RADIATION CHARACTERISTICS OF BOROSILICATE GLASS CONTAINING HIGH-LEVEL WASTE

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The article presents computational and analytical studies dealing with radiation characteristics of liquid high-level waste vitrified using borosilicate glass (BSS). To confirm the quality of glass on a time scale of up to 10^4 years and more, computational and experimental studies are performed to identify the dose loads on the BSS from all types of radiation. The paper presents the results of computational studies focused on radionuclide composition and radiation characteristics of a BSS produced during the reprocessing of spent nuclear fuel from VVER-1000 reactors based on a method developed by JSC Radium Institute named after V.G. Khlopin.

Keywords: *radioactive waste, borosilicate glass, spent nuclear fuel, mathematical models, energy release, radiation damage.*

Introduction

Vitrified high-level liquid waste contains radioactive isotopes the decay of which results in the irradiation of the matrix potentially causing its structural transformations. The effect of such transformations on the basic glass properties should be studied and contribute to the long-term stability assessment of glass matrices. Scientific and technical literature sources from abroad provide ample information on this matter, which basically confirms high radiation stability of the glass structure formed by strong ionic-covalent bonding via oxygen bridges.

Beta decay of fission products, primarily ^{137}Cs and ^{90}Sr and alpha decay of actinide elements (uranium, neptunium, plutonium, americium and curium) are considered as the main sources of ionizing radiation in vitrified HLW affecting the radiation and chemical stability of the glass matrix [1]. In the long-term, internal self-irradiation from the radionuclides contained in the RW matrix can affect its microstructural transformations, phase stability, thermodynamic properties and, consequently, the system performance in terms of HLW isolation from the biosphere. Beta decay of nuclides results in the release of particles (with energy level of about 0.5 MeV), low-energy recoil nuclei and gamma radiation. Alpha decay leads to the formation of particles with energy levels ranging from 4.5 to 5.5 MeV, energetic recoil nuclei with energy levels ranging from 70 to 100 keV, and some amount of gamma radiation. The released particles and gamma radiation interact with the electron shells of the atoms inside the matrix by transferring energy to electrons mainly through ionization processes producing electron-hole pairs. Alternatively, they can interact with the atomic nucleus via elastic collisions displacing them from their initial positions in the glass matrix. In addition, transmutation of recoil nuclei can occur with new nuclei, in turn, being subject to radioactive decay. According to various data, the effect associated with the beta decay of fission products prevails within the timeframe between 300 and 600 years [2, 3] causing high radioactivity and self-heating of glasses during their storage. The influence of alpha decay predominates in the subsequent years.

In Russia, alumophosphate glass (APG) is used for liquid HLW immobilization at RT-1 plant (FSUE PA Mayak) with large volumes of vitrified high-level RW (VHLW) from the reprocessing of spent nuclear fuel from WWER-440, BN-600, research reactors, transport and ship installations, etc. already immobilized enabling their further deep geological disposal [4].

Worldwide, borosilicate glasses have gained wider application due to a number of advantages as compared to APG, for example, BSS devitrification rate due to crystallization is lower than the one of APG. For this reason, during the development of MCC's pilot-demonstration center for SNF reprocessing (hereinafter, PDC), borosilicate glass was chosen as immobilization material for liquid HLW generated due to the reprocessing of SNF from VVER-1000 reactor units.

Taking into account the new glass matrix developed under this technology, computational and experimental studies of VHLW radiation characteristics should be performed based on the case study of borosilicate glass developed for the PDC [5].

Preliminary computational studies should become a basis for further planning and development of methodological approaches enabling the experimental work on the effects of beta, gamma and alpha irradiation of model BSS using accelerator technology and doping matrices with appropriate radionuclides.

Present research involved computational studies focused on the radionuclide composition and radiation characteristics of borosilicate glass resulted from SNF reprocessing according to the basic reprocessing method adopted at the PDC. Computational codes are used under such studies to simulate main radiation characteristics of spent nuclear fuel affecting the stages of its processing, HLW vitrification, as well as radiation and chemical parameters of the BSS under given conditions of fuel irradiation in the reactor taking into account different burnup ranges. This study aims to identify the radiation characteristics of BSS-based VHLW (activity, absorbed dose, energy release, yields and energy dependence of gamma and neutron sources) given various exposure times. Computer code TRACT was used to calculate the changes in fuel nuclide composition and radiation characteristics both during its irradiation and cooling [6–9]. Reference data presented in [10, 11] were partially used in the verification of SNF calculations.

Radiation characteristics of vitrified high-level waste

In accordance with the basic technology, the study focuses on HLW resulting from the reprocessing of SNF from a VVER-1000 reactor unit. The initial fuel is uranium dioxide UO_2 with ^{235}U enrichment of 4.35 %. Fuel composition was calculated in the approximation of a stationary reactor operation mode at a nominal power level: one fuel lifetime consisted of three micro-life-times lasting 305.9 days with 48-day off-load in between. SNF nuclide composition was calculated in the approximation of a uniform neutron flux distribution over the fuel assembly height corresponding to a fuel burnup of 50 GW·day/t U per life-time.

According to the PDC designs, the spent nuclear fuel is cooled down for 7 years and then reprocessed with uranium and plutonium isotopes being extracted. The generated radioactive waste can contain up to 0.01 % uranium and 0.025 % plutonium as compared to the initial SNF content. It should be noted that the basic SNF reprocessing technology applied at PDC incorporates some boundary parameters for fuel (in terms of fuel burnup and cooling time) the reprocessing of which will result in rather high BSS heat release and dose loads on the glass matrix. To date, ~6500 t of HM VVER-1000 SNF [12] with different burnups (from 8 to 50 GW·day/t U and more) and cooling times (from 10 to 30 and more years) have been accumulated in MCC storage facilities requiring reprocessing. A study [13] showed that accumulated and "fresh" SNF can be reprocessed jointly for ten or more years at PDC to enable multi-fold reduction in the BSS heat release. Therefore, such reprocessing scenario will result in much lower dose loads on the glass.

According to the initial data, the basic SNF reprocessing technology suggests that liquid HLW contains both fission products and actinides, as well as corrosion products of stainless steel (Fe, Cr, Ni) and sodium released into the stream as a result of extractant washing and regeneration. Availability of significant sodium amounts in liquid HLW is taken into account during the development of glass frit composition. Taking into account the specifics of such HLW with stable components in the waste, the total calculated inclusion for all oxides of the elements will account for some 20 wt%. According to calculations, such a high waste inclusion allows to produce some 100—110 dm^3 of high-level glass with a density of 2.76 g/cm^3 from 1 tone of reprocessed SNF.

Table 1 presents the estimated composition of vitrified RW Class 1 produced using the borosilicate glass developed by V. G. Khlopin Radium Institute [5].

Table 1. Calculated chemical composition of borosilicate RW matrix

Component	Content in the glass, %
SiO_2	45.60±10
Na_2O	13.35±10
B_2O_3	14.40±10
Li_2O	2.80±10
CaO	2.40±10
Al_2O_3	2.40±10
MnO_2	2.40±10
Fe_2O_3	0.25±10
NiO	0.12±10
Cr_2O_3	0.72±10
Oxides of radioactive elements	15.56±10

Changes in the activity (Figure 1) and residual energy release (Figure 2) were calculated for the given composition of vitrified RW. The data account for 1 m^3 of vitrified RW and cooling times of up to 10^4 years. Figure 2 presents the energy release for the three components: due to alpha and beta decay, as well as due to gamma radiation. During the cooling time period of 0 to ~ 100 years, it is beta and gamma radiation that mainly contributes to the energy release, whereas only in a long run accounting for over 120 years that alpha radiation becomes the main source of energy release.

SNF and RW neutrons result from spontaneous fission of actinides and reactions (α, n) on light nuclei. Figure 3 shows the yield of photons and neutrons for vitrified RW considering cooling times of up to 10^4 years. Neutron source intensity is several orders of magnitude less than the one of the gamma radiation source.

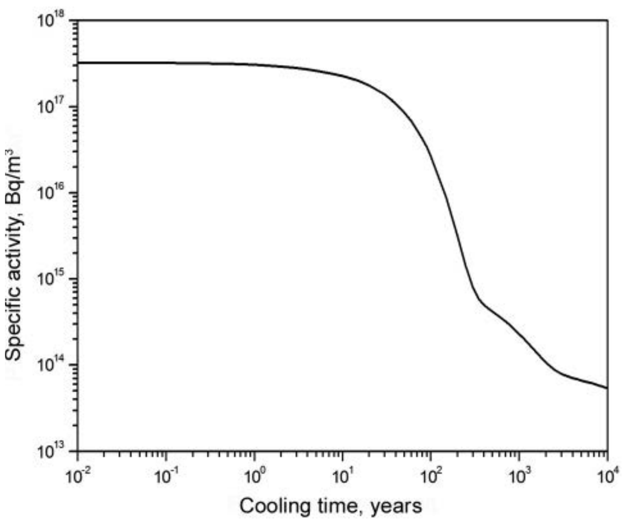

Figure 1. Specific activity of vitrified RW

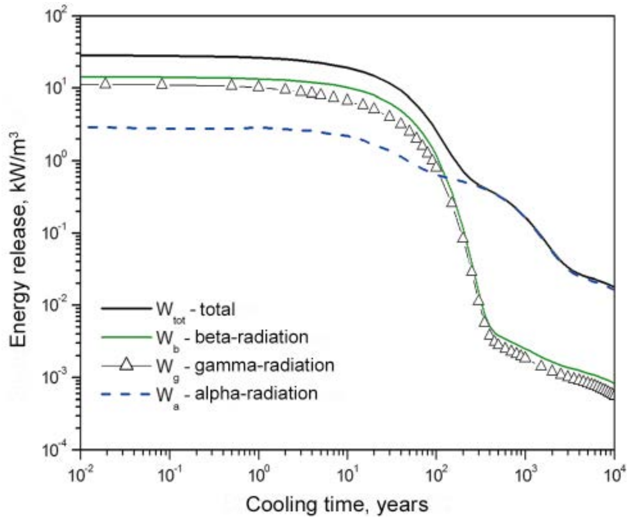

Figure 2. Energy release from vitrified RW

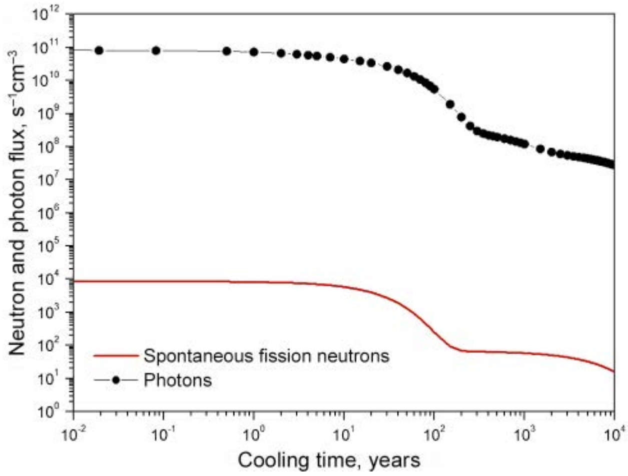

Figure 3. Cooling-time dependency of neutron and photon fluxes per BSS unit volume

Figure 4 shows the energy spectrum of a photon source given different cooling times. At the time of BSS production, the majority of photons was concentrated in the energy ranges of 0.6—1.0 MeV and 20—40 keV (solid line in Figure 4). With increasing cooling time, the photon spectrum shifts to the energy range from 10 to 100 keV. This should be taken into account in the assessment of energy release from a gamma-ray source considering leakage.

It should be noted that neutron radiation plays a minor role in the resulting radiation environment around RW compared to the one emitted by gamma quanta. However, compared to gamma quanta, fast neutrons are more dangerous for humans since they have a high penetrating capacity in many

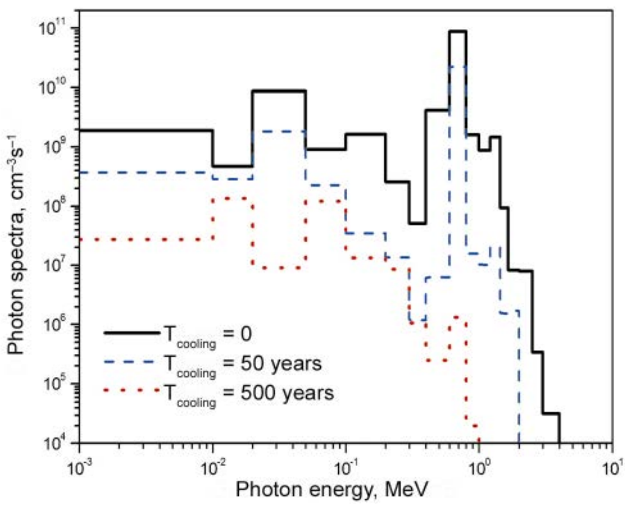

Figure 4. Energy spectrum of a bulk photon source considering different vitrified RW cooling times

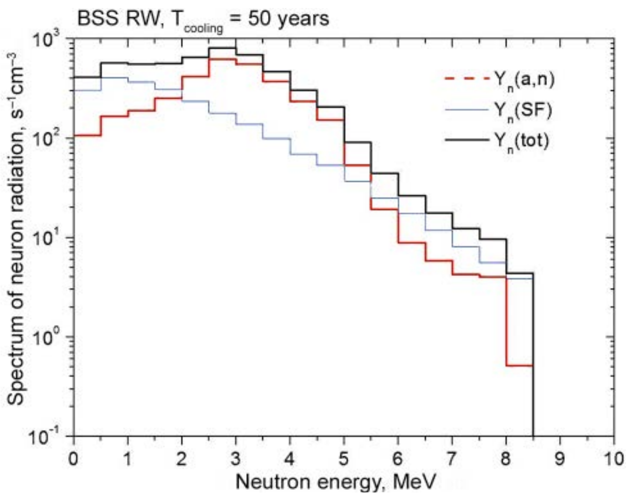

Figure 6. Energy spectrum of neutrons in vitrified RW and its constituent components due to spontaneous fission and (α, n) reaction given a cooling time $T_{cooling}$ = 50 years

substances. Increased fuel burn-up rate results in greater accumulation of americium and curium isotopes. For example, ^{244}Cm concentration increases proportionally to the depth of burnup to the power of four. Due to longer cooling time, short-lived fission products decay and actinides start to play a more important role in SNF and RW characteristics.

At cooling times of up to 100 years, curium isotopes (^{242}Cm, ^{244}Cm) provide major contribution to neutron radiation. In this case, the decisive contribution is the one associated with ^{244}Cm contributing to over 90 % of neutron radiation given a cooling time of 3 years. At longer cooling times of over 100 years longer-lived isotopes of minor actinides start to play increasingly important role.

Figure 5 shows the full neutron spectrum for vitrified RW for the initial period and its main components — the spectrum of spontaneous fission neutrons and the spectrum of neutrons due to (α, n) reaction. One can note that the number of reaction neutrons (α, n) in the neutron energy range of 2.5—5 MeV exceeds the number of spontaneous fission neutrons. Similar data are shown in Figures 6 and 7 for the full neutron spectrum of vitrified RW and its main components — spontaneous fission neutron spectra and spectra of neutrons due to (α, n) reaction given cooling times of 50 and 500 years, respectively.

Table 2 summarizes estimated specific neutron activities due to spontaneous fission processes and (α, n) reaction calculated for vitrified RW. It demonstrates that increased cooling time provides greater contribution of neutrons due to (α, n) reaction as compared to the one of spontaneous fission neutrons.

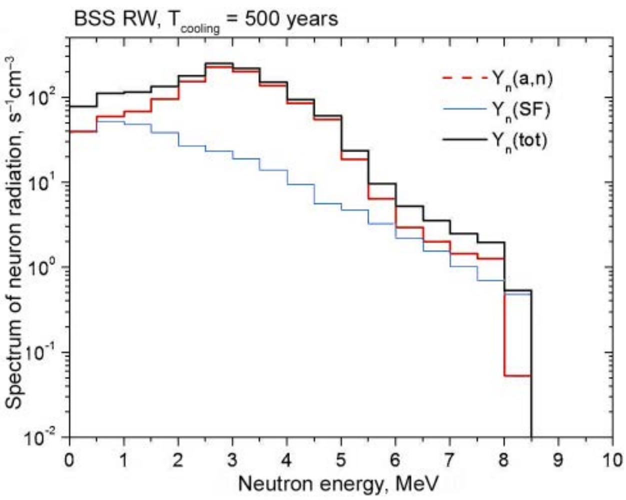

Figure: 7. Energy spectrum of neutrons in vitrified RW and its constituent components due to spontaneous fission and (α, n) reaction given a cooling time $T_{cooling}$ = 500 years

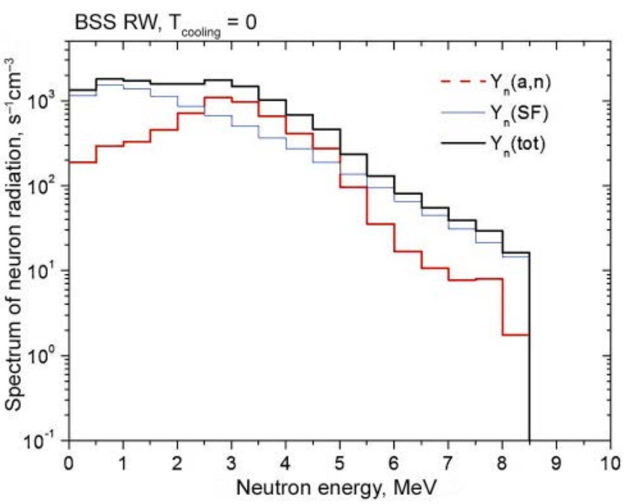

Figure 5. Energy spectrum of neutrons in vitrified RW and its constituent components due to spontaneous fission and (α, n) reaction given a cooling time $T_{cooling}$ = 0 years

Table 2. Neutron yields due to (α, n) reaction and spontaneous fission accounting for different vitrified RW cooling times

Cooling time, years	Relative neutron yields due to (α, n) reaction, %	Relative neutron yields due to spontaneous fission, %
0	39.7	60.3
10	39.6	60.4
20	42.7	57.3
50	58.3	41.7
100	79.7	20.3
500	79.9	20.1
1,000	66.6	33.4
10,000	76.8	23.2

Calculation of dose loads on borosilicate glass

The rate of absorbed dose buildup due to BSS internal irradiation due to neutron, alpha-, beta- and gamma radiation resulting from the decay of radionuclides contained in the glass can be calculated as the ratio of the energy absorbed in the sample to its mass: $P = W/m$ (P — Gy/s, W — J/s and m — kg).

It should be noted that the energies of alpha particles do not exceed 6.5 MeV and those of beta particles do not exceed 1 MeV. Given such energy levels, the maximum range of alpha and beta particles in the BSS will be no more than 30 μm and ~ 2 mm, respectively. Therefore, the approximation of point energy absorption sems to be quite applicable for alpha and beta particles. For neutrons and gamma quanta, processes associated with energy transfer in the BSS medium should be taken into account, i. e. take into account the release of neutrons and gamma quanta from the sample volume. This study focuses on an approximation assuming that gamma radiation with an average energy of less than 1 MeV is almost completely absorbed by BSS medium for a given size of a primary package: 100-liter capacity and a mass of about 280 kg. Figures 8 and 9 show the estimated absorbed dose rates and the integral accumulated dose depending on BSS cooling time.

Analysis of the data obtained shows:

- During the first ~ 200 years of RW matrix material cooling, the main dose effects are associated with beta and gamma radiation;
- after this period, it is the alpha radiation that mainly contributes to the dose rate, but the total integral dose changes only slightly. Figure 9 shows that the integral radiation dose for a period of 10^4 years has an order of $(3—5)\cdot10^9$ Gy. If the geometry of BSS can layout inside the storage facility is accounted for, contribution of gamma radiation can be estimated more accurately allowing to adjust the above estimates.

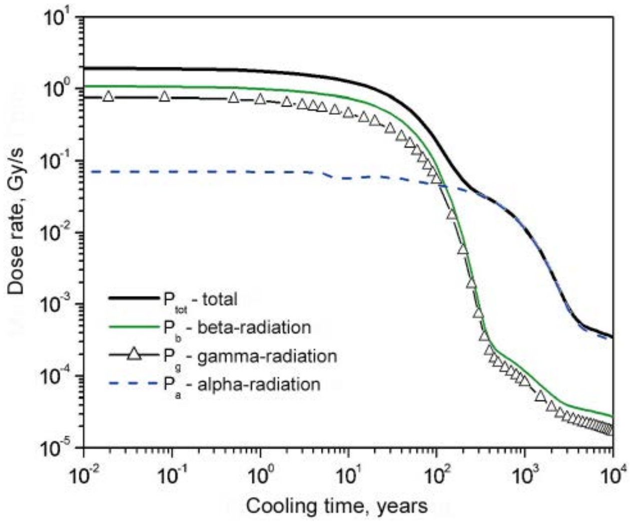

Figure 8. Dependence between the absorbed dose rate and BSS cooling time

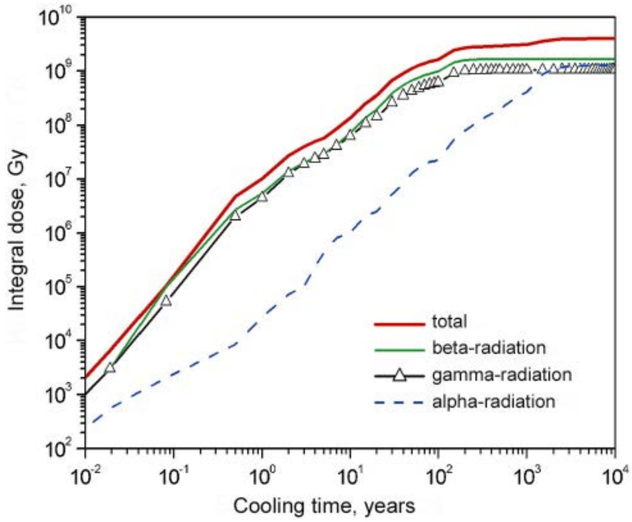

Figure 9. Dependence between cumulative radiation dose and BSS cooling time

Results provided in Table 2 and Figures 1—9 fully describe the alterations in BSS radiation characteristics and potential dose loads on the glass matrix considering a cooling period of up to 10^4 years. Resulting characteristics of neutron and photon radiation sources contained in vitrified RW can serve a basis for further analysis focused on the effects produced by various types of radiation on vitrified RW matrix properties.

Conclusion

The paper presents integral and differential dose loads estimated for borosilicate glass containing high-level waste considering a cooling time of up to 10^4 years. Energy spectra of neutron and photon radiation in vitrified RW have been estimated for various cooling times of up to 10^4 years.

During the experimental studies focused on BSS characteristics these data will be used to assess the expected effects.

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