CALCULATION OF GAMMA-RAY DOSE RATE FROM STORAGE CASK WITH RADIOACTIVE SLUDGE

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ABSTRACT

Sludge contaminated by ^{137}Cs and ^{60}Co collected from bottom of the spent fuel storage pool of the RA research reactor is conditioned and stored as low-level radioactive waste in specially designed casks. This paper describes an attempt to estimate the ambient dose equivalent rate of gamma-rays at the cask surfaces filled with the conditioned sludge by using the MCNP code and compare the result to the measuring data. Various data exist for measured activities of ^{137}Cs and ^{60}Co in sludge, depending on water content. Precise data for materials compositions and their fraction, used during sludge conditioning process, does not exist. Consequently, obtained calculation results show wide discrepancies compared to measured ambient equivalent dose rate of gamma-rays at surfaces of the storage cask. This indicates that better knowledge of composition of wet sludge and its mixture with cement, together with precise data for activity measurement conditions, is necessary to obtain more reliable calculation results.

Key words: MCNP, LLW, sludge, RA reactor storage pool, gamma-ray dose rate

1. Introduction

Spent nuclear fuel elements, created during operation of the RA reactor [1] from 1959 to 1984, are stored in the temporary storage pool within the reactor building. In the first years of the reactor operation, spent fuel elements were stored in 300 (made in Russia) stainless steel channel-type containers, filled with demineralised water. One stainless steel container was able to accept only one fuel channel with 10 fuel elements. New storage barrels with much higher fuel elements capacity were designed from aluminium in aim to increase the storage pool capacity in early sixties. These barrels were filled with de-mineralised water and sealed. Cadmium strips were inserted in few positions in the barrels to assure sub-criticality.

About 5000 oldest LEU spent fuel elements were repacked from stainless steel containers into 30 sealed aluminium barrels, from the beginning of sixties until 1984. About 1000 HEU spent fuel elements and 1600 LEU spent fuel elements remained in stainless steel containers in the storage pool, up to now. The total activity of all spent LEU and HEU fuel elements in the pool is estimated [2] at value of 2500 ± 250 TBq at end of 1988. Most of the activity originates from ^{137}Cs and ^{90}Sr nuclides (about 99%), while remaining activity (about 1%) is attributed to ^{85}Kr nuclide.

2. Sludge in the spent fuel storage pool of the RA reactor

In aim to provide radiological protection and cooling of the spent fuel elements, all four basins of the storage pool were filled by tap water since 1960 up to 1995. Chemical or mechanical purification of the water was not provided. There was none monitoring of chemical properties or radiation activities of the pool water during that period, i.e., there were no personnel within the RA reactor department or in the Institute responsible for the pool regular inspection. Written regulations rules regarding work, maintenance and monitoring within the RA spent fuel storage pool area were not existed as well.

Inspection of the storage pool in 1994 discovered thick corrosion deposits at surfaces of basins walls, aluminium barrels, stainless steel containers and sludge at the bottom of the pool. The situation was examined thoroughly in 1995-1996. Samples of the pool water and sludge were taken out from different location and measured to determine material composition, chemical parameters and activity. Analyses [3] have shown that the pool water is high corrosive to aluminium alloys (pH $\approx$ 8.5, conductivity in range 500 - 800 μS/cm). Pool water samples showed also specific gamma-ray activity of 80 – 90 kBq/L, originated primary from ^{137}Cs nuclide. Gamma-ray activities of various samples of wet sludge (‘ws’) and dry sludge (‘ds’) were measured by coaxial Ge gamma-ray spectrometers in the Vinča Institue [4, 6, 13] and in the IAEA laboratories [5]. Results of these measurements, together with basic description of the samples, are shown in Table 1, in the forms as they were reported.

Table 1. *Measured gamma-ray activity in the sludge samples*

Sludge sample (description)	Activity ± uncertainty (1σ) ^{137}Cs	^{60}Co
ws #1 [4]:	1.80 ± 0.20 kBq/mL	15.0 ± 1.5 kBq/L
ws #1 [5]:	1.83 ± 0.27 kBq/mL	
ws #2 [5]:	0.94 ± 0.14 kBq/mL	
ds #1 [6] (ρ (ds#1) = 0.64 g/cm^3):	13.4 ± 2.3 kBq/g	122.7 ± 21.2 Bq/g
ds #2&3 [5] (ρ (ds#2, ds#3) = 0.77-0.95 g/cm^3):	0.54 - 0.79 kBq/mL	1.9 - 7.2 Bq/mL
ds #4-6 [13] (ρ (ds#4-6) = 1.86 g/cm^3):	102 - 365 kBq/g ± 12%	1.2 - 1.95 kBq/g ± 10%
Avg(3 ds: #4-6):	247 ± 77 kBq/g	1.6 ± 0.2 kBq/g

Recent inspections of various water samples, taken from spent fuel aluminium storage barrels and stainless steel containers, confirmed that ^{137}Cs activity in the pool water originates from leakage of fission products. It led to conclusion that corrosion process damaged the aluminium cladding (1 mm thick) of the spent fuel elements and aluminium walls of storage barrels.

3 Conditioning and storage of the sludge

Total volume of the sludge in the pool is estimated to be about 3 m^3, based on average sludge height on the bottom of the pool and area of the pool surfaces. It was concluded that the sludge could be treated as the LLW, according to the results of activity measurement in the sludge samples.

A technology [7] was developed for sludge immobilisation and conditioning in a cement matrix, inside casks produced using the standard 200-litre metal barrels. Thickness of designed concrete walls and bottom layer is between 7 cm and 8 cm. A 1 cm thick plastic tube covers entire inner side of the cylindrical concrete wall. The tube serves as a first barrier in preventing radionuclides leaching from radioactive sludge, immobilised in a cement matrix. Inner wall was covered with thin layer of epoxy resin too, in order to prevent or reduce radionuclide leaching. Available volume for radioactive waste storage, in such designed casks, is 80 L ± 5 L.

The sludge is removed from the pool bottom by pump and inserted to the sedimentation vessel, placed in the storage pool area (Figure 1). Approximately 60 L ~ 65 L of sludge was poured at a time from the vessel into the cask. As soon as a cask is filled up, it is hermetically covered with a metal lid and transported to the laboratory for sludge conditioning where additional settling of sludge is allowed. Separated water is pumped out into a plastic can and returned back to the RA reactor spent fuel storage pool. Through the second stage of the sludge settling, volume of the sludge in the cask has been reduced to about 40 L.

The existing pilot cement mixer was reconstructed to enable simple placing a barrel containing the sludge on its platform without a risk of spilling. Rooms for conditioning the sludge in a cement matrix, supplied with independent ventilation system, and for storing the casks during the period needed for cement hardening, have been arranged. When the cask with settled sludge is placed on the platform of the mixer, an amount of about 90 kg of cement (PC-45 MPa) was added into the cask. Mechanical manipulator (Figure 2) was used to mix this mixture until a homogeneous substance (‘matrix’) was obtained [8]. This technology for sludge conditioning eliminates all the risks related to pouring the sludge into the concrete mixer and pouring the cement-sludge mixture into the metal barrel. The barrel with the homogenised mixture is removed from the mixer platform and placed in a separate room to harden mixture for about 2 days. In final stage of conditioning of sludge immobilised in a cement matrix, the cask is the covered with the concrete cork. When concrete cork is hardened, the cask is covered by a metal lid that is fixed by screws.

Fig 1 Sludge sedimentation vessel and the cask

Fig. 2. Pilot mixer in the cask

4. Description of the model used for calculations

Three-dimensional (3D) model of the sludge storage metal cask, filled with matrix of conditioned sludge-cement mixture, based on geometrical dimensions and known or assumed composition of materials, is developed. Vertical and horizontal cross-sections of the 3D model of the sludge storage cask used in calculation are given in Figures 3.

The cask is designed in the “standard metal 200 L barrel” that has outer diameter 57.0 cm, total height 87.5 cm and wall thickness of 0.1 cm. Volume of the barrel is about 220 L. Material composition of the barrel walls is not known and for the calculation purposes, a pure iron with theoretical density of 7.874 g/cm^3, (atom concentration of $8.491 \cdot 10^{22}$ cm^{-3}) is assumed

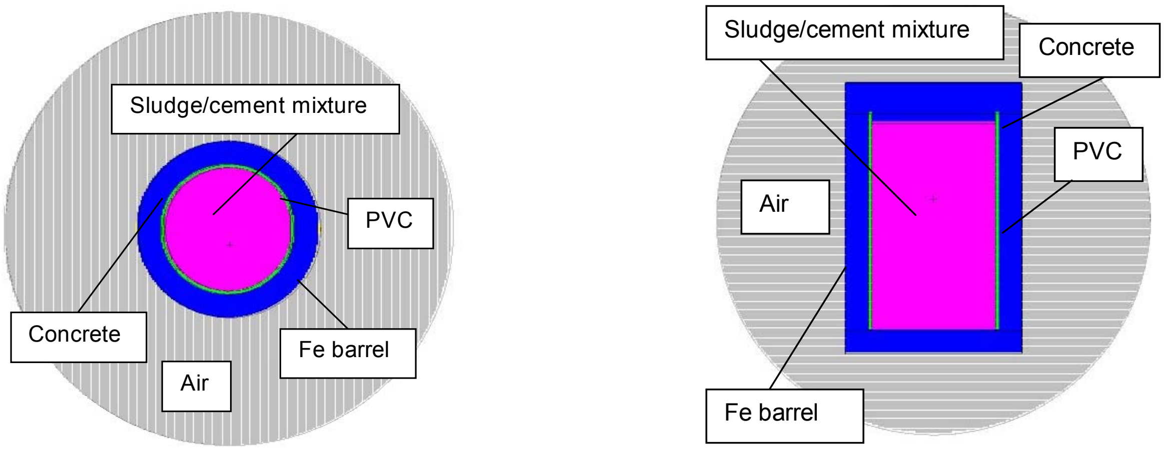

Fig. 3. Cross section of 3D cask model used in calculation

Inner sides and bottom of the barrel are filled by concrete which exact composition is not known. Density is measured as 2.35 g/cm^3. In the 3D cask model, “KENO code Regular Concrete Standard Mix” with 2.30 g/cm^3 density and nuclide composition [9] is used. Homogenous distribution of the concrete with uniform thickness of 7.5 cm is assumed. Space above matrix in the cask is completely covered by concrete (thickness 13.0 cm in the inner ring, above the matrix, and 10.0 cm above outer concrete ring). No air gaps are assumed to exist within cask in the 3D cask model.

Central zone of the cask is separated from the surrounding annular concrete ring by plastic cylindrical tube. Height of the tube is 67.0 cm, inner diameter is 40.0 cm, and wall thickness is 1.0 cm. The exact composition of the tube material used is not known. Pure PVC (C_2H_3Cl), with density of 1.65 g/cm^3 and nuclide composition given in [9], is chosen in the 3D cask model.

Sludge composition is determined by radiochemical analysis of few sludge samples in the IAEA laboratory [5]. The main constituent of the sludge is Fe_2O_3 (average 83.60% by weight). The remaining components of the sludge are (weight percents are given): Al_2O_3 (5.19%), SiO_2 (1.86%), Cr_2O_3 (1.86%), MnO (1.11%), PbO (1.78%) and CaO (4.60%, including other ‘minor impurities’). Exact density of ‘dry sludge’ is not known and depends of water bounded in ‘dry sludge’. Measured density values were in range from 0.64 g/cm^3 to 1.04 g/cm^3. Calculated maximum density value for “100% dry sludge” is 4.85 g/cm^3.

According to the sludge conditioning procedure, dried (but not 100% dry!) sludge is homogeneously mixed with cement in (assumed) volume equal ratio that gives the mass ration of 1:1.4. Procedure proposes the best mass ratio as 1:1.8. In reality, about 90 kg of cement is added to about 40 L of sludge/water mush (“dried sludge”), well mixed, and left to harden for few days. Volume of this mixture, prepared inside central space of the storage cask, is about 85 L, based on an assumption that there was no contraction of mass and volume of materials used for mixing. Measured density of one mixture sample of such conditioned sludge/cement, shaped as a cube with a 10 cm edge length, is (1.80 ± 0.05) g/cm^3.

Exact composition of the cement used for conditioning is not known either, except that it is Portland type cement. For the calculations purposes, the Portland cement with density of 2.1 g/cm^3 and composition given in [10] is chosen (weight percent are given): MgO (2.0%), SiO_2 (23.0%), Al_2O_3 (8.0%), Fe_2O_3 (4.0%) and CaO (63.0%). In this study, different to the previous one [13], a new assumption is adopted for water volume fraction in the wet sludge. Ratio of water to 100% dry sludge is set to 7:1, that is very near to measured value in one sample of the sludge. Volume fraction of 100% dry sludge in the mixture is, consequently, 7%, only. From these data, given above, density of sludge/cement mixture ('matrix') is calculated as 1.82 g/cm^3 and atom composition of the sludge/cement matrix is determined for use in calculations (Table 2).

Table 2. *Composition data for sludge/cement matrix*

Component	Density [g/cm^3]	Mass [kg]	Volume [L]
100% Dry Sludge	4.85	28.58	5.89
Water	1.00	36.30	36.30
Cement	2.10	90.00	42.10
Total	1.80	154.78	84.19

It is assumed, in 3D calculation model of the cask, that whole storage cask is surrounded by an idealised large air sphere (Figure 3). Air density of 1.20 kg/m^3 and nuclide composition given in [11] is used. Influence of gamma-ray reflections from ground surface and surrounding objects at gamma-ray spectrum and corresponding ambient equivalent dose rate in the calculating points is neglected in this 3D cask model. Atom concentrations of materials used in 3D model of the storage cask are given in Table 3.

Table 3. *Atom composition of materials used in 3D model*

Atom	Atom concentration [10^{24} cm^{-3}]			
	Air	PVC	Concrete	Matrix
H		0.04771	0.01374	0.028453
C		0.03180		
N	$4.336\ 10^{-5}$			
O	$1.019\ 10^{-5}$		0.04606	0.032458
Na			0.00175	
Mg				0.000316
Al			0.00175	0.001206
Si			0.01662	0.002503
Cl		0.01590		
Ar	$1.653\ 10^{-7}$			
Ca			0.00152	0.007323
Cr				0.000050
Mn				0.000032
Fe			0.00035	0.000244
Pb				0.000016

It is accepted that, according to measuring data, source of gamma-rays in the sludge of the storage cask originates from ^{137}Cs nuclide (E_γ = 0.662 MeV, yield = 0.851) and ^{60}Co nuclide (E_γ = 1.1732 MeV, yield = 0.99857 and E_γ = 1.3325 MeV, yield = 0.99983), only. Homogeneously distributed source of gamma-rays within volume of the sludge/cement matrix in the cask is assumed in the 3D model. Intensity of particular gamma-ray line in the sludge/cement mixture is determined according to the measured activity data obtained for the different sludge samples and given in Table 4.

Table 4. *Intensity of gamma-ray source (I_γ) in the cask*

Gamma-ray Source		I_γ [gamma-ray per second]		
Nuclide	E_γ [MeV]	Ref. [13] Avg	Ref. [5]	Ref. [6]
^{137}Cs	0.662	$6.082 \cdot 10^{9}$	$6.449 \cdot 10^{7}$	$3.285 \cdot 10^{8}$
^{60}Co	1.1732	$4.711 \cdot 10^{7}$	$4.204 \cdot 10^{5}$	$3.512 \cdot 10^{6}$
	1.3325	$4.716 \cdot 10^{7}$	$4.209 \cdot 10^{5}$	$3.516 \cdot 10^{6}$
Total intensity:		$6.177 \cdot 10^{9}$	$6.533 \cdot 10^{7}$	$3.355 \cdot 10^{8}$

5. Results of calculations by MCNP code

Calculations are carried out by using MCNP [12], well-known Monte Carlo based code, with the MCPLIB continuous energy photon data library. The gamma-ray flux is determined in 13 energy groups in range from 10 keV to 1.5 MeV. It is calculated at two points in air near the cask surfaces, at top and at side (Figure 4). For that purpose, the F4 (track length estimator) and the F5 (detector) tallies of the MCNP code are used. Gamma-ray flux is converted to the gamma-ray ambient dose equivalent rates using ICRP-21 conversion factors [12]. The code is run for 50 million gamma-ray histories to obtain satisfactory low statistical errors in group spectrum (< 1%) and ambient dose equivalent rate (< 0.1%). Calculations with 5 million gamma-ray histories are carried out for the cube (10 cm edge length) of the sludge/cement mixture, too. Values of calculated gamma-ray ambient dose equivalent rates are compared to the measured values at the same spots of surfaces of the cube.

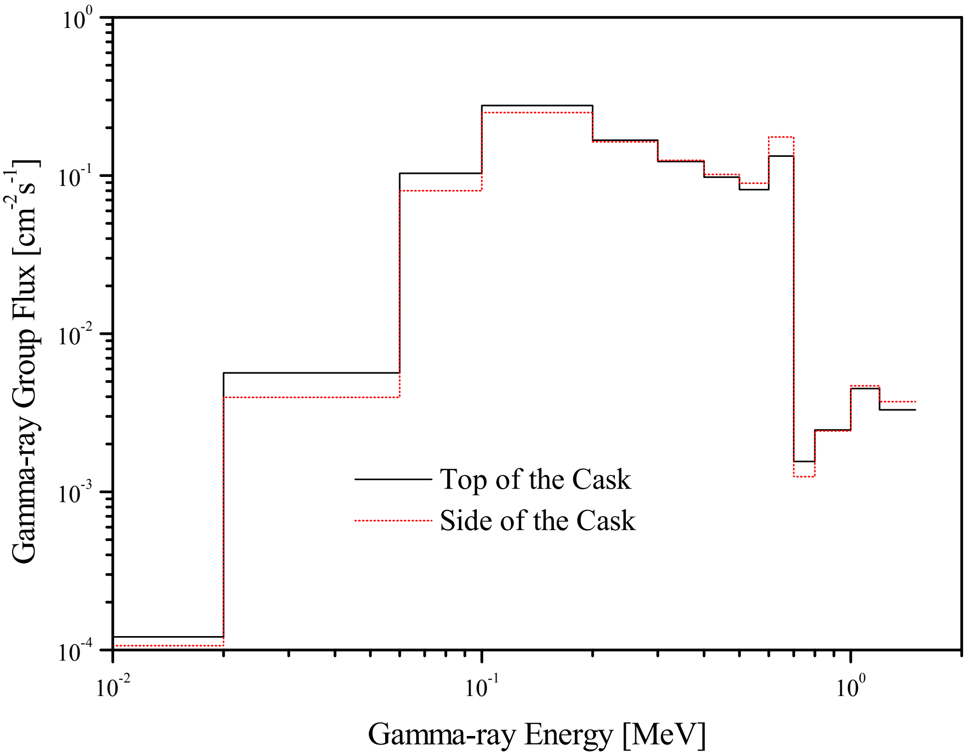

Fig. 4. Calcualted gamma-ray spectrum at the cask surfaces

Table 5. Measured and calculated values

Case	Source Activity Ref.	Ambient Dose Equivalent Rate [μ Sv/h]			
		Top surface		Side surface	
		Calculated	Measured	Calculated	Measured
Cube	[5]			7.3	8 -10
	[13] Avg			626.1	
	[6]			34.1	
Cask	[5]	2.7	50 - 60	10.7	80
	[13] Avg	10.1		240.5	
	[6]	13.5		55.9	

As can be seen form Table 5, calculation results, compared to measured ones, show wide dispersion due to large discrepancies in measured activities and unknown exact material composition of the sludge and sludge/cement matrix. Best agreement is achieved for the 'cube' and activity determined in Ref. [5] and for the 'cask' and activity determined in Ref. [6]. For better agreement, more reliable composition of the '100% dry sludge' – water mixture and sludge/cement matrix should be determined obviously.

6. Conclusion

Values of calculated gamma-ray ambient dose equivalent rates are compared to the measured values at the same spots of the cask with conditioned sludge from the RA spent fuel pool. But, acceptable agreements were found too, in spite of wide spread of reported data for the gamma-rays source activity and relatively large uncertainties in compositions and fractions of materials used in the calculations carried out by applying developed 3D model of the waste storage cask in the MCNP code.

Acknowledgements

This research is supported by Ministry of Science, Technologies and Development of Republic Serbia through Project no. 1958/2002 "Transport of particles in fission and fusion systems".

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