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# CALCULATION OF THE HIGH-LEVEL WASTE MAXIMUM ACTIVITY OF THE CHORNOBYL NPP OPERATING ORIGIN DURING STORAGE IN KTZV-0.2 CONTAINERS

This paper decrypts the conceptual design of the cask's control radiation system of high-level waste (HLW). This system is intended for the solid radioactive waste processing plant at Chornobyl NPP and is capable of forming a passport for a batch of HLW, based on the measured surface dose rate (DR) for KTZV-0.2 protective container. DR for primary packaging and KTZV-02 container were calculated using Monte Carlo simulation code MCNP 6.2. The typical material compositions of the Chornobyl NPP radioactive waste, contaminated by <sup>137</sup>Cs and <sup>60</sup>Co, were considered, and the corresponding surface DR was calculated. The simulation results were used for the development of the HLW cask load criterion for the radiation loading control system. The paper shows that HLW load criterion could be safely increased from 80 to 280 mSv/h.

Keywords: radioactive waste, maximum activity, container, modeling, simulation, Monte Carlo method, MCNP.

### 1. Introduction

There are two main types of radioactive waste (RAW) at the Chornobyl Nuclear Power Plant (ChNPP) site. The first one is RAW generated during normal operation of NPP and the second one is RAW generated as a result of the accident at Unit 4 as well as during elimination of the accident consequences [1]. Based on localization criteria, the following classification is possible:

- 1. RAW accumulated and placed in existing storage facilities on the NPP site;
- 2. Operational RAW produced during operation of the remaining NPP units;
  - 3. RAW of the Shelter object;
  - 4. RAW formed during decommissioning;
- 5. Solid RAW formed during the "Ukryttya" object transformation into an ecologically safe system.

Liquid and solid RAW are also differentiated in the classification. There is also another option – classification according to activity: low-activity, medium-activity, and high-activity.

Currently, liquid RAW at Chornobyl is undergoing an immobilization process. During this process, RAW is condensed to the cement mixture density. The immobilized RAW is packed into 200-litre drums, loaded into a concrete containment container KT-0.2, and transferred to storage.

Solid radioactive waste (SRAW) is located in the operational storage facilities of the ChNPP. Their total volume is about 2500 m<sup>3</sup>. Due to the lack of an appropriate instrumental and methodological basis,

the radionuclide composition of SRAW has not been determined during the storage process. However, SRAW sampling and laboratory analysis of radionuclide specific activity were carried out during preparatory measures to characterize accumulated SRAW for disposal at the solid radioactive waste treatment plant (PPSRAW) [2].

The automated radiation monitoring system was tested during the preparations for the PPSRAW. As a result of the evaluation of the system's performance, the characterization of the RAW batch failed. At the same time, the RAW management procedure sets the acceptance criteria for RAW disposal in a specially equipped near-surface storage facility for solid radioactive waste (SENSFSRW) [3]. According to the procedure, each package (batch) sent for disposal in PPSRAW from ChNPP requires a passport with the indication of the specific activity of each of the controlled radionuclides (more than 20 radionuclides) and other characteristics, including the total activity of radionuclides in the batch.

This investigation is aimed to determine the maximum specific activity of SRAW in the primary packaging so that the maximum dose rate (DR) at a distance of 10 cm from the surface of the KTZV-0.2 shielding container is below the permissible level. This was achieved by calculating the coefficients of activity-to-dose conversion for the primary packaging (165 l drum) and the protective container KTZV-0.2. The calculations were performed using Monte Carlo transport code MCNP version 6.2 [4] with ENDF/B-VII.1 nuclear data library. Using available data on the physical properties and

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composition of SRAW materials, together with contamination levels of <sup>60</sup>Co and <sup>137</sup>Cs, we obtained appropriate activity values for the primary repository, satisfying the surface DR acceptance criterion of 2 mSv/h. The calculated conversion factors were then used to determine the surface dose of the loaded KTZV-0.2 container.

# 2. High-level waste (HLW) properties used for simulation

To calculate activity-to-dose conversion factors, we used data obtained from laboratory studies of samples from the Chornobyl site [2]. According to these studies, HLW can be divided into the following categories:

1. pressable SRAW;

- 2. unpressable SRAW;
- 3. combustible SRAW.

The cask loading often leads to a significant degree of porosity in the stored SRAW. We have accounted for this effect in our modeling by varying the density of the SRAW according to the expected porosity value. We considered values of porosity up to 50 %, where the value of porosity was defined as the ratio of the volume of voids to the total volume of the barrel.

The following materials were considered as pressable SRAW: shell rock, fiberglass, copper wire, aluminum products, and fragments of building materials (gypsum). Table 1 shows the elemental composition, density, and relative weight of such SRAW determined according to the data of the studies [5].

Table 1. Elements	l composition of	f pressable SRAW
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	3.6 6 2			3.6 6	
Element	Mass fraction		Element	Mass fraction	
Shell rock (density 2.8 g/cm <sup>3</sup> )			Copper wire (density 8.96 g/cm <sup>3</sup> )		
C	0.120		Cu	1.000	
0	0.480		Aluminum products (density 3.97 g/g		
Ca	0.400		О	0.470749	
Fiberglass (density 2.49 g/cm <sup>3</sup> )			Al	0.529251	
В	B 0.018579		Gypsum (density 2.32 g/cm <sup>3</sup> )		
0	0.478631		Н	0.023416	
Na	0.059171		О	0.557572	
Mg	0.018037		S	0.186215	
Al	0.021107		Ca	0.232797	
Si	0.302924		_	_	

For the modeling we assumed that the pressed SRAW, loaded in a 165 l cask is a mixture of the materials from Table 1. The corresponding volumetric composition of the SRAW, used in the simulation is given in Table 2.

Table 2. SRAW volumetric composition used for simulations

Material	Volume fraction
Shell rock	0.50
Gypsum	0.30
Fiberglass	0.10
Aluminum products	0.05
Copper wire	0.05

Rubber, plastic, and wood with densities of 0.92, 0.93, and 0.64 g/cm<sup>3</sup> respectively were considered as combustive SRAW. The corresponding element compositions were taken according to the document [5]. In the modeling we assumed that the combustible SRAW consist of equal volumes of rubber, plastic, and wood. The calculated density of such SRAW was 0.830 g/cm<sup>3</sup>.

The unpressurised SRAW was a composite of metal and concrete with a density of 2.18 g/cm<sup>3</sup>. The corresponding elemental composition was taken according to the document [5].

# 3. Technical characteristics of the KTZV-0.2 container

KTZV-0.2 container is intended for the transporttation and storage of HLW. Its construction consists of two main elements: the primary packaging – a cylindrical cask made of stainless steel with a capacity of 165 l for RAW storage, and KTZV-0.2 protective container. KTZV-0.2 protective container (Fig. 1) has a cylindrical shape and consists of three protective layers: the outer shell of steel, concrete housing, and steel inner shell.

The following building materials were used for the modeling: stainless steel with a density of 7.8 g/cm<sup>3</sup> and concrete with a density of 4.23 g/cm<sup>3</sup> with an element composition according to document [5].

## 4. Modeling of primary packaging and container KTZV-0.2

The purpose of the computer modeling of the KTZV-0.2 container and the primary package was to calculate the activity-to-surface dose conversion factors for both the KTZV-0.2 container with the package inside and the package alone. The actual modeling was performed using the Monte Carlo transport code MCNP version 6.2 [4].

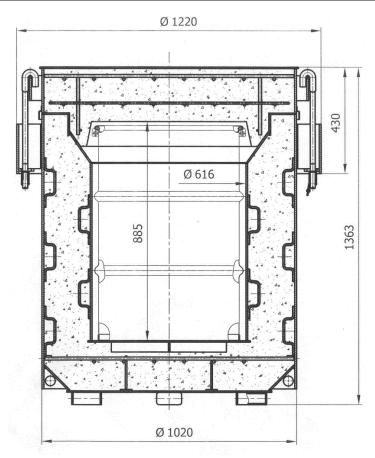
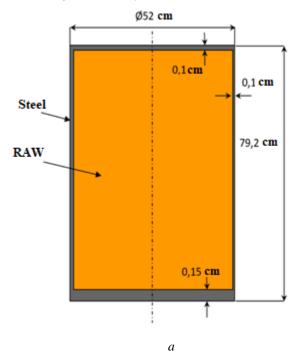
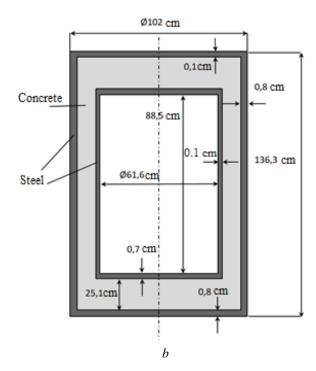


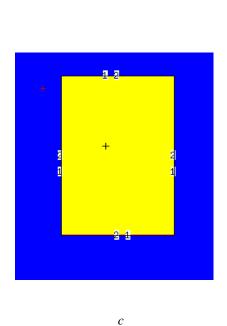
Fig. 1. Packing set with the KTZV-0.2 protective container.

The Monte Carlo simulations were based on the three-dimensional models of the KTZV-0.2 container and the primary packaging. Using the real container design (see Fig. 1), we developed the corresponding models within MCNP framework. The model for KTZV-0.2 was a cylindrical shell consisting of three layers: steel, concrete, and steel.

The primary packaging was modeled as a cylindrical stainless-steel shell and installed so that its centerline coincided with the centerline of the container. The dimensions of the modelled forms, together with the corresponding representations in the MCNP system, are shown in Fig. 2.







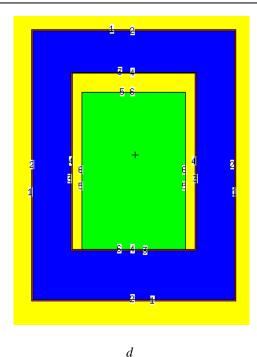


Fig. 2. Models of primary packaging and container KTZV-0.2 were used for simulation: a – geometric dimensions of the primary packaging; b – geometric dimensions of the container KTZV-0.2; c – the model of primary packaging for simulation in MCNP6 code; d – the model of the KTZV-0.2 container with primary packaging for simulation in MCNP6

The SRAW was modeled as a homogeneous cylindrical isotropic source that fills the inner volume of the primary package. According to the results of the studies [2] the radiation properties of SRAW are defined by the radiation of <sup>137</sup>Cs and <sup>60</sup>Co isotopes. Such a source was simulated by emission of 661.66 keV photons with the probability of 0.85 per decay in the case of <sup>137</sup>Cs and emission of two photons with energies of 1.17323 MeV and 1.33249 MeV per decay in the case of <sup>60</sup>Co. In each case, 10<sup>7</sup> initial photons were taken for calculations.

code.

During the modeling the DR was calculated at two points:

- 1. at a distance of 10 cm from the middle of the side surface of the package or container;
- 2. at a distance of 10 cm up along the centerline from the top surface.

The side surface and top surface DR were calculated using F5 tally, which corresponds to ring and point detectors. In this way, the calculated values are statistically valid. Possible DR were estimated using ICRP-116 dose conversion factors for AP (anteroposterior) geometry [6]. The corresponding coefficients are presented in Table 3.

Table 3. Dose conversion factors according to ICRP-116 for AP geometry

Energy, MeV	Conversion rate, pSv·cm <sup>2</sup>		Energy, MeV	Conversion rate, pSv·cm <sup>2</sup>	
0.01	0.0685		0.6	2.91	
0.015	0.156		0.662	3.17	
0.02	0.225		0.8	3.73	
0.03	0.313		1	4.49	
0.04	0.351		1.117	4.9	
0.05	0.37		1.33	5.59	
0.06	0.39		1.5	6.12	
0.07	0.413		2	7.48	
0.08	0.444		3	9.75	
0.1	0.519		4	11.7	
0.15	0.748		5	13.4	
0.2	1		6	15	
0.3	1.51		6.129	15.1	
0.4	2		8	17.8	
0.5	2.47		10	20.5	
0.511	2.52		_	_	

#### 5. Results and discussion

The results of calculations of activity-to-dose conversion factors for the KTZV-0.2 container are

shown in Table 4. In all cases, the statistical uncertainty of the obtained dose coefficients was less than 5 % in accordance with the recommendations of the MCNP guidelines.

Table 4. Radiation characteristics of the KTZV-0.2 container

Porosity	sity Density, g/cm <sup>3</sup> Dose coefficient		Activity for DR 2 mSv/h,	
rolosity	Delisity, g/cili	Side, mSv/h/TBq	Top, mSv/h/TBq	TBq
		Pressable SRAW,	<sup>137</sup> Cs source	
0	2.992	0.128	0.085	15.610
0.1	2.692	0.139	0.091	14.404
0.2	2.393	0.156	0.102	12.822
0.3	2.094	0.179	0.122	11.178
0.4	1.795	0.209	0.134	9.552
0.5	1.496	0.243	0.160	8.220
		Pressable SRAW,	<sup>60</sup> Co source	
0	2.992	2.955	2.028	0.677
0.1	2.692	3.266	2.177	0.612
0.2	2.393	3.637	2.446	0.550
0.3	2.094	4.113	2.713	0.486
0.4	1.795	4.678	3.077	0.428
0.5	1.496	5.482	3.582	0.365
		Combustible SRAW	V, <sup>137</sup> Cs source	
0	0.830	0.359	0.231	5.573
0.1	0.747	0.389	0.253	5.137
0.2	0.664	0.426	0.262	4.695
0.3	0.581	0.462	0.284	4.329
0.4	0.498	0.506	0.310	3.949
0.5	0.415	0.556	0.349	3.598
		Combustible SRAV	V, <sup>60</sup> Co source	
0	0.830	7.667	4.999	0.261
0.1	0.747	8.209	5.275	0.244
0.2	0.664	8.799	5.648	0.227
0.3	0.581	9.432	6.100	0.212
0.4	0.498	10.137	6.665	0.197
0.5	0.415	10.932	7.248	0.183
		Unpressable SRAW	V, <sup>137</sup> Cs source	
0	2.180	0.168	0.119	11.875
0.1	1.962	0.192	0.124	10.435
0.2	1.744	0.212	0.139	9.439
0.3	1.526	0.239	0.158	8.385
0.4	1.308	0.275	0.177	7.267
0.5	1.090	0.323	0.198	6.187
		Unpressable SRAV	V, <sup>60</sup> Co source	
0	2.180	3.948	2.606	0.507
0.1	1.962	4.312	2.892	0.464
0.2	1.744	4.796	3.122	0.417
0.3	1.526	5.369	3.513	0.373
0.4	1.308	6.081	3.889	0.329
0.5	1.090	6.898	4.448	0.290

The analysis of the results leads to the following conclusions:

- 1. Dose conversion factors increase with increasing porosity levels. This is a consequence of decreasing density and, hence, decreasing self-absorption of radiation in the bulk of the primary packaging.
- 2. The higher decay activity of the isotope <sup>60</sup>Co imposes more significant limitations on the permissible activity of the SRAW charge in the primary package.
- 3. The dose conversion coefficients for the side surface are higher than those for the top surface. This

is a result of the geometry of the cask, which favours greater self-absorption in the axial direction. Thus, the dose coefficients for the side surface should be used as the DR limit criteria at the surface of the KTZV-0.2 container. The DR limit is 2 mSv/h at a distance of 10 cm from the container, and the corresponding SRAW activity values calculated using the dose conversion factors are given in the last column of Table 4.

For the primary packaging (165 l cask) the DR near the surface could be calculated using appropriate

activity-to-dose conversion factors. These factors were calculated by Monte Carlo simulation of the primary package loaded with SRAW with no containment canister. The corresponding values of dose coefficients are presented in Table 5. The last two columns of Table 5 contain DR, calculated for the corresponding activity values from Table 4. For instance, the DR of 1751.38 (side) and 2122.22 mSv/h (top) for <sup>137</sup>Cs contaminated pressed SRAW with zero porosity correspond to the activity of 15.610 TBq from Table 4, obtained for 2 mSv/h DR near the side surface of KTZV-0.2 container.

Table 5. Radiation characteristics	of primary packaging (	(cask 165 l)
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	Dose coefficient							
Porosity	Density, g/cm <sup>3</sup>	DR (side),	DR (top),	DR (side), mSv/h	DR (top), mSv/h			
		mSv/h/TBq	mSv/h/TBq					
	Pressable SRAW, <sup>137</sup> Cs source							
0	2.992	110.17	135.25	1751.38	2122.22			
0.5	1.496	202.83	224.51	1667.30	1845.50			
	Pressable SRAW, <sup>60</sup> Co source							
0	2.992	537.60	629.73	363.88	426.25			
0.5	1.496	945.19	993.56	344.84	362.49			
		Combustible SRA	W, <sup>137</sup> Cs source					
0	0.83	293.87	301.89	1637.61	1682.33			
0.5	0.415	392.11	380.22	1410.98	1368.22			
		Combustible SRA	W, 60Co source					
0	0.83	1272.76	1262.15	331.99	329.22			
0.5	0.415	1620.38	1552.06	296.45	283.95			
Unpressable SRAW, <sup>137</sup> Cs source								
0	2.18	150.34	177.21	1785.29	2104.37			
0.5	1.09	259.90	273.41	1608.09	1691.69			
Unpressable SRAW, <sup>60</sup> Co source								
0	2.18	712.40	796.03	360.86	403.23			
0.5	1.09	1159.29	1169.55	336.10	339.08			

Document [7] imposes a 4.32 TBq limit on the maximum HLW activity value for the primary packaging and a 180 kg limit on its weight. At the same time, according to the document [8], the criterion for surfactant loading into a drum is 80 mSv/h.

Our calculations have shown that even within the most conservative approach it is possible to increase the HLW loading control level to 280 mSv/h. This value was derived from the lowest calculated DR for the primary packaging (see Table 5), accounting for the 5 % error in dose measurement by the monitoring system.

The calculated dose conversion coefficients provide the basis for an efficient algorithm for determining the limit criterion for HLW cask loading. The developed algorithm consists of the following steps:

- 1. Determine  $^{137}$ Cs  $(x_1)$  and  $^{60}$ Co  $(x_2)$  fractions in the total activity of HLW.
- 2. Calculate the maximum possible activity of  $^{137}$ Cs (A<sub>1</sub>) and  $^{60}$ Co (A<sub>2</sub>) in the cask with HLW before conditioning, assuming a 30 % error of the radiation

monitoring system, as follows:

$$A_1 = 4.32 \cdot x_1/130 \text{ TBq},$$
  
 $A_2 = 4.32 \cdot x_2/130 \text{ TBq}.$  (1)

- 3. Choose SRAW type (pressable, combustible, unpressable) and the porosity level.
- 4. Take dose conversion factors for  $^{137}$ Cs ( $K_1$ ) and  $^{60}$ Co ( $K_2$ ) from the third column of Table 5 and calculate the limiting DR:

$$DR_{max} = (K_1A_1 + K_2A_2)/1.05 \text{ mSv/h}.$$
 (2)

The resulting  $DR_{\text{max}}$  value is the limit criterion for the cask loading of HLW.

#### 6. Conclusions

This work considers the sci-tech problem of operating an automated radiation process control system for SRAW processing plants. Namely, the methodology for determining the maximum

permissible HLW loading without exceeding the DR limit for both the primary packaging and the protective container was developed. In order to calculate the activity to dose conversion factors for the primary packaging and the KTZV-0.2 container, a Monte Carlo simulation was performed using the MCNP 6.2 transport code. Three types of SRAW were considered during modeling: pressable, unpressable, and combustive. The possible partial loading of the primary packaging was described through the porosity parameter that varied from 0 to 50 %. Assuming that SRAW radiation properties are defined by <sup>137</sup>Cs and <sup>60</sup>Co radiation, we calculated dose conversion factors for various combinations of SRAW type, porosity, and gamma-emitting isotope.

Using the 2 mSv/h DR limits near the surface of the container established by the regulations, together with the calculated dose conversion factors, we obtained the maximum allowable activity stored inside the container. The corresponding set of dose conversion factors for the primary packaging gives the DR only for packaging which is subject to radiological control upon acceptance for storage.

Obtained dose conversion coefficients allowed us to develop the procedure for determining the limit criterion of HLW cask loading for the DFF system. This procedure could be implemented for solid waste processing at the ChNPP SRAW processing plant. This could significantly reduce the number of HLW containment containers required, which are currently localized in the storage facilities at the ChNPP.

#### REFERENCES

- Law of Ukraine "On the National Program for Decommissioning of the Chornobyl NPP and Transformation of the "Shelter" Facility into an Ecologically Safe System". Vidomosti Verkhovnoyi Rady Ukrayiny 24 (2009) art. 300. With changes introduced in accordance with Laws No. 3960-VI of 21.10.2011. Vidomosti Verkhovnoyi Rady Ukrayiny 23 (2012) art. 231; No. 2595-VIII of 16.10.2018. Vidomosti Verkhovnoyi Rady Ukrayiny 47 (2018) art. 376. (Ukr)
- Report of laboratory tests of SRAW samples from "light" compartments of the SSRAW building 85 PPSRAW of SSE ChNPP (eastern compartment). Approved 07/28/18. (Chornobyl, SSE ChNPP, 2018) 35 p. (Rus)
- Criteria for the acceptance of radioactive waste for disposal in a specially equipped near-surface storage facility for solid radioactive waste (SENSFSRW). The first stage of SENSFSRW operation (Chornobyl, 2009) 38 p. (Ukr)

- T. Goorley et al. Initial MCNP6 release overview. LANL Report LA-UR-11-07082 (Los Alamos National Laboratory, 2011).
- R.J. McConn (Jr.) et al. Compendium of Material Composition Data for Radiation Transport Modeling (PNNL-15870 Rev. 1) (USA, Richland, Washington: Pacific Northwest National Laboratory, 2011) 375 p.
- Conversion Coefficients for Radiological Protection Quantities for External Radiation Exposures. ICRP Publication 116. Annals of ICRP 40(2-5) (2010).
- 7. 58E-DPRAW. Operating instructions for the control and accounting system for SWRF and PPSRAW packages. [S. 1.] (Rus)
- 8. 25E-RSS Instruction on the operation of the SIT of radiation parameters of the system of radiation technological control of the industrial complex for the treatment of solid radioactive waste. [S. 1.] (Rus)

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### РОЗРАХУНОК МАКСИМАЛЬНОЇ АКТИВНОСТІ ВИСОКОАКТИВНИХ ВІДХОДІВ ЕКСПЛУАТАЦІЙНОГО ПОХОДЖЕННЯ ЧАЕС ПРИ ЗБЕРІГАННІ В КОНТЕЙНЕРАХ КТЗВ-0,2

Розроблено та запропоновано підхід для вирішення науково-технічної проблеми функціонування автоматизованої системи радіаційно-технологічного контролю на заводі із переробки твердих радіоактивних відходів Чорнобильської АЕС, при формуванні паспорта на партію високоактивних відходів (ВАВ) та визначенню максимально допустимої питомої активності з умовою не перевищення допустимої потужності дози (ПД) на поверхні захисного контейнера КТЗВ-0,2. Розроблений підхід базується на розв'язанні оберненої задачі методом Монте-Карло за допомогою програмного коду МСNР 6.2. Виконано розрахунок максимальної активності ВАВ та обрано сценарій визначення критерію завантаження твердих радіоактивних відходів у контейнер КТЗВ-0,2. Проведено моделювання та аналіз первинної упаковки та контейнера КТЗВ-0,2 та на його основі визначено фактори, що впливають на питому активність ВАВ та значення ПД від контейнера. За результатами моделювання програмним кодом МСNР будо розроблено алгоритм встановлення критерію завантаження бочки з ВАВ на системі радіаційного контролю завантаження, що дало змогу підвищити цей критерій з рівня 80 до 280 мЗв/год.

*Ключові слова*: радіоактивні відходи, максимальна активність, контейнер, моделювання, метод Монте-Карло, MCNP.

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