

VITRIFICATION OF A SIMULATOR OF VAT RESIDUES FROM LIQUID RADIOACTIVE WASTE[†]

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The study on the possibility of the use of the optimal glass compositions for vitrification of an imitator of vat residues of liquid radioactive waste from nuclear power plants with VVER-1000 reactors was carried out. The main process parameters such as vitrification temperature, strength, corrosion resistance, absence of crystalline phases, minimization of glass-forming additives and inclusion the maximum amount of waste were analyzed. It has been established that the melting temperature of lead-borosilicate glass matrices was 1150 °C, which satisfies the requirements for vitrification of low- and medium-level waste. The ultimate compressive strength of the obtained samples of glass matrices was 136.0 MPa. In addition, it has been shown that lead-borosilicate glass matrices are the most resistant to leaching. The cesium leaching rate was $1.5 \cdot 10^{-5}$ g/cm²·day.

Keywords: liquid radioactive waste; vat residue; vitrification; strength, leaching rate

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INTRODUCTION

At present, the problem of environmental safety during the process of the radioactive waste immobilization is very challenging for further nuclear power industry development. It is well known that exploitation of nuclear power plants (NPPs) results in accumulation of liquid radioactive wastes (LRW) of low and middle activity. Ukrainian NPPs currently accumulate a large amount of LWR, represented by vat residues with a salt content of 200 to 600 g/l, salt melt with a higher salinity, spent filter materials and sludge. According to the rules for ensuring radiation safety, liquid radioactive waste cannot be transferred for long-term storage and disposal without additional processing. Various technologies for processing such types of waste to a state suitable for disposal are under consideration [1].

The most common and fairly cheap method of LRW conditioning is cementing [2, 3]. However, cement matrices are characterized by increased solubility in an aqueous medium, and the volume of final waste forms significantly increases [4]. Some authors argue that the most optimal method for solidifying LRW is inclusion in glass with subsequent burial in near-surface storage facilities [5, 6]. The prospect of using glass as a protective matrix is due to the high ability to include elements in its composition, regardless of their charge and atomic size, resistance to radiation damage, and proven production technology [7]. In addition, vitrification of waste reduces their volume by several times, and thus saves expensive storage space. The cost of disposal is very high, even in its simplest near-surface variant, which is used only for low- and medium-level short-lived nuclear waste. For example, in France, the cost of storing one cubic meter of low- and medium-level short-lived waste in a near-surface storage facility is about 2200 € [8].

The main problems of the vitrification method are associated with the use of high-temperature processes. In addition, the following difficulty arises during vitrification process: for radioactive waste with a variable composition, it is necessary to change the composition of the glass frit accordingly in order to obtain a sufficiently high-quality product. Since radioactive waste contains many different elements, their introduction into glass changes its parameters and affects its stability. Moreover, some components are practically not included in glass at their certain content in radioactive waste.

However, despite the above limitations, the vitrification method is considered as one of the promising methods for converting LRW into a solid state. The high stability of the glass, combined with a sufficiently large reduction in the volume of waste, encourages the use of LRW vitrification as an alternative to cementing. A significant advantage of vitrification is the fact that there are no significant restrictions on the concentration of salts in LRW, as well as on the content of organic materials and surfactants, compared with the use of cement.

Vitrification of radioactive waste, especially of high-level activity, has been carried out in several countries for almost 40 years. Teams of specialists from the United States, France, Russia, Great Britain, Belgium, and others are carrying out numerous works on the development and improvement of methods for the vitrification of radioactive waste [8–10]. However, research on the composition and long-term behavior of vitrified high and intermediate level wastes, mainly borosilicate compositions, is still ongoing. The main attention is paid to the composition, chemical and

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radiation resistance of glass matrices in order to ensure the safe conditioning of wastes of various compositions and demonstrate the safety of their disposal [11].

Thus, in [12], various glass and glass-ceramic materials are considered that are capable of immobilizing waste with a high load. The emphasis here is on the importance of the solubility of the waste components and the consequent undesirable crystallization compared to intentional crystallization of the glass-ceramic waste forms.

According to the authors of [13], glass is considered an acceptable form for curing low-level waste with a high boron content, generated at Korean nuclear power plants. The advantages of the process are ease of handling and unique ability to accept a wide variety of waste streams. The study involved development of glass structure and producing of several glass frits which were evaluated to determine their suitability for LRW vitrification. The fly ash generated in coal-fired power plants with the main components SiO_2 and Al_2O_3 , is a material necessary for the formation of glass mesh. For selected compositions using fly ash, detailed evaluations of the final product were carried out, including the amount of waste, uniformity, chemical resistance, viscosity, etc. Up to 30 wt. % of the waste was included in the composition of glass welded at 1200 °C using fly ash after adding 5–10 wt. % Na_2O .

Works have been carried out and are being carried out to develop glass compositions for the immobilization of LRW from nuclear power plants with pressurized water reactors (PWR), characterized by a high concentration of sodium and boron [7, 14–17]. Borosilicate glass matrices with high insulating properties are considered as promising candidates.

The aim of the work is an experimental study of the vitrification of the simulant of vat residues of LRW from nuclear power plants with VVER-1000 and the effect of the glass composition on the strength properties and corrosion resistance of the obtained glass matrices.

MATERIALS AND METHODS

Experimental studies of the process of vitrification of liquid radioactive waste were carried out on simulators of vat residues (VR) of LRW from nuclear power plants with VVER reactors. The following salts and solutions were the initial materials for obtaining the VR simulator: H_3BO_3 , NaOH , HNO_3 , NaCl , KOH , NH_4OH , $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$, CsNO_3 . A solution imitating the vat residue was obtained by dissolving weighed portions of the required amount of salts in distilled water to provide the appropriate concentration, according to the data on the average chemical composition of the vat residue of liquid radioactive waste from NPPs with VVER-1000 reactors. The vat residue has the following composition, g/l: Na^+ cations – 140.9; K^+ – 37.9; NH_4^+ – 1.0; Fe^{3+} – 4; Cu^{2+} – 0.74, anions BO_3^{3-} – 150.8; NO_3^- – 156.2; SO_4^{2-} – 6; Cl^- – 2.8. Specific activity for the main isotopes: ^{137}Cs – $5.2 \cdot 10^{-5}$ Ci/l, ^{134}Cs – $2.8 \cdot 10^{-5}$ Ci/l, ^{60}Co – $6.1 \cdot 10^{-6}$ Ci/l [18]. The amount of cesium in the solution-simulator was increased by several orders of magnitude in relation to the calculated amount in accordance with the activity of its isotopes. In this case, it became possible to quantitatively determine cesium in a solution-simulator of VR by mass spectrometric methods.

The study of the composition of the resulting solution-simulator of VR was carried out using a high-resolution mass spectrometer with ionization in inductively coupled plasma ICP-MS ELEMENT 2.

Glass samples were melted in aluminum oxide crucibles in air in a Nabertherm P310 high-temperature electric furnace.

To grind the powders, a Pulverisette 6 planetary monomill was used.

Diffraction studies were carried out on a DRON-4-07 X-ray diffractometer in copper $\text{Cu-K}\alpha$ radiation using a Ni selective absorbing filter. The diffracted radiation was recorded by a scintillation detector. Phases were identified using the database of crystalline compounds ICDD PDF-2 (2004). To determine the vitrification temperature of the VR simulator, differential thermal (DTA) and thermographic (TG) analyzes were performed on an SDT Q600 V20.9 Build 20 thermal analyzer in the temperature range of 20–1200°C, heating rate 20°C/min.

Elemental analysis of samples of vitrified VR simulators was carried out on a high-resolution EMAL-2 laser mass spectrometer with double focusing according to Mattauch-Herzog. The random error of the analysis results is characterized by a relative standard deviation of 0.1–0.3.

The study of the microstructure of the obtained samples was carried out on a scanning (SEM) electron microscope JSM-7001F.

An IKS-29 IR spectrophotometer (LOMO) was used to record absorption spectra in the IR range.

The apparent density of the obtained samples was determined by hydrostatic weighing.

To prepare a solution-simulator of the vat residue, the required amounts of salts H_3BO_3 , NaOH , NaCl , KOH , $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ was dissolved in distilled water, then nitric acid HNO_3 and a solution of ammonia hydrate NH_4OH were added. The density of the model solution imitating the vat residue is 1300 g/l, pH = 11.

In order to determine the compliance with the composition of the VR simulator and real vat residue of NPPs with VVER-1000 reactors, the concentrations of the following elements in the obtained solution-simulator were measured: B, Na, K, S, Cl, Fe, Cu, Cs. The studies were carried out using a high-resolution mass spectrometer with ionization in inductively coupled plasma ICP-MS ELEMENT 2. The initial probe of the simulator solution was diluted 150,000 times. The results of the analysis, together with the targeted composition of the VR, are presented in Table 1. There is a fairly good agreement between the measured and the targeted composition of the VR. A slight discrepancy between the targeted and measured amounts of elements is probably due to the difficulty in taking a representative sample and the presence of insoluble precipitate in the simulant solution.

Before calcination, the obtained solution imitating the vat residue was dried at a temperature of 90 °C for 35 hours. The calcination was carried out in air at a temperature of 600 °C for 1 hour. After heat treatments, the mass of the obtained calcinate decreased by more than 3 times compared with the mass of the initial solution-simulator of VR. Then, the calcinate was ground in a planetary monomill at a speed of 300 rpm within 30 minutes. The resulting powder consisted of particles $\leq 10 \mu\text{m}$ in size.

Table 1. Targeted and measured composition of the vat residue solution

Element	Targeted concentration, g/l	Measured concentration, g/l
B	27.7	<16.8
Na	140.9	<114.6
K	37.9	<22.2
S	2.0	<1.7
Cl	2.8	<1.1
Fe	4.0	<4.2
Cu	0.7	<2.4
Cs	<0.1	<0.45

The process of vitrification of the VR simulator consisted of mixing powdered calcinate with glass-forming powder additives and subsequent melting of glass in aluminum oxide crucibles. Taking into account the fact that, in terms of radiation and corrosion resistance, borosilicate glass is one of the optimal matrices for localizing radioactive waste, various compositions of borosilicate matrices for vitrification of salt boron-containing wastes were studied. Since the content of Na and B in the vat residues of LRW from NPPs with VVER reactors is quite high, there is no need to add components containing these elements to obtain borosilicate glass. SiO₂ powder in the form of quartz sand was used as the main glass-forming additive. As is known, one of the main requirements for the vitrification of medium-level waste is the use of a minimum amount of additives in the melting of glass matrices [10]. Therefore, in the course of experiments on vitrification of the VR simulator, lead oxide, barium oxide, and calcium fluoride were added separately. It is known that the vat residues of NPPs with VVER-1000 reactors include Na₂SO₄ and NaCl components, which during the process of immobilization in borosilicate glasses may cause phase separation associated with the immiscibility of silicate and sulfate (chloride) systems [7, 19]. In order to prevent phase separation and the appearance of a sulfate-chloride phase during the vitrification of sulfate- and chloride-containing LRW, it is necessary to add PbO, BaO, and CaF₂ additives [20-22]. The following glass matrices SK45, SK45-Pb10, SK45-Ba10, and SK45-CF10 were obtained (Table 2).

Table 2. Chemical composition (initial) and melting temperature of glass matrices

Glass matrix	Compounds, wt. %					Melting temperature, °C
	SiO ₂	PbO	BaO	CaF ₂	VR	
SK45	45				55	1150
SK45-Pb10	45	10			45	1150
SK45-Ba10	45		10		45	1100
SK45-CF10	45			10	45	1150

The glass melting temperature and time were determined from the results of experiments to obtain the highest quality glass according to visual observations and with an acceptable viscosity for pouring the resulting glass from a crucible into metal molds (Fig. 1a). The optimal temperature-time conditions for vitrification of VR simulators were determined: 1100 – 1150°C, holding time 1 hour. The cube-shaped metal molds with the resulting glasses were annealed and cooled to room temperature in the switched off furnace. The samples of glass matrices SK45, SK45-Pb10, SK45-Ba10 and SK45-CF10 obtained after extraction from the metal mold are shown in Fig. 1b. As can be seen, samples-cubes with an edge length of 2 cm of all the above compositions of glass matrices are transparent and characterized by high-quality penetration.

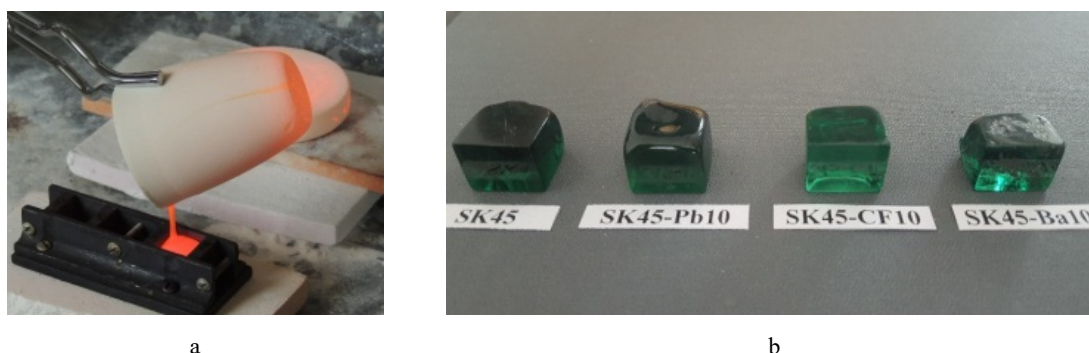


Figure 1. Spill (a) and appearance of samples of glass matrices (b).

RESULTS AND DISCUSSION

Study of the phase composition of the mixture to obtain the SK45 glass matrix after heat treatment at a temperature of 600°C for 1 hour showed the presence of the following phases: sodium nitrate NaNO_3 (JCPDS 36-1474), sodium tetraborate $\text{Na}_2\text{B}_4\text{O}_7$ (JCPDS 29-1179) and glass-forming silicon oxide SiO_2 (JCPDS 39-1425). Sodium nitrate and sodium tetraborate are the main components of the vat residues. In addition, in the region of angles $2\theta \approx 24\text{--}35^\circ$, there is a diffraction halo, which indicates the presence of an amorphous phase in the sample (Fig. 2a).

The presence of an amorphous phase in the material of the SK45 glass matrix indicates that at a temperature of 600°C the process of glass formation is already underway. This fact is confirmed by the analysis of thermograms taken with the SDT Q600 V20.9 Build 20 thermal analyzer (Fig. 3). It can be seen that for the glass matrices SK45 and SK45-Pb10, an endothermic effect is observed, which starts at a temperature of about 430°C [23]. This effect corresponds to the beginning of the glass transition process, which causes the presence of an amorphous phase at higher heat treatment temperatures.

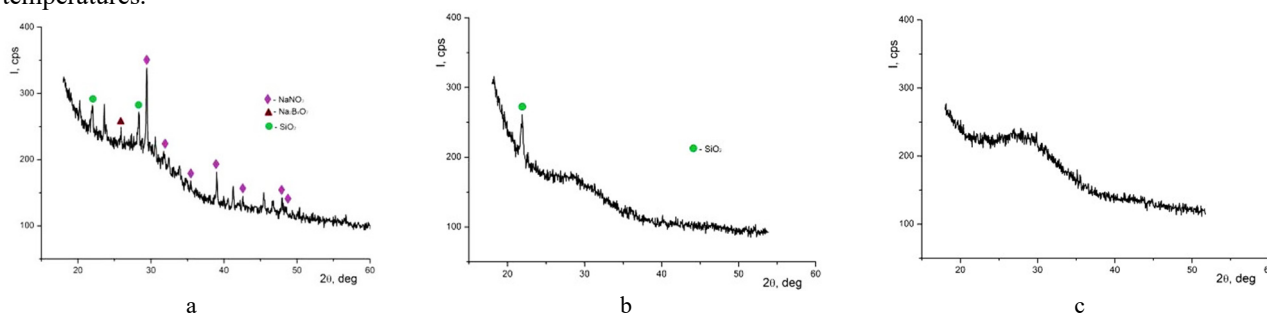


Figure 2. Diffractograms of SK45 glass matrix after heat treatment: 600°C, 1 hour (a), 700 °C, 1 hour (b), 1150°C, 1 hour (c)

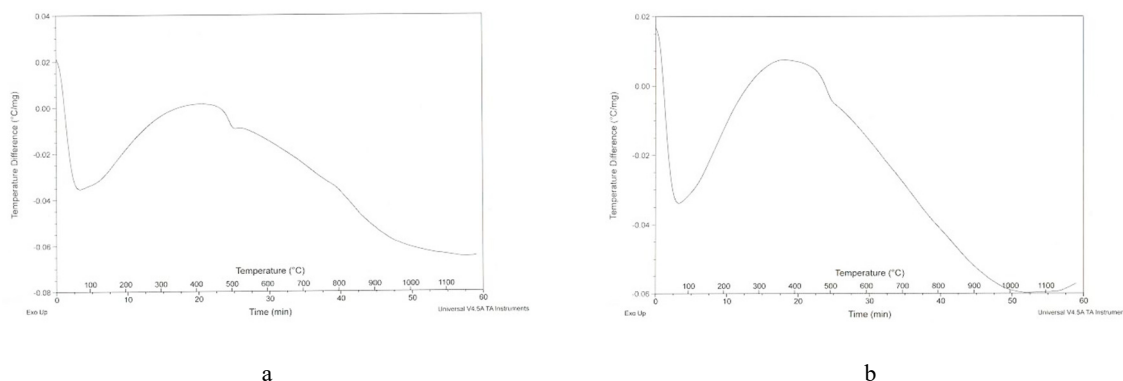


Figure 3. Thermograms of samples of glass matrices SK45 (a) and SK45-Pb10 (b).

An increase in the melting temperature of the SK45 glass matrix material to 700 °C leads to the disappearance of reflections of the components present in the distillation residues, namely sodium nitrate and sodium tetraborate. A decrease in the number of silicon oxide reflections to one is also observed (Fig. 2b). Further, after melting glass at a temperature of 1150°C for 1 hour, according to X-ray phase analysis, only a diffraction halo in the region of angles $2\theta \approx 24 \div 35^\circ$ is present in the diffraction pattern of the glass matrix SK45, i.e. the sample consists entirely of the amorphous phase (Fig. 2c). It should be noted that, according to XRD analysis, all obtained glass matrices SK45, SK45-Pb10, SK45-Ba10, and SK45-CF10 are X-ray amorphous.

Also, the absence of crystalline inclusions and segregation zones is confirmed by the results of scanning electron microscopy. The homogeneity of the glass matrix material and the absence of segregation zones are also confirmed by data on the uniformity of the distribution of glass matrix elements and cesium, obtained using X-ray cartography (Fig. 4).

Since glass is a brittle material, resistance to mechanical failure is an important criterion for evaluating the physical stability of a solidified waste form. In this regard, the mechanical properties of glassed LRW undoubtedly require quantitative evaluation, and their values must comply with the acceptance criteria for storage and disposal.

The strength test under uniaxial compression of samples of vitrified VR simulators was carried out on an electromechanical press of the “ZD 10/90” brand. During testing, the obtained samples-cubes of glass matrices with an edge length of 2 cm were installed with one of the selected faces on the lower support plate of the press in the center relative to its longitudinal axis. Next, loading was carried out and the forces of destruction of glass samples were determined. The determination of the compressive strength was calculated by dividing the breaking force by the working area of the glass sample. The results of tests to determine the compressive strength of the obtained samples is shown in Table 3. In addition, the values of the apparent density of the obtained glass matrices are presented.

The apparent density values increase significantly with the addition of lead oxide to the glass composition. Also, samples of the glass matrix SK45-Pb10 are characterized by the maximum value of the compressive strength. Sufficiently

high compressive strength values are also observed for glass matrix samples SK45, SK45-Ba10, and SK45-CF10. Thus, the authors of [8] provide data on the compressive strength of borosilicate glass for radioactive wastes in the range of 22-54 MPa. In addition, the obtained values of the ultimate compressive strength significantly exceed the norm for mechanical strength, which characterizes the quality of hardened high-level waste (9 MPa) [24].

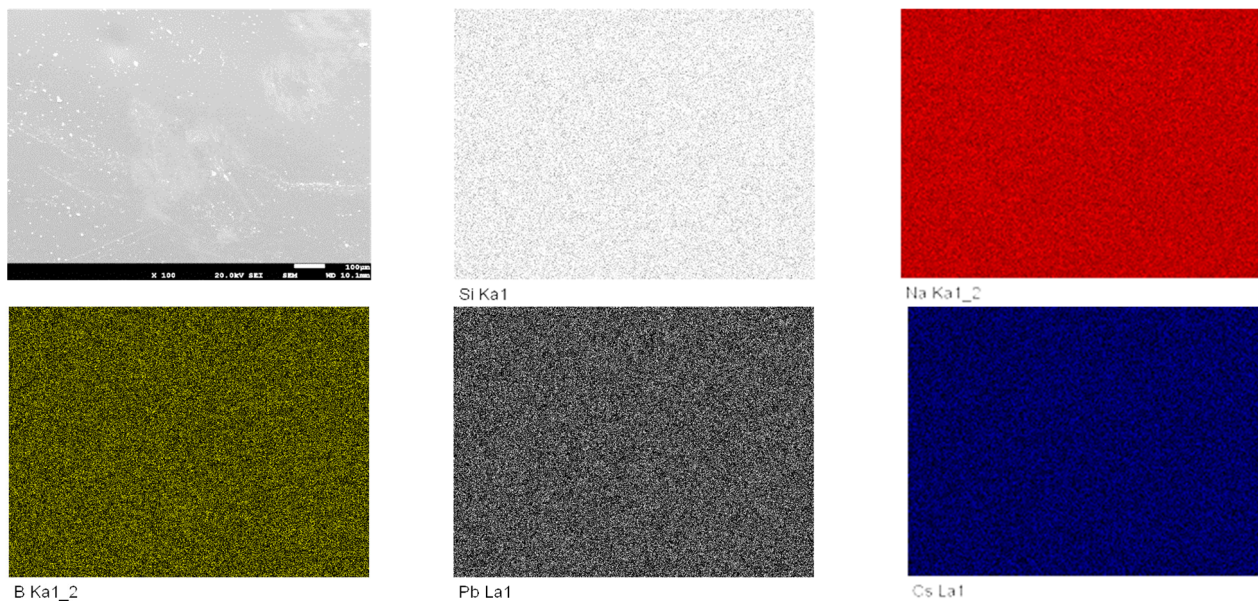


Figure 4. X-ray cartography of a cleavage of glass matrix sample SK45-Pb10: a – structure, b – Si, c – Na, d – B, e – Pb, f – Cs elemental distribution

Table 3. Density and compressive strength of glass matrix samples

Glass matrix	Density, g/cm ³	Compressive strength, MPa
SK45	2.62	78.5
SK45-Pb10	2.84	136.0
SK45-Ba10	2.59	59.5
SK45-CF10	2.27	72.6

To ensure reliable isolation of radionuclides included in the glass matrix for a long time, the vitrified waste form must have certain physical and mechanical characteristics. Since the main route of radionuclides entry from vitrified forms of waste into the environment is leaching by groundwater, it is necessary to determine their corrosion resistance [15].

The main parameter characterizing the corrosion resistance of the glass matrix is the leaching rate of radionuclides and matrix elements. The loss of total mass during leaching has the same function as the rate of leaching under the same test conditions. Mass loss is fairly easy and quick to measure. Therefore, this parameter was initially used to control the corrosion resistance of the glass matrix in order to determine the optimal glass composition for vitrification of the vat residue simulators.

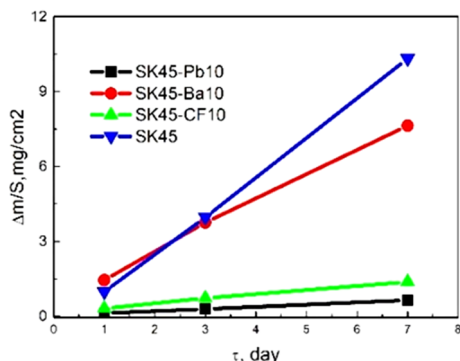


Figure 5. Dependence of mass loss of glass matrix samples on leaching time

As can be seen, the best indicators of corrosion resistance are observed on samples SK45-Pb10. The mass loss of the SK45-Pb10 sample on the 7th day of leaching is $6.4 \cdot 10^{-4}$ g/cm². As is known, the presence of a small amount of lead oxide significantly reduces the mobility of sodium ions in a glass frame. Sufficiently high corrosion resistance is demonstrated by samples of the glass matrix SK45-CF10. However, the weight loss of the SK45-CF10 glass matrix

The leaching test was carried out in Teflon containers at a constant temperature of 90 °C for 1, 3 and 7 days. The vitrified samples of the VR simulator had the shape of a cube with an edge of 2 cm. The mass of the samples was measured before and after leaching. The loss of the total mass related to the total surface of the samples $\Delta m/S$ (g/cm²) was calculated by the formula:

$$\Delta m / S = \frac{(m_0 - m_t)}{s} \quad (1)$$

where m_0 (g) is the initial mass of the sample, m_t (g) is the mass of the sample after t days of leaching, S (cm²) is the total surface of the sample.

The results of leaching of glass matrix samples SK45, SK45-Pb10, SK45-Ba10 and SK45-CF10 are shown in Fig. 5.

samples on the 7th day of leaching is $1.37 \cdot 10^{-3} \text{ g/cm}^2$, which is more than twice the weight loss of the SK45-Pb10 sample. According to the authors of [20], the addition of fluorine ion has a destructive effect on the silicon-oxygen framework of the glass matrix. As a result, spatial cavities are formed in the glass matrix to accommodate sulfate ions.

In turn, a noticeable destruction of the silicon-oxygen framework can lead to the degradation in the corrosion resistance of the glass. Also, finely dispersed crystalline phases of fluorine-containing compounds are formed with a certain concentration of fluorine in glass. The fluorine-containing crystalline particles act as nuclei for heterogeneous glass crystallization.

A more noticeable destruction of the silicon-oxygen framework of the SK45-CF10 glass matrix compared to SK45-Pb10 is observed in the IR spectra of samples of these glass matrices (Fig. 6). The IR spectrum of the SK45-Pb10 glass matrix sample has a broad band in the region of 470 cm^{-1} and a broad doublet band with weak maxima at 980 and 1020 cm^{-1} . These bands refer, respectively, to the bending and stretching vibrations of O-Si-O in the silicon-oxygen glass network. The spectrum exhibits a weak peak at 700 cm^{-1} associated with symmetric stretching vibrations of the Si-O-Si bridges, as well as a peak at 1590 cm^{-1} (deformation vibrations of OH in water molecules) and a doublet at 3400 and 3450 cm^{-1} due to the presence of two types of H_2O : structurally bound and absorbed [25–28]. The spectrum of the glass matrix sample SK45-CF10 also contains a number of broad bands with a sharp maximum at 470 cm^{-1} and a diffuse maximum in the region of 980 cm^{-1} , which are due to bending and stretching vibrations of the Si-O bonds. However, there is no second maximum of the 1020 cm^{-1} doublet. In addition, there are differences in the spectral band associated with stretching and bending vibrations of structurally bound and absorbed water molecules and hydroxyl groups.

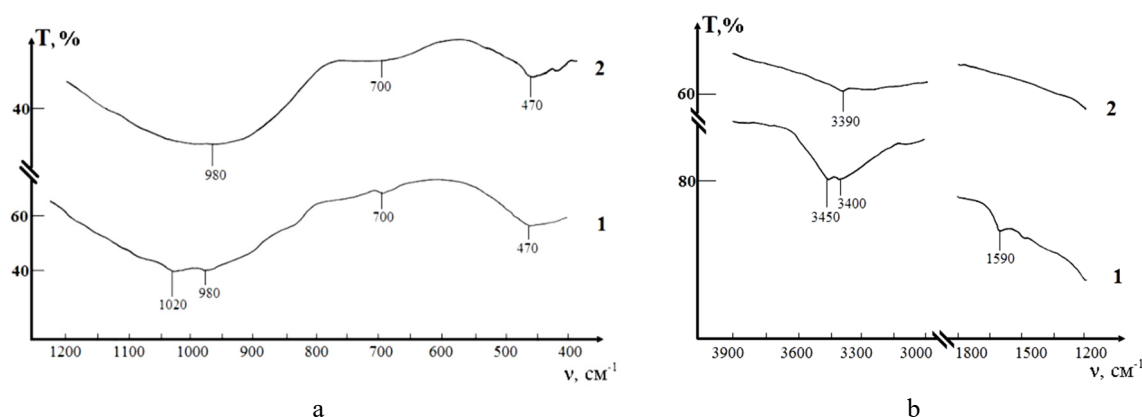


Figure 6. IR absorption spectra of glass matrices (curve 1 – SK45-Pb10, curve 2 – SK45-CF10) in the range: a – $400\text{--}1200 \text{ cm}^{-1}$, b – $1200\text{--}4000 \text{ cm}^{-1}$

After the adding of barium oxide into the composition of borosilicate glass, an increase in mass loss during leaching test is observed. The greatest weight loss during leaching is detected on samples of the SK45 glass matrix. The reduced corrosion resistance of the SK45 sample compared to other tested glasses is explained by the fact that the SK45 glass matrix is welded from 45 wt.% silicon oxide and 55 wt.% VR simulators. While all other glass matrices are welded with a load of 45 wt.% of the VR simulators. This fact means that SK45 glass matrix contains the largest amount of easily soluble sodium and potassium compared to other glass matrices, and explains the degradation of the corrosion properties of the SK45 glass matrix.

Conducted tests for corrosion resistance with an increase in leaching time up to 28 days showed a decrease in the weight loss of samples of glass matrices with time. The mass loss of the S45-Pb10 sample on the 28 day of leaching is $1.06 \cdot 10^{-3} \text{ g/cm}^2$, and that of the S45-CF10 sample is $2.83 \cdot 10^{-3} \text{ g/cm}^2$. The weight loss of the S45-Pb10 sample is more than two times less than the weight loss of the SL-1 glass matrix sample ($2.44 \cdot 10^{-3} \text{ g/cm}^2$) at the same waste load (45 wt.%), which was proposed as the matrices for immobilization of borate waste from PWR reactors [4].

The leaching rate of SK45-Pb10 and SK45-CF10 glass matrix elements during leaching tests was determined using the following equation:

$$v = \frac{m_n^i}{f_i \cdot SA \cdot t_n} \quad (2)$$

where:

v is the leaching rate ($\text{g/cm}^2 \cdot \text{day}$),

m_n^i is the mass of the i -th element (g) leached over a given time interval,

f_i is the mass fraction of the i -th element in the initial sample (g/g),

SA is the open geometric surface of the sample (cm^2),

t_n is the duration of the n -th leaching period (day).

The mass of each element in the leach for the n th leaching period was determined using a high-resolution mass spectrometer with ionization in inductive plasma ICP-MS ELEMENT 2 and f_i - EMAL-2 laser mass spectrometer.

The values of the leaching rates of the elements Si, B, Na, K, Pb, Ca, and Cs from the glass matrices SK45-Pb10 and SK45-CF10 are shown in fig. 7. For the glass matrix SK45-Pb10, during the first day, the highest leaching rate is observed for cesium, potassium, and sodium, and the lowest for lead. With an increase in leaching time, the rate of potassium and cesium leaching decreases significantly, but the rate of sodium leaching remains relatively high. Thus, the leaching rate of Cs after 14 days is $1.5 \cdot 10^{-5}$, K – $5.22 \cdot 10^{-6}$, Na – $2.53 \cdot 10^{-5}$, Pb – $1.72 \cdot 10^{-6}$ g/cm²·day. The glass matrix SK45-CF10 is characterized by the highest leaching rates for sodium ($5.8 \cdot 10^{-5}$) and boron ($5.1 \cdot 10^{-5}$ g/cm²·day). The smaller values of leaching rate are observed in the case of cesium ($3.8 \cdot 10^{-5}$) and potassium ($2.4 \cdot 10^{-5}$ g/cm²·day). The lowest leaching rates are detected for calcium ($2.5 \cdot 10^{-6}$ g/cm²·day). It should be noted that the cesium leaching rate from the SK45-CF10 glass matrix is more than 2 times higher compared to the SK45-Pb10 glass matrix.

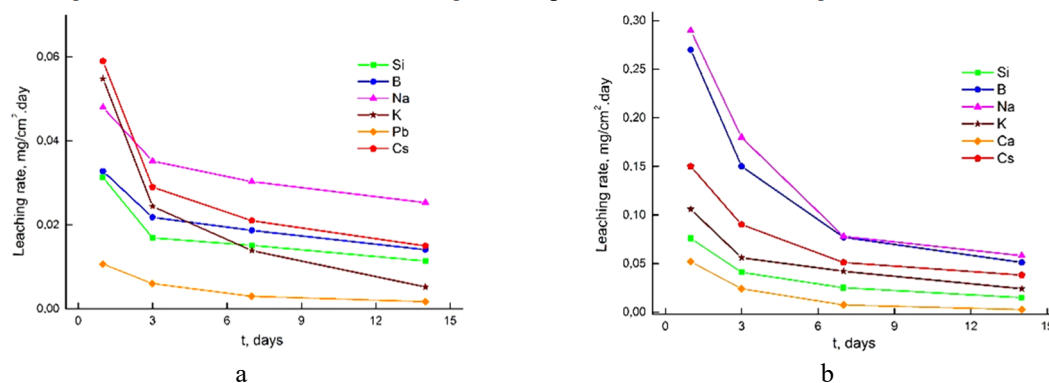


Figure 7. Leaching rates of glass matrix elements: (a) SK45-Pb10, (b) SK45-CF10

The obtained rates of cesium leaching at a temperature of 90 °C from a vitrified simulator of VR are consistent in order of magnitude with the data given in the literature on the leaching rate of ¹³⁷Cs from vitrified wastes of low and medium activity at the same (45 wt.%) or lower waste load [28]. In general, these values indicate a sufficiently high corrosion resistance of glasses, which are quite acceptable for the immobilization of LRW vat residues.

Previously, the absorbed dose which is accumulated by borosilicate glass matrices with included vat residues during 300 years of storage was calculated, and studies of the effect of simulated X-ray irradiation on their physical and mechanical properties were also carried out. It was shown that a lead-borosilicate glass matrix has a high radiation resistance [29]. The obvious advantage of the obtained glass matrices is the relatively low melting point along with the high content of included waste. Thus, the glass matrix S45-Pb10, containing 45% imitators of LRW vat residues, in combination with high strength, corrosion and radiation characteristics, is promising candidate for curing of the boron-containing salt LRW.

CONCLUSION

1. Experimental studies of the process of immobilization of the imitator of vat residues residues of LRW from nuclear power plants with VVER-1000 reactors in borosilicate glass were carried out. To obtain borosilicate glass matrices, quartz sand, oxides of lead, barium, and calcium fluoride were used.
2. Tests for mechanical strength under uniaxial compression of cubic samples of borosilicate glass matrices of various compositions were carried out. It has been established that the maximum compressive strength of lead-borosilicate matrix specimens is 136.0 MPa. Samples of this glass matrix have maximum density values – 2.84 g/cm³.
3. According to the results of SEM and XRD studies of lead-borosilicate matrices, the prepared samples are X-ray amorphous, characterized by a dense and uniform structure, and have no crystalline inclusions.
4. Studies of the corrosion resistance of the obtained samples of glass matrices by leaching in water were carried out. It has been established that lead-borosilicate matrices are the most leaching-resistant glass matrices for immobilization of the vat residue simulators. The cesium leaching rate is $1.5 \cdot 10^{-5}$ g/cm²·day.
5. It is shown that the obtained lead-borosilicate glass matrices are the most optimal candidates for immobilization of the vat residues of LRW from NPP. These glass matrices are characterized by high mechanical strength, corrosion and radiation resistance and include the maximum amount of LRW simulator – up to 45 wt.%.

Competing interests. The authors declare that they have no competing interests.

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ОСКЛУВАННЯ ІМІТАТОРА КУБОВИХ ЗАЛИШКІВ РІДКИХ РАДІОАКТИВНИХ ВІДХОДІВ

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В роботі проведено дослідження та обґрунтування використання оптимальних складів скла для осклування імітатора кубових залишків РРВ АЕС з реакторами ВВЕР-1000 в залежності від таких параметрів, як температура осклування, міцність, корозійна стійкість, відсутність кристалічних фаз, мінімізація скло-утворюючих добавок та включення максимальної кількості відходів. Встановлено, що температура варіння свинцево-боросилікатних скло-матриць становить 1150 °С, що задовольняє вимогам щодо осклування низько- та середньо-активних відходів. Межа міцності на стиск отриманих зразків скло-матриць становить 136.0 МПа. Також показано, що свинцево-боросилікатні скло-матриці є найбільш стійкими до вилугування. Швидкість вилугування цезію становить $1,5 \cdot 10^{-5}$ г/см²·добу.

Ключові слова: рідкі радіоактивні відходи; кубовий залишок; осклування; міцність; швидкість вилугування