq \xi^2 \qquad (4)$$

where: D_i - registered counting rate dispersions;

ξ^2 - a small enough value giving the statistical criterion significance level.

Algorithm of the F($\bar{f}$) functional minimization is based on the gradient integration method allowing to build the iteration process. For calculational formulae in the algorithm a somewhat changed writing form is used instead of (4):

$$F(\bar{f}) = \frac{1}{2}(\hat{A}\bar{f}, \bar{f}) - (\bar{B}, \bar{f}) + C \qquad (5)$$

where: $\hat{A}$ - (n×m) matrix with $A_{ij} = 2\sum_{i=1}^{n} K_{ki} \cdot K_{kj} \cdot D_k^{-1}$ elements;

$\bar{B}$ - vector with components

In these designations the main formulae of the minimization algorithm by the method of integrated gradients are:

$$\bar{f}^{l+1} = \bar{f}^{l} + a^{l+1} \cdot \bar{V}^{l+1}$$

$$\bar{V}^{l+1} \equiv \mathrm{grad}\, F(\bar{f}^{l}) = (\hat{A}, \bar{f}^{l}) - B \qquad (6)$$

In the algorithm shortened iterations are used, with the coefficients a^{l+1} being determined as:

$$a^{l+1} = -\frac{(\bar{V}^{l}, \bar{V}^{l})}{(\hat{A}\bar{V}^{l}, \bar{V}^{l})} + \sqrt{\left[\frac{(\bar{V}^{l}, \bar{V}^{l})}{(\hat{A}\bar{V}^{l}, \bar{V}^{l})}\right]^2 - 2\frac{F(\bar{f}^{l}) - \varepsilon^2}{(\hat{A}\bar{V}^{l}, \bar{V}^{l})}} \qquad (7)$$

The quality of neutron spectra reproduction depends to a large extent on the choice of the initial approach spectrum $\bar{f}^{\circ}$. In our problem for initial approach the uranimum neutron fission spectrum behind the iron/water shield and californium - 252 spectrum from a ϕ 600 mm iron sphere /5/ were taken.

The iteration process ceases when the condition (4) is satisfied or when the relative velocity of the functional $F(\bar{f}^{l})$ change becomes less than a given parameter α, i.e.:

$$\left| \frac{F(\bar{f}^{l}) - F(\bar{f}^{l+1})}{F(\bar{f}^{l+1})} \right| \leqslant \alpha \qquad (8)$$

The calculations assume $\alpha = 0.01$

If in the iteration process a vector $\bar{f}^{l}$ component f_j^{e} becomes negative, then variation against this component ceases and the null value is assigned to it. This procedure allows to provide the nonnegativity condition of the neutron spectrum reproduced.

To avoid oscillations resulted from the use of limited set of moderators in the multisphere spectrometer it was found expedient (and this is accounted in the algorithm) to correct the spectrum shape by its smooting. Different smooting procedures - against 5 or 7 values are used. For the smoothness indication of the spectrum to be reproduced the Y value estimated by the formula:

$$Y=\sum_{j=1}^{K}\left(f_{j+1}-f_j\right)^2 / \sum_{j=1}^{K} f_j^2 \qquad (9)$$

is accepted:
where: K - number of the ΔU_j interval locking the smoothing area.

The index Y is calculated for the $\bar{f}^e$ vector on which the next minimization cycle of the functional $F(\bar{f}^e)$ ceases (8). The spectrum reproduction process is over when the respective rate of the Y index change per minimization cycle becomes less than a given parameter β , i.e. when the conditions are specified:

$$\frac{Y^{e_1}-Y^{e_2}}{Y^{e_2}} \leq \beta \qquad (10)$$

where: l_1 and l_2 are the numbers of iterations on which two last minimization cycles ceased.

The resulted vector $\bar{f}$ describes the shape of the neutron spectrum to be reproduced and satisfies to some extent all the requirements of the statistical approach.

The algorithm allows to calculate the following values based on the spectrum reproduced:

1) Distributions of different energy neutron dose commitments:

$$\frac{\Delta D(E_i)}{D_\Sigma} = \frac{h_m(E_j)\cdot f(E_j)}{\sum_{j=1}^{m=46} h_m(E_j)\cdot f(E_j)} \qquad (11)$$

2) Correction factors for different neutron dose rate radiometer indications

$$K_{non} = \frac{\sum_{j=1}^{m=46} f(E_j)\cdot K_{np.}(E_j)}{\sum_{j=1}^{m=46} f(E_j)\cdot h_m(E_j)} \qquad (12)$$

where $K_{\text{пр}}$ - instrument sensitivity.

3) Values of the average isotropy factors

$$\bar{J} = \frac{\sum_{j=1}^{m=46} h_m(E_j)\cdot f(E_j)}{\sum_{j=1}^{m=46} \frac{h_m(E_j)\cdot f(E_j)}{J(E_j)}} \qquad (13)$$

where $I(E_j)$ - isotropy factor for monoenergy neutron of energy E_j /I/.

For spectra reproduction energy dependencies of moderator sensitivies as recommended by ICRP /6/ were used, which were scaled for the dimensions of moderators and thermal neutron detector ДМН-29Б used. The multisphere spectrometer was calibrated against isotope sources Pu-Be and ^{252}Cf.

Figs. 1 and 2 show typical shapes of the reproduced neutron spectra for the TK-10 cask with the jacket not filled with water (measuring point N2) as well as with the water-filled jacket of the neutron shield (measuring point N5) and distribution of different energy neutron dose commitments. Table 1 shows neutron dose commitment for three energy groups ($10^{-2} - 10^{4}$eV, $10^{4} - 1.6\times10^{6}$eV and higher than 1.6×10^{6}eV) for some measuring points as well as average isotropy factors $\bar{I}$ calculated on the basis of the spectra measured.

The analysis of the spectra found shows that in case of the neutron shield (water in the cask jacket) the neutron spectrum "softens", the share of thermal neutrons and that of intermediate energy neutrons as well as their dose commitment increase from 5-7% to 24-27%. At the same time, with efficient attenuation of intermediate energy neutron flux and significant decrease of neutron dose rate as a whole (see Tab.2) 120mm water layer is not sufficient for fast neutron thermalization, which results in their respective dose commitment increase. It should be also noted, that the proper choose of spectra fo as the initial approaches gave sufficiently good agreeing shapes of neutron spectra.

Indications of different neutron dose rate radiometers, КДН-2, РУС-У8, 2202-Д ("Afora" company), were compared with the data calculated on the basis of the spectra. Table 2 shows correction factors for radiometer indications. For the КДН-2 radiometer K_{corr} equals in average 0.59 and 0.45 for the neutron spectra from the TK-10 cask with the neutron shield jackets without water filling and with water filling, respectively. For the РУС-У8 radiometer $K_{corr.}$ equals in average 0.39 and 0.29 and for 2202-Д radiometer it is about 1.03.

It should be noted that though the 2202-Д radiometer indications are to the least extent effected by the neutron spectra differences due to its (radiometer) similar to the Sneider curve energy dependence of the sensitivity function, this radiometer is of high sensitivity anisotropy.

Using the scintillation gamma-spectrometer with a NaI (Te) $\varnothing$ 40×40mm crystal the TK-10 cask gamma-radiation spectra were investigated. Fig.3 shows typical instrument spectra, and Fig.4 - distribution of different energy gamma-radiation dose commitment. Investigations of gamma-spectra showed, that water filling of the cask neutron shielding jacket though it results in increase of capture gamma-radiation on hydrogen and iron by 20 and 60%, respectively, at the same time it significantly decreases the neutron flux from the cask (by~10^2) and gamma-radiation from oxygen ([illegible]).

Neutron and gamma-radiation intensity from the [illegible] cask changes along the cask length with the maximum in the middle of the cask (see Figs. 6, 7, 8). Maximum values of the ionizing radiation dose rate equivalent from the TK-10 cask loaded with spent fuel of the parameters shown above are not high than the permissible values of the international regulations. Scaled values of radiation levels for fuel with the maximum design parameters for the WWER-1000 reactor do not exceed the permissible transport standards either.

LITERATURE

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Table 1

Different energy neutron dose commitments (%) for TK-10 cask

Number of measuring point	Coordinates of measuring point			Energy range, eV			Isotropy factor
	Axis	Distance, m	Height m	$10^{-2}-10^{4}$	$10^{4}-1.6\times10^{6}$	10^{6}	
	Neutron shielding jacket without water filling						
1	III	2	1	7.2* 7.3	92.8 92.7	- -	2.91 2.98
2	I	2	3	5.1 5.4	94.9 94.6	- -	2.96 2.94
	Neutron shielding jacket with water filling						
3	III	on surface	1	2.38	76.2	-	3.3
4	IY	on surface	1	23.9	75.6	0.5	3.28
5	I	on surface	3	25.2	60.2	14.6	2.98
6	I	2	3	27.7	52.8	19.5	2.96

* The upper figures mean that for initial approximation, 252Cf spectrum from an iron sphere is taken.

The lower figures mean that for initial approximation, the fission spectrum behind the iron/water shielding is taken.

Table 2

Neutron dose rate values in spectrum measuring points according to indications of РУС-38, КДН-2 and 2202-Д radiometers and correction factors values

Coordinates of measuring point			Radiometer type								
			РУС-У8			КДН-2			2202Д		
Axis	Distance, m	Height, m	Experim value, µrem/s	$K_{corr.}$	Corrected value, nSv/s	Experim value, µ rem/s	$K_{corr.}$	Corrected value, nSv/s	Experim value, µ rem/s	$K_{corr.}$	Corrected value, nSv/s
Neutron shielding jacket without water filling											
III	2	1	16	0.386	62	14	0.593	83	30	1.05	315
I	2	3	13	0.390	51	14	0.596	83.4	35	1.06	371
Neutron shielding jacket with water filling											
III	on surface	1	0.3	0.288	0.86	0.45	0.447	2	0.6	1.01	6.06
IY	on surface	1	0.2	0.289	0.58	0.22	0.448	0.99	0.45	1.01	4.55
I	on surface	3	0.5	0.300	1.5	0.40	0.459	1.84	0.80	1.03	8.24
I	2	3	0.15	0.287	0.43	0.22	0.439	0.97	0.27	1.034	2.8

Neutron spectrum from TK-10 cask with a jacket without water filling (a) and different energy neutron dose commitment (b) at a point on the axis III 2m distance from surface at 1m height (the initial approximation is the Uranium fission neutron spectrum behind the iron/water shielding)

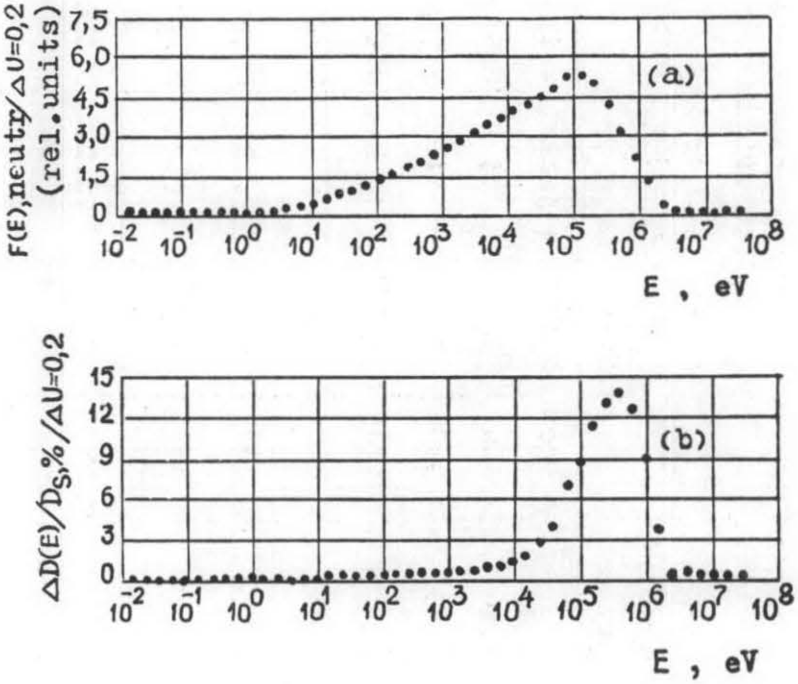

Fig.1

Neutron spectrum from TK-10 cask with a water-filled jacket(a) and different energy neutron dose commitment (b) at a point on the axis Ⅲ on the cask surface at 1m height (the initial approximation is the Uranium fission neutron spectrum behind the iron/water shielding)

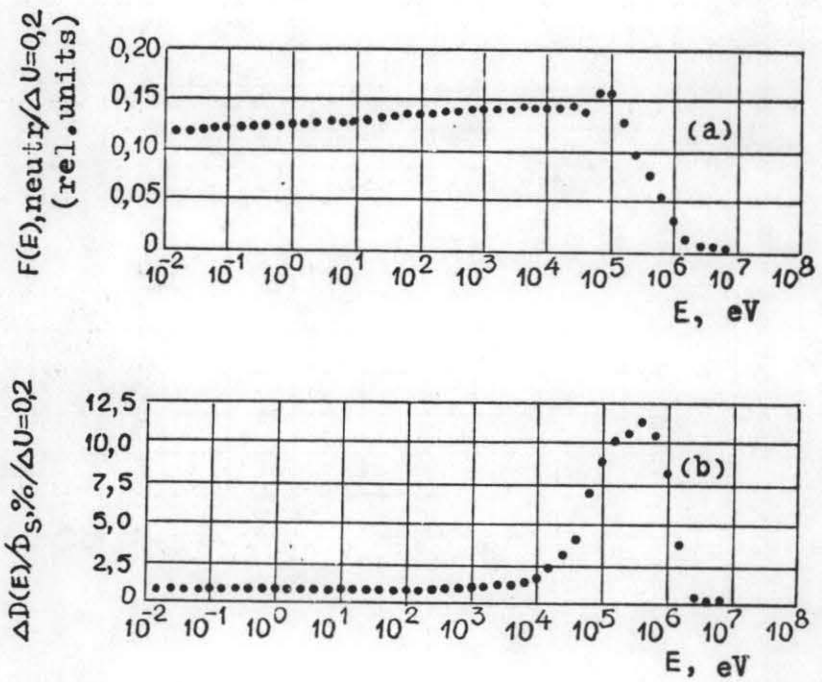

Fig.2

Instrument gamma-spectra measured on the axis IY 2m distance from the cask surface at 1m height

1 - a jacket without water filling;

2 - a jacket with water filling;

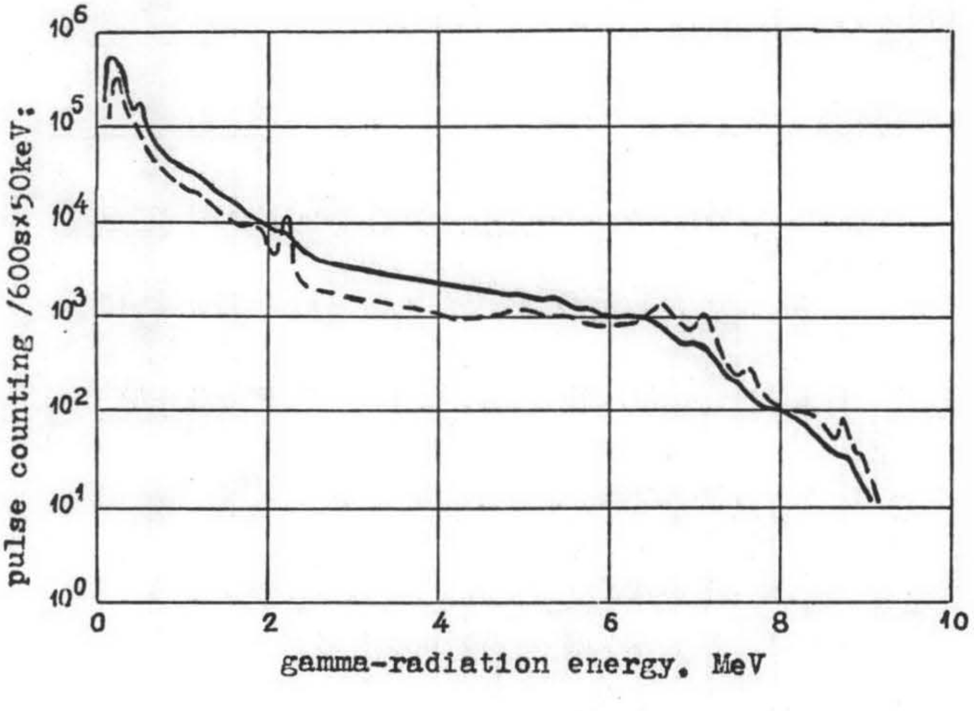

Fig.3

Energy distributions of dose commitments for gamma-radiation from TK-10 cask (the energy range width is 0.4 MeV)

——————— a jacket without water filling

--------- a jacket with water filling

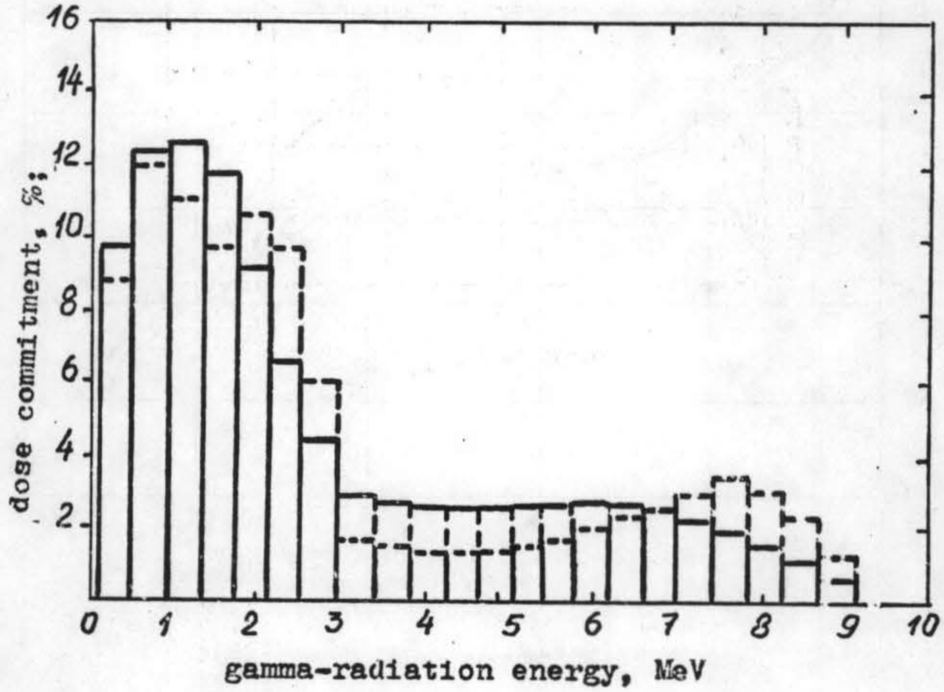

Fig.4

Neutron dose rate distribution (corrected indications of КДН -2 radiometer) on the side surface of TK-10 cask on the axis IY with the neutron shielding jacket with water filling

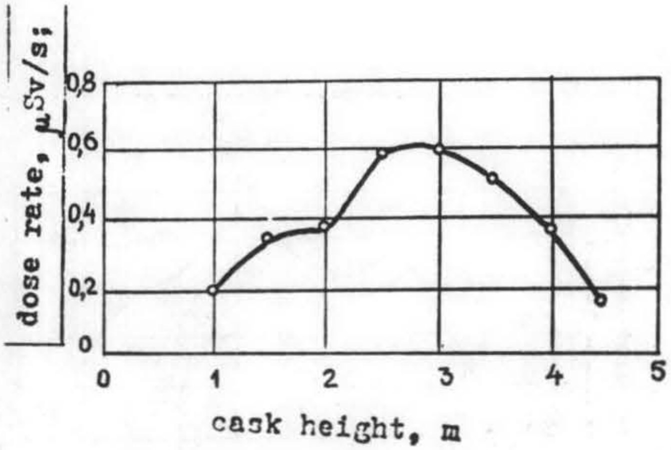

Fig.5

Neutron dose rate distribution (corrected indications of КДН -2 radiometer) on the side surface of TK-10 cask on the axis IY with the neutron shielding jacket without water filling

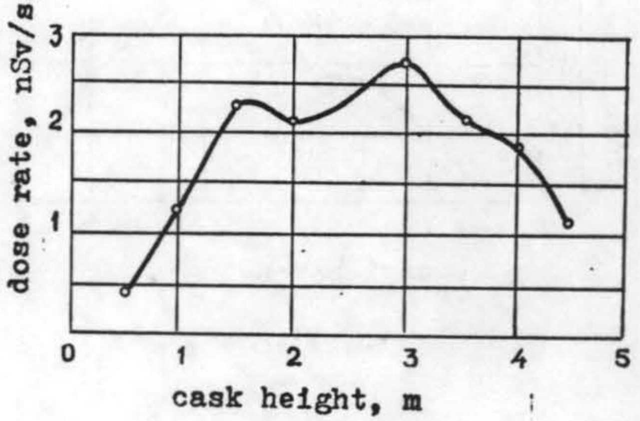

Fig.6

Gamma-radiation dose rate distribution on the side surface of TK-10 cask according to indications of ДРГ-01 instrument

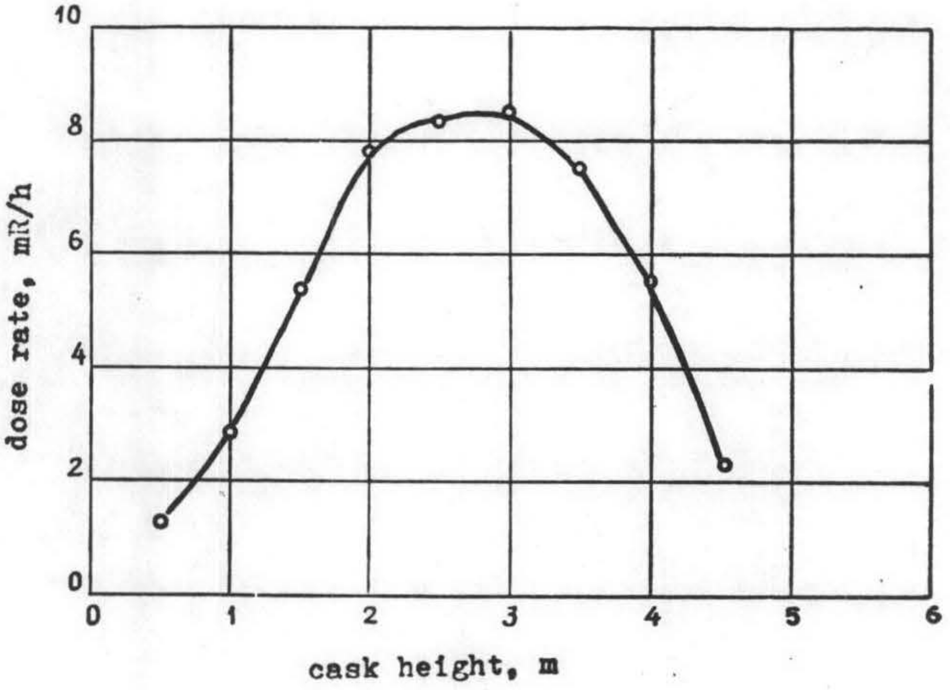

Fig.7

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Cask Materials