



Original Article

Solidification of high level waste using magnesium potassium phosphate compound

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ABSTRACT

Compound samples based on the mineral-like magnesium potassium phosphate matrix $\text{MgKPO}_4 \times 6\text{H}_2\text{O}$ were synthesized by solidification of high level waste surrogate. Phase composition and structure of synthesized samples were studied by XRD and SEM methods. Compressive strength of the compounds is 12 ± 3 MPa. Coefficient of thermal expansion of the samples in the range 250–550 °C is $(11.6 \pm 0.3) \times 10^{-6} 1/^\circ\text{C}$, and coefficient of thermal conductivity in the range 20–500 °C is 0.5 W/(m × K). Differential leaching rate of elements from the compound, g/(cm² × day): Mg - 6.7×10^{-6} , K - 3.0×10^{-4} , P - 1.2×10^{-4} , ¹³⁷Cs - 4.6×10^{-7} ; ⁹⁰Sr - 9.6×10^{-7} ; ²³⁹Pu - 3.7×10^{-9} , ²⁴¹Am - 9.6×10^{-10} . Leaching mechanism of radionuclides from the samples at the first 1–2 weeks of the leaching test is determined by dissolution (¹³⁷Cs), wash off (⁹⁰Sr) or diffusion (²³⁹Pu and ²⁴¹Am) from the compound surface, and when the tests continue to 90–91 days - by surface layer depletion of compound. Since the composition and physico-chemical properties of the compound after irradiation with an electron beam (absorbed dose of 1 MGy) are constant the radiation resistance of compound was established.

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1. Introduction

Liquid radioactive waste (LRW) formed from the reprocessing of spent nuclear fuel (SNF) must be immobilized into a stable solid form suitable for long-term and safety disposal. Vitrification is today the only technology of high level waste (HLW) management brought to industrial implementation [1]. The main drawbacks of such high temperature technology are high cost and short operation lifetime of the melting equipment, and low chemical and crystallization resistance of the glass-like compound. It should be noted especially that liquidation of the melting equipment after finishing of operation lifetime is a major and still unsolved radiological problem.

Crystalline (mineral-like) phosphate matrices [2], including the magnesium potassium phosphate (MKP) matrix $\text{MgKPO}_4 \times 6\text{H}_2\text{O}$ [3–13], obtained at room temperature during chemical interaction between magnesium oxide and potassium dihydrogen phosphate in aqueous solution and being a synthetic analogue of the natural mineral K-struvite [14], are considered as an alternative to glass for HLW immobilization. This article presents the results of studying

the composition, structure, and main characteristics (thermal, mechanical, hydrolytic and radiation resistance) of a low-temperature phosphate compound obtained by solidification of the surrogate of industrial HLW formed from the reprocessing of SNF with a high fuel burn-up.

2. Material and methods

The chemicals used in the experiments were of no less than chemically pure grade. The HLW surrogate was prepared by dissolving the element nitrates in an aqueous solution of nitric acid, and molybdenum was added in the form of MoO₃. The chemical and radionuclide composition of HLW surrogate is shown in Table 1. The content of HNO₃ in the obtained HLW surrogate is 3.2 mol/L, and the density is about 1.21 kg/L. Preparation of HLW surrogate for solidification was carried out by neutralizing it to pH 7.5 ± 0.5 with sodium hydroxide solution at a concentration of 15.0 ± 0.1 mol/L. Magnesium oxide (MgO, LLC «RusHim», precalcined at 1300 °C for 3 h, particle size is no more than 50 μm, specific surface area is 6.6 m²/g) and potassium dihydrogen phosphate (KH₂PO₄, LLC «ChimMed», particle size of 0.15–0.25 mm) were used to solidify HLW surrogate. Compounds were obtained with the following reagent ratio, g: MgO: H₂O in HLW surrogate: KH₂PO₄ = 1: 2: 3. To reduce the reaction rate, boric acid in the

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Table 1
The content of elements in HLW surrogate (* - neodymium as a simulator of the REE in HLW).

Element	Content, g/L	Element	Content, g/L	Radionuclide	Specific activity, Bq/L
Na	13.3	Ba	6.4		
Sr	3.9	Nd*	28.8	²³⁹ Pu	3.8×10^8
Zr	7.6	Fe	1.0	²⁴¹ Am	5.2×10^7
Mo	0.9	Cr	2.8	¹³⁷ Cs	2.2×10^8
Pd	5.4	Ni	0.5	⁹⁰ Sr	7.3×10^7
Cs	9.3	U	2.8		

amount of $1.2 \pm 0.1\%$ by weight of the compound was also added to the mixture. The obtained mixture was placed in fluoroplastic molds measuring $2 \text{ cm} \times 2 \text{ cm} \times 2 \text{ cm}$ and stay for at least 15 days to set the strength of the compound.

Previously it was shown [3,7,15] that the stability of the compound can be enhanced by the introduction of various mineral fillers. Therefore, samples were also prepared using wollastonite (FW-200, Nordkalk) or zeolite («Sokirnit», clinoptilolite content 70%, Transcarpathian deposit), filler particle size - $0.07\text{--}0.16 \text{ mm}$, filler content in the samples varied from 10 to 30 wt%.

In addition, an effective method for increasing the stability of the compound to cesium leaching by its preliminary binding in the form of a ferrocyanide compound was proposed in Ref. [12]. Thus, to determine the hydrolytic durability of the compounds, samples were also prepared after the addition of nickel nitrate and potassium ferrocyanide to the neutralized HLW surrogate in a stoichiometric amount for the synthesis of cesium nickel ferrocyanide in an amount of 1.0% by weight of the compound.

The phase composition of the resulting compounds was determined by X-ray diffraction (XRD) (Ultima-IV, Rigaku, Tokyo, Japan). The X-ray diffraction data was interpreted using the specialized Jade 6.5 program package (MDI, Livermore, CA, USA) with PDF-2 powder database. The samples structure was studied by scanning electron microscopy (SEM) (LEO Supra 50 VP, LEO Carl Zeiss SMT Ltd, Oberkochen, Germany) and X-ray spectral microanalysis (X-MAX 80, Oxford Instruments plc, Abingdon, England).

One of the requirements for compounds with solidified HLW is the homogeneity of the compound, which is assessed by the uniformity of the distribution of macrocomponents in its volume in the margin of $\pm 10\%$ [16]. For this, the cubic compound samples with dimensions of $2 \text{ cm} \times 2 \text{ cm} \times 2 \text{ cm}$ divided (sawing or/and crushing) no less than 10 separate fragments of arbitrary shape with a mass of 0.3 ± 0.1 and up to $1.7 \pm 0.1 \text{ g}$ were used. Next, these fragments were completely dissolved in a solution of nitric acid ($6.0 \pm 0.1 \text{ mol/L}$) and the elements content in the resulting solutions was determined.

The compressive strength of the compounds, including after their heat treatment at $450 \text{ }^\circ\text{C}$ during 4 h, heating rate $7 \text{ }^\circ\text{C/min}$ (according to Ref. [16]) was determined using a universal test machine AG-X Plus (Shimadzu, Japan). Compound samples were heat treated in the muffle furnace (SNOL 30/1300, UMEGA, Lithuania), cooling of the samples occurred in the disconnected furnace with its direct cooling.

Investigations of compounds by thermogravimetric analysis and differential-scanning calorimetry (TG/DSC) were carried out in an air atmosphere at temperatures up to $800 \text{ }^\circ\text{C}$ (STA 409 PC Luxx, Netzsch, Germany). The coefficient of thermal expansion (CTE) of the MKP compounds was determined by dilatometry (DIL 402 C, Netzsch, Germany), and the coefficient of thermal conductivity (CTC) was determined using the laser flash unit (LFA 457/2/G MicroFlash, Netzsch, Germany).

The experiments on determination of hydrolytic durability of the compounds was carried out in accordance with the semi-dynamic standard [17] in the fluoroplastic containers with a cover.

Leaching conditions: monolithic compound; temperature $23 \pm 2 \text{ }^\circ\text{C}$; leaching agent - bidistilled water ($\text{pH } 6.6 \pm 0.1$, 200 mL); periodic replacement of the leaching agent after 1, 3, 7, 10, 14 etc. days, the total duration of the test was limited to 90–91 days. Earlier in Ref. [7], we showed that the quantitative removal of the bound water from the compound was achieved by heat treatment of the samples to constant weight at $180 \text{ }^\circ\text{C}$ for 6 h (heating rate $4 \text{ }^\circ\text{C/min}$), which allows to minimize the risk of explosive radiolytic hydrogen formation under conditions of temporary storage and disposal of HLW. For this reason, not only the initial samples were tested for hydrolytic durability, but also the samples after heat treatment by the above method.

The content of radionuclides in solutions was determined by radiometric method (α - and γ -ray spectrometers Canberra, USA; α - β radiometer UMF 2000, LLC RPE «Doza», Russia), and the content of structure-forming elements was determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) (iCAP - 6500 Duo, ThermoScientific, Waltham, MA, USA).

The leaching mechanism of the compound components was evaluated according to the de Groot and van der Sloot model [18], that can be expressed by the linear dependence $\log(B_i)$ on $\log(t)$, where B_i – the total yield of an element i from the sample during contact with water, mg/m^2 ; t – contact time, days. Values of the line slope of this dependence correspond to the following leaching mechanisms of elements from the compounds [19,20]: >0.65 - surface dissolution; $0.35\text{--}0.65$ - diffusion transport; <0.35 - surface wash off (or a depletion if it found in the middle or at the end of the test). The B_i calculation was carried out according to the equation $B_{i n} = C_{i n} \cdot (L/S) \cdot (t_n)^{1/2} / ((t_n)^{1/2} - (t_{n-1})^{1/2})$, where $C_{i n}$ - the content of the element i in solutions after leaching at the end of the period n , mg/L ; L/S - the ratio of the volume of the contact solution and the surface area of the sample, L/m^2 ; t_n and t_{n-1} - the total contact time for the period n and before the beginning of the period n , respectively, day.

The radiation resistance of the compounds was evaluated after their irradiation at the UELV-10-10-C-70 linear accelerator (the energy 8 MeV, IPCE RAS) with a vertically scanning electron beam.

3. Results and discussion

3.1. Composition and structure of the compound

The X-ray diffraction pattern of the MKP compound obtained by solidification of HLW surrogate is shown in Fig. 1. The base of the studied samples of compound is the crystalline phosphate phase - the synthetic analogue of the natural mineral of K-struvite with composition of $\text{MgKPO}_4 \times 6\text{H}_2\text{O}$ (the interplanar spacings are 4.26, 4.14, 3.27, 2.91, 2.78, 2.70, 1.95 Å). Earlier in Refs. [4,7], we found that potassium in the $\text{MgKPO}_4 \times 6\text{H}_2\text{O}$ matrix is partially replaced by LRW surrogate metals and it leads to formation of mixed hydrated phosphates with approximate composition of $(\text{Mg, K, Na, Cs, Sr, Nd} \dots)\text{PO}_4 \times 6\text{H}_2\text{O}$, that is confirmed by the presence of the potassium nitrate phase - KNO_3 (niter, the interplanar spacings are 3.78, 3.74, 3.04 Å) in the samples. It is noted (Fig. 1) that the

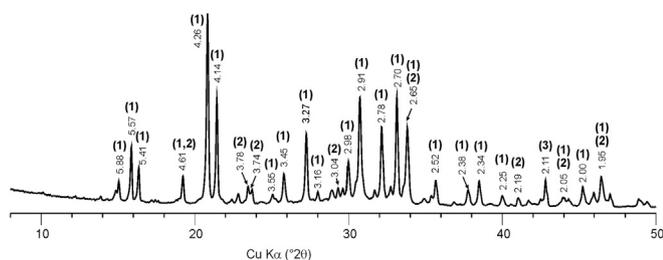


Fig. 1. X-ray diffraction pattern of the MKP compound obtained by solidification of HLW surrogate.

1 – $\text{MgKPO}_4 \times 6\text{H}_2\text{O}$ (K-struvite); 2 – KNO_3 (niter); 3 – MgO (periclase).

individual crystalline phases containing metals of HLW surrogate in the studied samples were not identified. In addition, MgO phase (periclase, the interplanar spacing is 2.11 Å) is present in the compound, which is associated with an excess of the oxide in an amount of about 10 wt% according to the compound synthesis technique [7].

According to SEM data, it was established that the compound consists of two main phases: crystalline hydrate of the mixed phosphate with the basic formula $\text{Mg}_x(\text{K,Na})_y\text{PO}_4 \times (3-6)\text{H}_2\text{O}$, where $2x + y = 3$, like K-struvite, and also potassium nitrate (Fig. 2, MKP and N phases, respectively). Due to the multicomponent composition of HLW surrogate, the quantitative characteristics of the single particles with size range from hundreds of nm to tens of μm in the compound were significantly complicated. In general, it was found that the compound contains chaotically distributed phosphate phases (e.g., light incorporations in Fig. 2), primarily cesium compounds with the general formula $\text{Mg}_x(\text{Cs,K})_y\text{PO}_4 \times n\text{H}_2\text{O}$ (it confirms by the authors' suggestion in Ref. [21] about cesium binding in the MKP matrix) and also anhydrous or hydrated orthophosphates of cesium, strontium and neodymium, or mixtures thereof. Data on the calculation of the composition of incorporations in No. 1 and 2 in Fig. 2 enriched in the above metals are given in Table 2. Thus, it is possible to assume that the chemical binding of Cs, Sr, actinides and rare earth elements (REE) in poorly soluble phosphates during solidification of HLW using MKP matrix will occur, which should provide high stability of the compound to leach of radionuclides. It is also noted that uranium, barium, iron and chromium in the compound are in single incorporations distributed uniformly throughout the volume

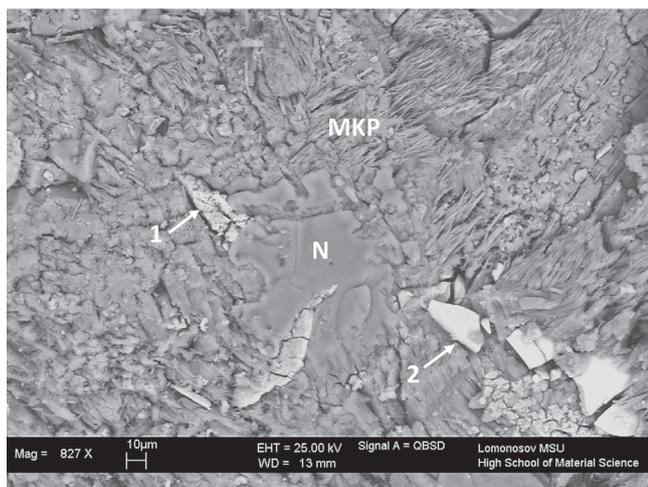


Fig. 2. SEM image of the MKP compound obtained by solidification of HLW surrogate (in back-reflected electrons).

of the compound.

The uniform distribution of macrocomponents (K, Mg, P, Na), as well as radionuclides (^{137}Cs , ^{90}Sr , ^{241}Am) and most other components of HLW surrogate (Nd, Cs, Sr, U, Ba, Cr, Fe, Ni) was established when analyzing the solutions obtained after dissolving of the compound fragments. Changes of element concentration in individual fragments are within $\pm 6\%$ of their average content in the compound. The nonuniform distribution of ^{239}Pu , Zr and Mo was an exception, the cause of which may be the precipitation of these elements under preparing the HLW surrogate for solidification.

3.2. Thermal and mechanical resistance of the compound

According to the TG/DSC analysis, as well as mass spectrometry of the formed gas phase (Fig. 3), it is established that mass of compounds is reduced under heating up to 800 °C (Fig. 3a, TG), first by removing bound water in the range of 100–360 °C (Fig. 3b, spectrum H_2O), and then, obviously, as a result of decomposition of nitrates at 450–720 °C (Fig. 3b, spectra NO and NO_2). Peaks of endothermic effects correspond to these processes in the indicated temperature ranges (Fig. 3a, DSC). The maximum endothermic effect of water removal is in the range 115–120 °C (Fig. 3a), that is confirmed by the data in Refs. [3,7]. Obviously, the main phase of the MKP compound passes into the anhydrous mixed phosphates of compound's elements based on $\text{Mg}(\text{K,Na})\text{PO}_4$, that was noted in Ref. [22].

The compressive strength of the compound samples was about 12 ± 3 MPa. At the same time, the incorporation of mineral fillers - wollastonite and zeolite - into the compound led to a significant increase in its strength. For example, the strength of the compound increased almost linearly with the incorporation of wollastonite and achieved about 40 MPa with a filler content of about 30 wt%. In addition, the presence of fillers allowed to keep the compressive strength of the compounds at the level of requirements for vitrified HLW (9–13 MPa) even after heat treatment at 450 °C. Thus, it is necessary to add mineral filler during solidification of HLW, the heat release of which can lead to the heating of the compound to 300–400 °C. It was found that the optimal amount of wollastonite and zeolite is from 23 to 28 wt%. The elastic modulus (Young's modulus) of the compound containing 28 wt% wollastonite after its heat treatment at 180 °C to remove water was 7.2×10^9 N/m², as well as coefficient of thermal expansion of compound in the range 250–550 °C is $(11.6 \pm 0.3) \times 10^{-6}$ 1/°C and the coefficient of thermal conductivity in the range 20–500 °C is 0.5 W/(m × K).

3.3. Hydrolytic and radiation resistance of the compound

Leaching tests on the MKP compound were carried out according to the standard [17] for 90–91 days. It was established that the pH of the solutions after leaching increases in the first 2 weeks of contact of the compound with bidistilled water and achieves ~10.0, that may be due to the hydrolysis of readily soluble phosphate formed in the compound (probably, Na_3PO_4). When the tests continue, the pH of the solutions after leaching monotonically decreases to ~8.9, which is explained by the gradual dissolution of these formations and, thus, the depletion of the surface layer of the compound.

Kinetic curves of leaching of the main structure-forming elements of the compound (magnesium, potassium and phosphorus) are given in Fig. 4. Differential leaching rate of Mg, K and P on 91st day of contact compound with water was 6.7×10^{-6} ; 3.0×10^{-4} and 1.2×10^{-4} g/(cm² × day), respectively. A higher leaching rate of potassium is associated with the dissolution of the KNO_3 phase formed in the compound (as shown in Section 3.1). Release of phosphorus from the compound occurs due to the dissolving

Table 2
The elemental composition of the particles of the MKP compound with immobilized HLW surrogate.

No.	Content of elements, at.%								Calculated composition of compound particles
	Mg	K	P	O	Na	Nd	Cs	Sr	
1	10.2	1.4	9.3	73.2	0.2	–	5.7	–	$Mg_{1.05}Cs_{0.62}K_{0.15}Na_{0.02}PO_4 \times 3.9H_2O$ or mixture $3.8Mg_{1.33}Na_{0.03}K_{0.19}PO_4 \times 4.9H_2O + Cs_3PO_4$
2	2.0	4.1	9.5	74.6	4.1	5.0	–	0.7	$Mg_{0.21}Sr_{0.07}(Na,K)_{0.43}Nd_{0.53}PO_4 \times 3.8H_2O$ or mixture $Mg_{0.5}NaKPO_4 \times 6.0H_2O + 1.3Sr_{0.12}Nd_{0.93}PO_4 \times 2.1H_2O$

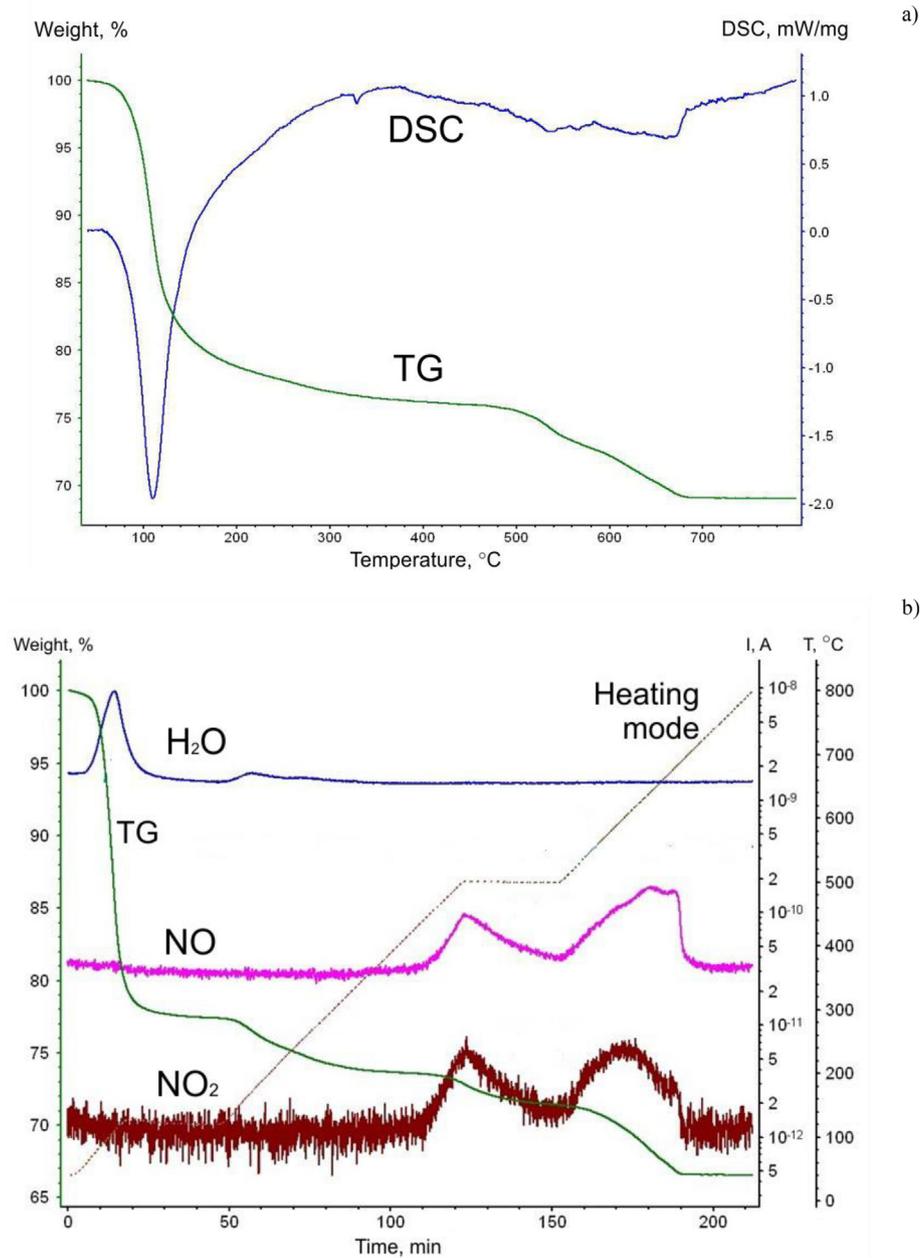


Fig. 3. Thermal behavior of the MKP compound obtained by solidification of HLW surrogate by thermogravimetry methods (a) and mass spectrometry of the formed gas phase (b).

readily soluble phosphate formations, especially Na_3PO_4 . Previously it was established that phase Na_3PO_4 (olypite) is present in the samples during the solidification of an aqueous solution of sodium nitrate [7]. However, the Na_3PO_4 phase was not detected in the samples after the solidification of HLW surrogate by XRD and SEM methods, probably due to its small amount and small-sized crystallites.

Data on the leaching rate of radionuclides from the MKP compound containing 23 wt% wollastonite, including after removal of bound water from the compound at 180 °C (index « term»), are shown in Fig. 5. Differential leaching rate of radionuclides on 90–91st days of contact compound with water was for the ^{137}Cs - 4.6×10^{-7} ; ^{90}Sr - 9.6×10^{-7} ; ^{239}Pu - 3.7×10^{-9} and ^{241}Am - 9.6×10^{-10} g/(cm² × day). It should be noted that the leaching rate

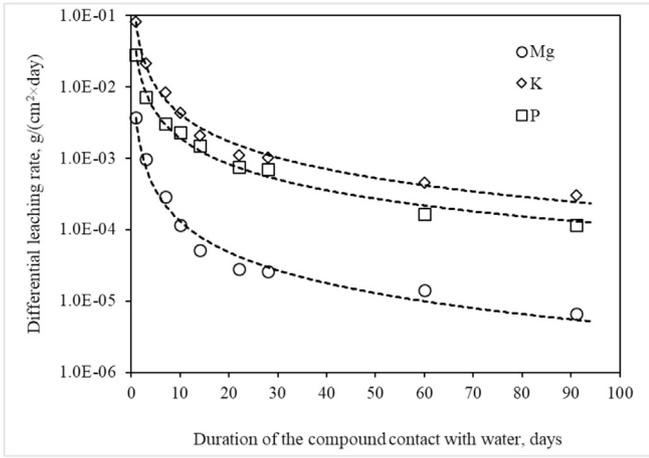


Fig. 4. Kinetic curves of leaching of the basic structure-forming elements (magnesium, potassium, phosphorus) of the MKP compound.

of radionuclides does not change after the removal of bound water from the compound (Fig. 5). Thus, the hydrolytic durability of the MKP compound corresponds to the requirements in Russia for a glass-like compound for HLW immobilization [16] (leaching rate of ¹³⁷Cs, ⁹⁰Sr and ²³⁹Pu must be $\leq 1.0 \times 10^{-5}$, 1.0×10^{-6} and 1.0×10^{-7} g/(cm² × day), respectively).

Based on the results of application of the model [18] at determining the hydrolytic durability of the compound, it was established that the mechanism of radionuclide leaching depends both on their nature and on the contact time of the compound with water (Fig. 6). In the first 14 days of the compound contact with water, the coefficient of equation of linear dependence log (B_i) on log (t) for ¹³⁷Cs is 1.04 (Fig. 6), which corresponds to the release of unbound ¹³⁷Cs from the surface layer of the compound upon dissolution. The further contact of the compound with water up to 91 days leads to the changes of ¹³⁷Cs leaching mechanism, and the release of ¹³⁷Cs occurs due to depletion of the surface layer of the compound (−1.71, Fig. 6). The leaching of ⁹⁰Sr is determined by its wash off from the surface of the compound in the first 7 days, and

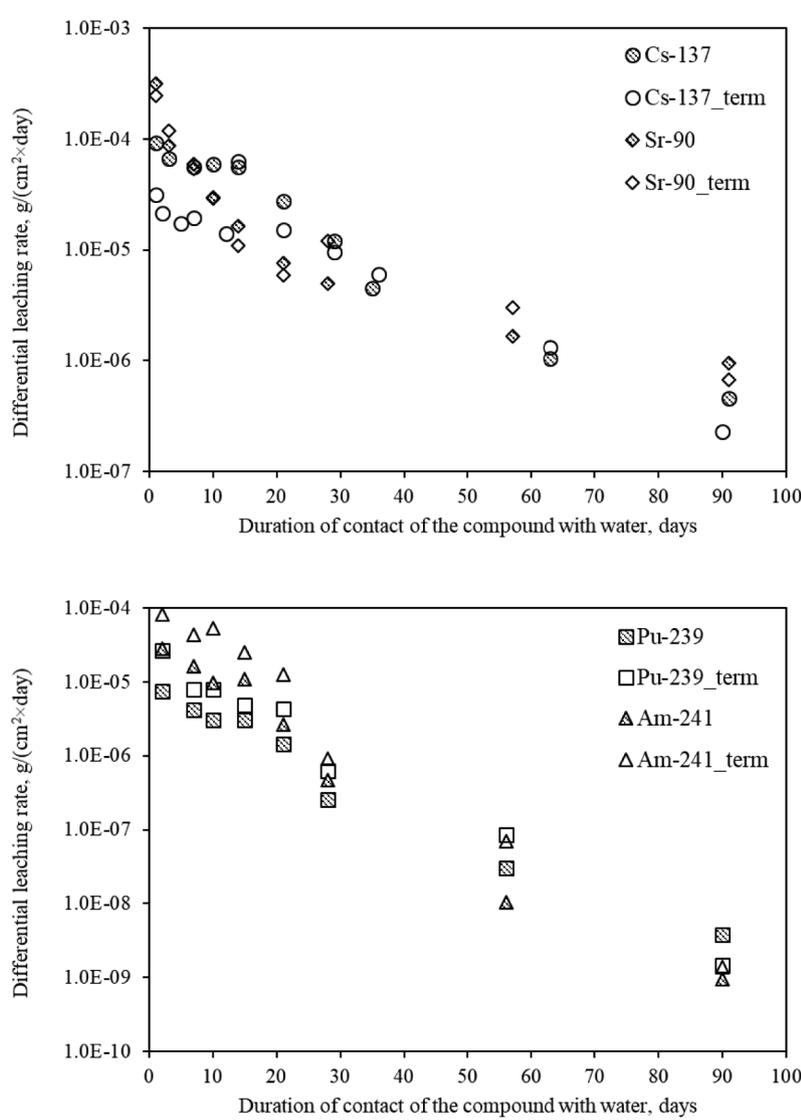


Fig. 5. Kinetic curves of the radionuclides leaching from the MKP compound containing 23 wt% wollastonite, including after removal of bound water during heat treatment (index « term»): a) ¹³⁷Cs and ⁹⁰Sr; b) ²³⁹Pu and ²⁴¹Am.

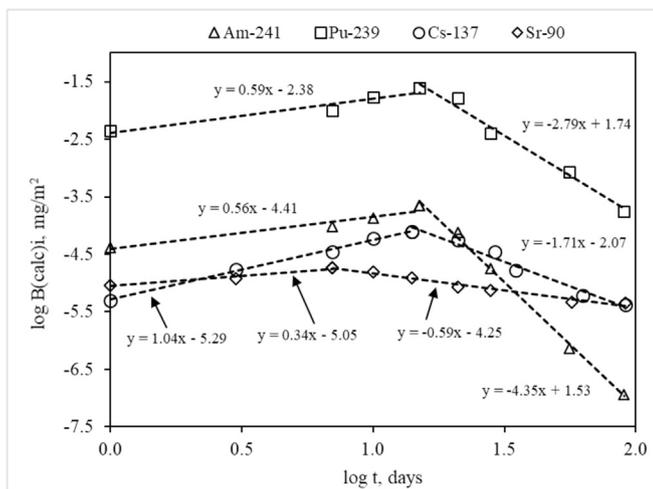


Fig. 6. Logarithmic dependence of the radionuclide release from the MKP compound containing 23 wt% wollastonite on contact time with water.

by depletion of its surface layer for the test duration to 91 days (0.34 and -0.59 , respectively, Fig. 6). The release of ^{239}Pu and ^{241}Am in the first 2 weeks of the compound test is due to diffusion (0.59 and 0.56, respectively, Fig. 6), but then similar to ^{137}Cs and ^{90}Sr due to depletion of the surface layer of the compound (-2.79 and -4.35 , respectively, Fig. 6). The observed sharp decrease in the release of radionuclides during the contact of the compound with water is probably due to the formation of a stable surface layer of the compound depleted of unbound radionuclides and preventing their further leaching. The radionuclide leaching is suggested in future to be determined by their diffusion from the inner layers of the compound with a coefficient of ~ 0.5 according to the model [18].

Much attention was given to the study of the radiation resistance of the compound. It was found that the phase composition, structure and physico-chemical properties of the compound remain uniform under irradiation of the compound by a beam of accelerated electrons with an absorbed dose of 1 MGy.

4. Conclusion

As a result of the carried out researches it was shown that the using of the MKP matrix allows to obtain compounds with high physico-chemical and hydrolytic durability. This allows to consider the MKP compound as a promising material for industrial HLV solidification.

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