NUMERICAL CALCULATION OF ABSORBED DOSE RECEIVED BY GLASS MATRICES FOR IMMOBILIZATION NUCLEAR WASTE AND ITS SIMULATED IRRADIATION

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Background and Goal of the present work

The state of liquid radioactive waste (LRW) management at Ukrainian nuclear power plants (NPPs) is characterized by the lack of a completed technological cycle from processing to obtaining the final product suitable for further long-term storage or disposal. Resources for the storage are close to exhaustion.

Therefore, the development of technologies and materials for LRW solidification is an urgent need and aims to ensure the processing of LRW to a solid state that will meet the acceptance criteria for disposal in centralized storage facilities.

One of the effective methods of LRW solidification is their vitrification. The main advantage of vitrification is that during the vitrification process the volume of waste is reduced by several times and this saves expensive storage space.

The goal of this work is to calculate how much absorbed dose that borosilicate glass matrices with included bottom residue (BR) will accumulate over 300 years of storage, and to study the effect of simulated γ-irradiation on their physical and mechanical properties.

Initial data

The software toolkit GEANT4 was used for the caluclation. The 2x2x2 cm cube with radioactie impuritites was modelled during simulation. The chemical composition of the cube was the following (% mass): B_2O_2 - 11.49, Na_2O - 27.64, MgO - 0.43, Al_2O_3 - 0.98, SiO_2 - 45.67, P_2O_5 - 0.072, SO_2 - 0.45, Cl_2O_3 - 0.21, K_2O - 3.55, CaO - 0.94, TiO_2 - 0.037, Fe_2O_3 - 0.36, Cu_2O - 0.036, Cs_2O - 0.58, PbO - 7.55. As radioactive sources, spheres of with a diameter of 0.5 mm, located inside the cube and with the following coordinates (mm): 1st source (^{137}Cs) - {0, 2, 2}, 2nd source (^{60}Co) - {0, 0, 0}, 3rd source (^{134}Cs) - {0, -2, -2}. Initial specific activity and other initial data are given in Table 1. The half-lives and decay constants are given in Table. 2. The number of simulated decays of radioactive elements is 10^7. Volume of

Table 1: Initial data

Parameters	Value
Specific activity of isotopes:	
^{137}Cs, $\cdot 10^{-6}$ Ki/l	52
^{134}Cs, $\cdot 10^{-6}$ Ki/l	28
^{60}Co, $\cdot 10^{-6}$ Ki/l	6,1
Period of simulation, years	300
Dimensions, cm x cm x cm	2 x 2 x 2

Table 2: The half-lives and decay constants ^{137}Cs, ^{134}Cs and ^{60}Co.

Isotope	Half-lives, days	Decay constant, λ
^{137}Cs	11 000 ±90	0,0000630
^{134}Cs	754 ±0,7	0,0009197
^{60}Co	1 925,3 ±0,4	0,0003601

Numerical calculation

Calculation method. The software toolkit GEANT4 was used for the calculation. Period of simulation was 300 years. High-altitude years were not taken into account. The recalculation of the activity I of radioactive elements was carried out for every six months according to the following formulae:

$$I = I_0 \cdot e^{(-\lambda \cdot t)}, \quad (1)$$

where, I_0 – initial activity, Ki; λ – decay constant; t – time elapsed since the initial moment of time, days.

Absorbed dose calculated according to the following formula

$$D = \frac{k \cdot d \cdot \Sigma I \cdot V}{n_0}, \quad (2)$$

where $k = 3,7 \cdot 10^7$ – number of decays per 1 second (corresponds to 1 Ci); d – calculated absorbed dose; ΣI – total activity of all radioactive sources at the time of calculation (taking into account radioactive decay), Ci/m^3; V – volume of the target being modeled, m^3; n_0 – the number of decays (10^7) that was modeled.

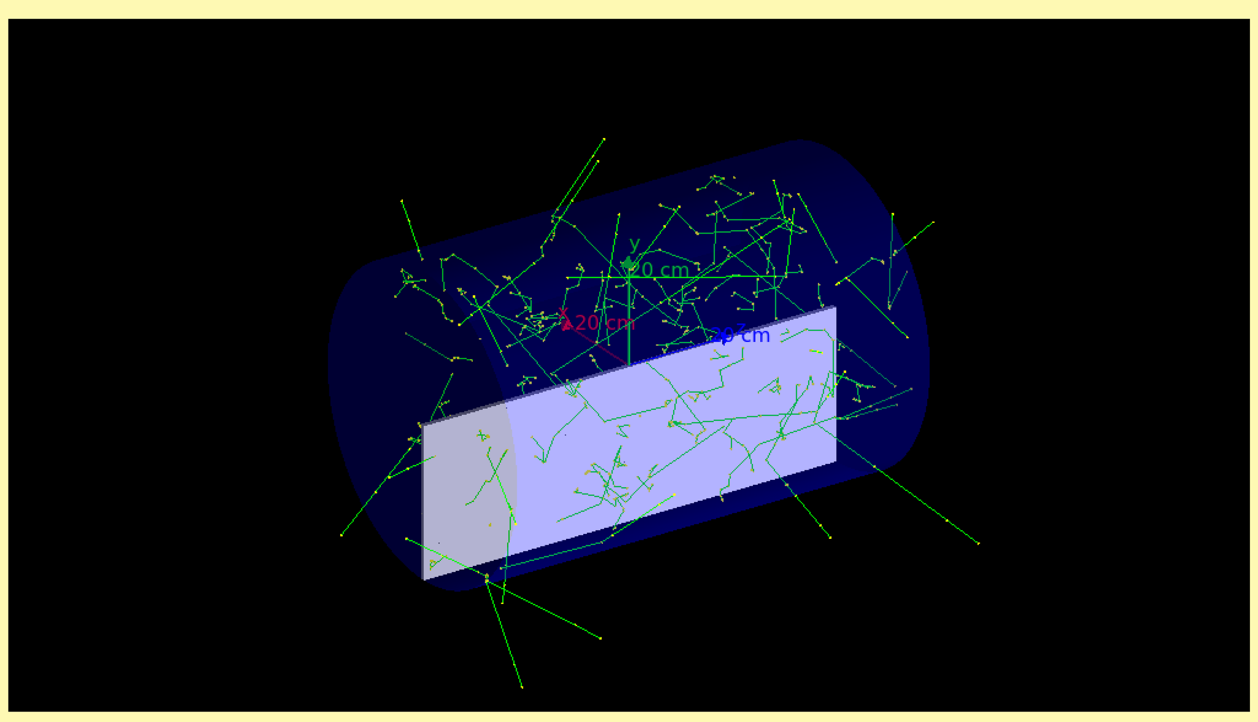

Fig. 1. Scheme of calculation setup.

Choosing of a toolkit for the simulation of the passage of particles through matter

GEANT4 [2] was chosen as a toolkit for the simulation of the radiation treatment because of

1. GEANT4 has the multithreading mode (LINUX version).
2. GEANT4 uses modern programming language C++.
3. This toolkit is free
4. A long-time development
5. Good technical support
6. GEANT4 has possibility to input complex geometry of irradiated objects

References

[1] Poole, C. M., Cornelius, I., Trapp, J. V., & Langton, C. M. (2012). A CAD interface for GEANT4. Australasian physical & engineering sciences in medicine, 35(3), 329-334.

[2] Agostinelli, Sea, et al. "GEANT4—a simulation toolkit." Nuclear instruments and methods in physics research section A: Accelerators, Spectrometers, Detectors and Associated Equipment 506.3 (2003): 250-303.

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Result of calculations

The results of calculations for 300 years of long-term storage of nuclear waste are given on Fig. 1. The initial data are given in the Tables 1 and 2. The absorbed dose in cylinder (height - 0.8 and radius - 0.28 m) is equal to 627,9 Gy.

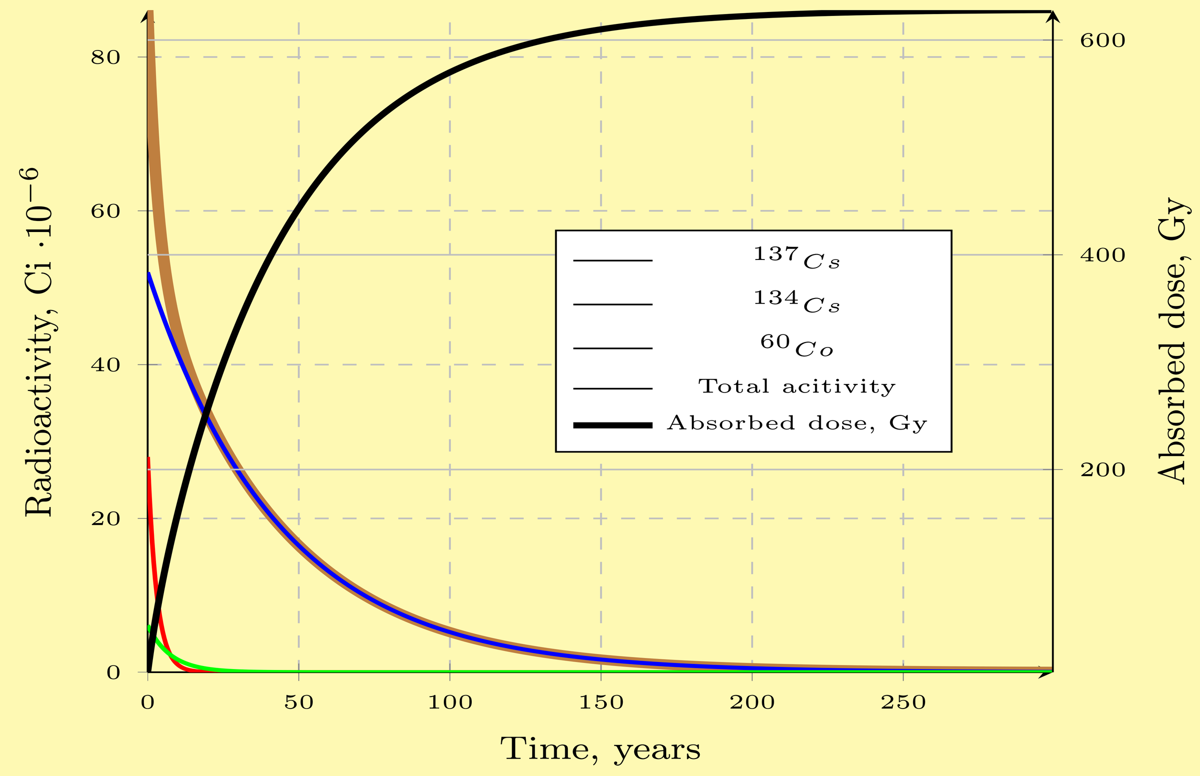


Рис. 1. Radioactivities of ^{137}Cs, ^{134}Cs, ^{60}Co, total radioactivity, absorbed dose vs time.

Simulated irradiation

Simulated irradiation of glass samples S45-Pb10 and SK45-CF10 was carried out at electron beam accelerator LU-10 (NSC KIPT) by X-ray with average energy 10.4 MeV with dose rate equal to 1.09 kGy/h. Sample appearance before irradiation and after irraidation is shown on Fig. 2.

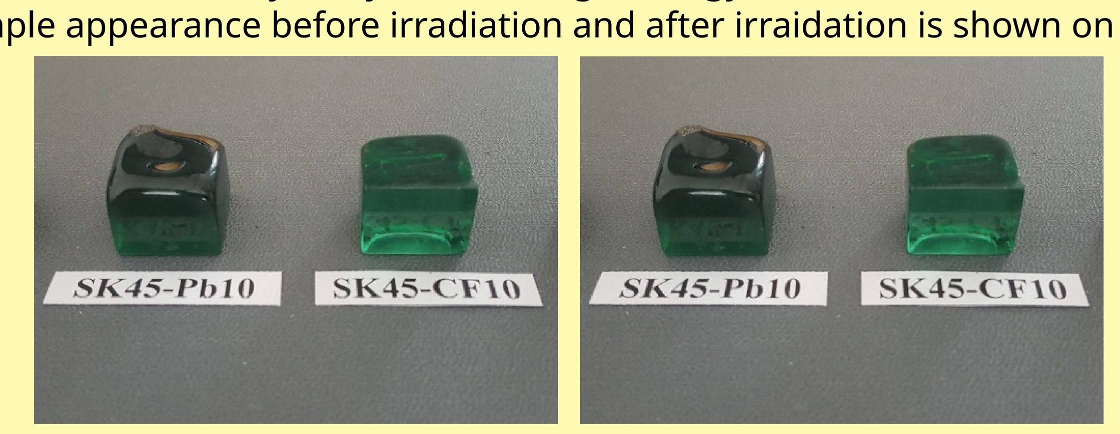
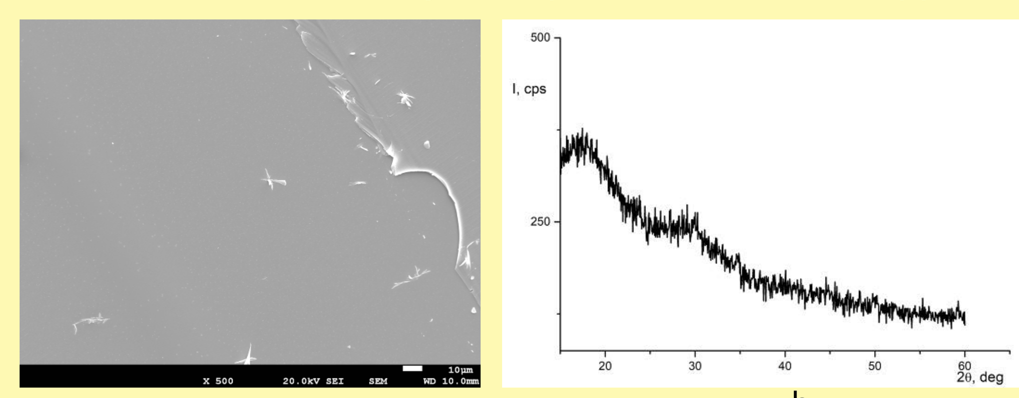


Fig. 2. Samples appearance before (left) and after (right) irradiation 10 kGy.

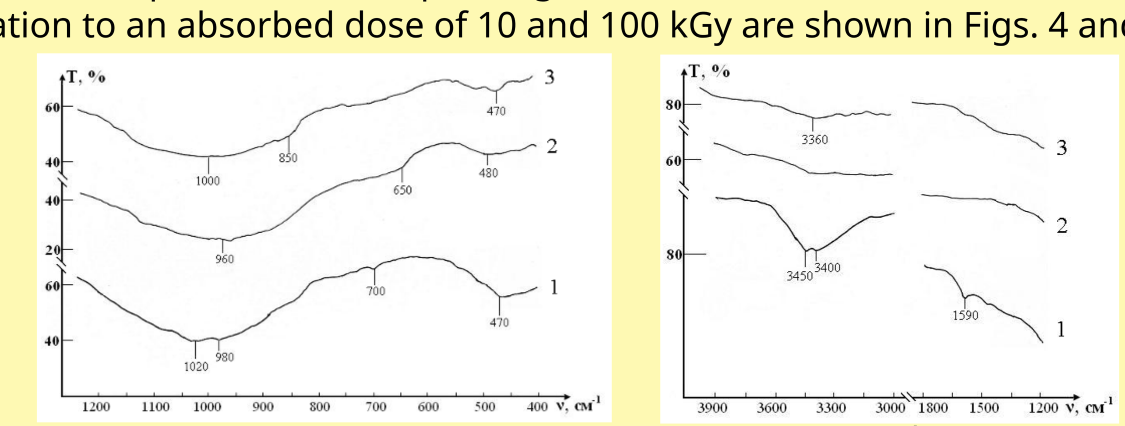
There are no signs of destruction and the appearance of cracks and other defects on the samples after irradiation. Also, no changes were observed in the structure (Fig. 3a) and phase composition of the SK45-Pb10 glass matrix samples after X-irradiation up to an absorbed dose of 100 kGy (Fig. 3b). The glass matrix material remains X-ray amorphous without the appearance of crystalline phases.



a b

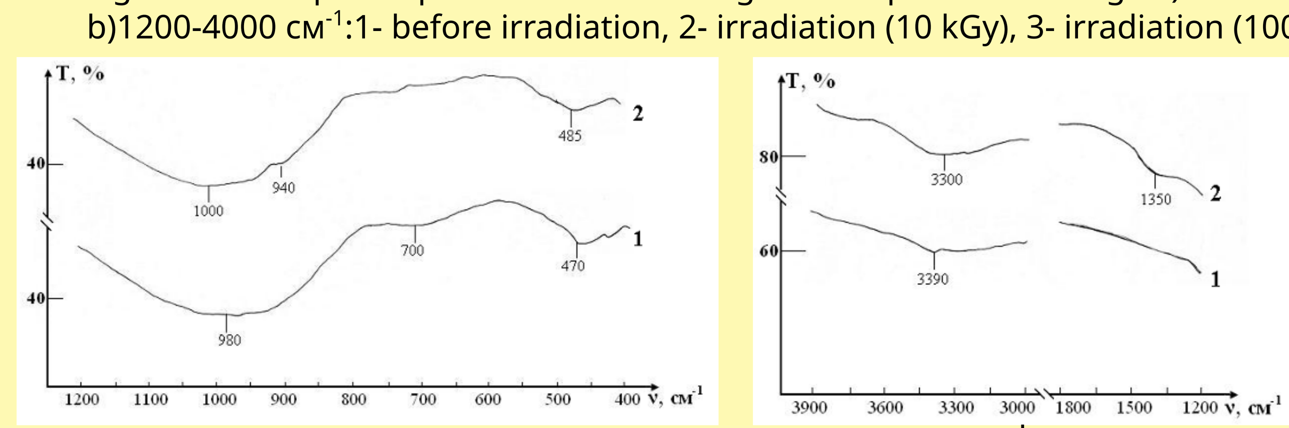
Fig. 3. Structure and phase composition of SK45-Pb10 glass matrix samples after X-ray irradiation: a – SEM image, b – diffraction pattern

Small differences were found in the IR spectra of glass matrix samples before and after X-ray irradiation. The IR spectra of a sample of glass matrices SK45-Pb10 and SK45-CF10 before and after irradiation to an absorbed dose of 10 and 100 kGy are shown in Figs. 4 and 5.



a b

Fig. 4. IR absorption spectra of SK45-Pb10 glass samples in the range a) 400-1200 cm^{-1} b)1200-4000 $cм^{-1}$:1- before irradiation, 2- irradiation (10 kGy), 3- irradiation (100 kGy)



a b

Fig. 5. IR absorption spectra of SK45-CF10 glass samples in the range a) 400-1200 cm^{-1}: b)1200-4000 $cм^{-1}$:1- before irradiation, 2- irradiation (10 kGy), 3- irradiation (100 kGy)

Conclusion

The numerical calculation of absorbed dose received by glass matrices for immobilization nuclear waste were peroformed. Calculated absorbed dose for 300 years of disposal is equal to 627,9 Gy. **The simulated X-ray irradiations of glass samples** (SK45-Pb10 and SK45-CF10) were carried out on EB accelerator with the average energy 10,4 MeV up to doses 10 and 100 kGy with dose rate equal to1.09 kGy/h. Scanning electron microscopy, X-ray crystallography, infrared spectroscopy show small differences between irradiated and not-irradiated samples. However, the detected minor differences do not affect the composition, structure and chemical bonds of the obtained glass matrices, and, accordingly, their insulating properties. Thus, the SK45-Pb10 and SK45-CF10 glass matrices have sufficient radiation resistance, which can ensure its integrity and stability during long-term storage.