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BEHAVIOUR OF MAGNESIUM POTASSIUM PHOSPHATE CEMENT UNDER LEACHING

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The effectiveness of the use of magnesium potassium phosphate cements (MKPC) as protective matrices for radioactive waste was investigated in this work from the point of view of the main elements leaching. Fly ash and blast-furnace slag were used as MKPC fillers. Leaching tests were performed on monolithic MKPC samples according to ANSI/ANS 16.1 test. Based on the obtained data, the cumulative leaching fractions, leaching mechanisms, effective diffusion coefficients and leaching indices K, Mg, P, B, Si, Al, and Ca were determined. The values of the calculated leaching indices (>10) of the constituent elements indicate that the characteristics of the studied MKPC matrix are acceptable for radioactive waste immobilization.

INTRODUCTION

Treatment of low- and intermediate-level waste (LLW/ILW) currently includes cementation in steel barrels. Ordinary Portland Cement (OPC) is traditionally used, mixed with additional binders such as lime, fly ash, ground blast furnace slag, and others [1, 2]. Cement matrices have many advantages: they are inexpensive, easy to use, low tech and suitable for a wide range of radioactive waste. However, there are problematic waste that are not suitable for curing into OPC-based cement matrices. One of them is salt (salt content 200…600 g/l) wastes containing sodium nitrates and borates obtained after the purification of radioactive technological liquids from corrosion products and chemical impurities at water treatment plants. As is known, the use of cement for curing such borate-containing waste is limited by the negative effect of boron compounds on the hydration of Portland cement, which leads to a slowdown in setting, and worsens the strength and chemical resistance of the final product [3].

Therefore, new types of cements are being developed and researched, which have higher rates compared to OPC in terms of strength and durability. There is especially important for protective matrix materials. Unlike OPC, alternative cements are also attractive due to the more effective chemical interaction of hardening products with radionuclides.

Magnesium potassium phosphate cement (MKPC) is being considered as an alternative matrix material for LLW/ILW conditioning. MKPC binder is an acid-base cement system with almost neutral pH, low water consumption, fast solidification and high early compressive strength [4]. Considering that in these cements expensive raw materials are used, in the case of the MKPC matrices mineral additives are proposed such as fly ash, blast furnace slag, silica fume and others. The use of such additives makes it possible to reduce heat generation, increase the solidification time, and increase mechanical strength and corrosion resistance.

The effectiveness of the certain forms waste using is evaluated by the results of leaching tests. It is known that leaching is a complex process and many factors can influence the release of specific components from waste forms over a period of time. The main factors include elemental chemistry, pH, redox potential, complex formation, liquid-to-solid ratio, contact time, etc. [5]. In order to study the behavior of protective matrices in contact with an aqueous medium, the semi-dynamic leaching test ANSI/ANS 16.1 is widely used [6]. In semi-dynamic leaching tests, the sample (monolithic form) is subjected to a leaching test in which the leaching agent is periodically renewed. To compare the relative mobility of various harmful substances and components of the protective matrix, the leaching index LI is used, which is defined as the negative logarithm of the effective diffusion coefficient D.

According to Environment Canada, the criterion for the use and disposal of solidified hazardous waste implies that if LI > 9, the waste may be accepted for "controlled disposal", such as backfilling quarries, closing sumps, roadbeds, or similar uses. If LI > 8, the solidified waste can be disposed in sanitary landfills or separate landfills. If LI < 8, the solidified waste is not considered suitable for disposal [7].

The US Nuclear Regulatory Commission has defined a minimum value of 6 for the leaching index as a criterion for the acceptability of nuclear waste for disposal at controlled sites [8].

In many works [9–16], devoted to the study of the leaching of magnesium-potassium-phosphate cement-stabilized/solidified wastes, the leaching indices of radionuclides and hazardous metals are presented. Thus, depending on the state of aggregation, the chemical composition of the waste, the content of radioactive waste and other factors, various values of LI of radionuclides were obtained: Cs – 15 [9], 11.4…13.0 [10], 11.5…11.7 [11], 11.45 [15], 9.1…9.7 [16]; Sr – 15.66 [15], 12.4…13.1 [16]; Co – 17.63 [15]; Tc – 9.6…9.5 [12], 9 [13], 11.3…14.4 [14]; I > 8.4 [12], 11 [13]; Pu – 13…14 [10], Np – 13…14 [10], Am – 13…14 [10]. The LI values for heavy metals and

mercury are as follows [9]: Pb – 20, Ni – 21, Cd – 22, Cr – 22, and Hg – 18.

An important characteristic for the efficiency of the MKPC matrix is the evaluation of the degree of leaching of its main elements: K, Mg, and P in the aquatic environment. The aim of the study was the calculation of leaching indices based on semi-dynamic leaching data, determination of leaching control mechanisms and evaluation of the applicability of MKPC for radioactive waste immobilization.

MATERIALS AND METHODS

Leaching tests of the MKPC samples in 3x3x3 cm cubes of the following composition: MgO – 6.47, KH_2PO_4 – 21.85, Sand – 28.33, filler – 28.33, Boric acid – 0.57, Water – 14.45 wt.% [17] were carried out. The samples differed in the composition of the filler: MKPC-F with fly ash (FA) from Burshtyn TPP as a filler, MKPC-S with blast-furnace slag (BFS) from the Mariupol Metallurgical Plant, and MKPC-FS - ½ FA + ½ BFS. The chemical composition of fly ash from the Burshtyn TPP and blast furnace slag from the Mariupol Metallurgical Plant is shown in Table.

Chemical composition of fly ash and blast-furnace slag

Industrial waste	Weight content %										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	C	MnO	TiO_2	Na_2O+K_2O	Other components
Fly ash	46.12	18.00	22.17	4.09	1.46	0.21	2.50	0.14	1.78	2.10	1.43
Blast furnace slag	38.20	4.60	1.65	48.5	4.3	–	–	0.81	–	0.82	1.12

The test procedure following the ANSI/ANS 16.1 test is a 90 day semi-dynamic leaching experiment. The procedure consists of immersing a monolithic sample (with a fixed geometry) in distilled water with a fixed volume of liquid to the area of the geometric surface of a solid body ((10±0.2) cm) and sampling the leached solution after a fixed time interval: 2, 5, 17 h, four 24-hourly and 14, 28, and 43-day (total time 90 days). The chemical analysis of the leached solution is carried out using ICP-MS. The obtained values are used to calculate the cumulative fraction of the leach CFL, the effective diffusion coefficient D and the leachability index LI:

$$D = \pi \left[\frac{a_n/A_0}{(\Delta t)_n}\right]^2 T \left[\frac{V}{S}\right]^2, \quad (1)$$

where D is the effective diffusion coefficient (cm^2/s), V – sample volume, cm^3; S is the geometric surface area of the sample, calculated from the measured parameters of the sample, cm^2;

T – average leaching time (s), $T = \left[\frac{1}{2}\left(t_n^{\frac{1}{2}} + t_{n-1}^{\frac{1}{2}}\right)\right]^2$;

a_n – quantity of a given element released from the sample during the leaching interval n-th interval; A_o is total quantity of a given element in the sample before leaching; Δt_n is the duration of the n-th interval (s).

The cumulative leaching fraction CFL is calculated as follows: $CFL = \left(\sum a_n/A_0\right)\left(V/S\right)$.

To determine the concentration of elements in the solution after leaching, a high-resolution mass spectrometer with ionization in inductive plasma ICP-MS ELEMENT 2 was used. The range of measured masses was from 2 to 264 a.e.m.

The phase composition of MKPC was studied by X-ray diffraction analysis (DRON-4-07 in copper Cu-Kα radiation using a Ni selective absorbing filter). The diffraction database ICDD PDF-2 (2004) was used for phase identification.

The study of the microstructure of MKPC samples was carried out on a Superprobe scanning electron microscope (JEOL, Japan)

RESULTS AND DISCUSSION

For the leaching test for each MKPC sample, 1 L polyethylene container with a specially equipped lid were used. The container was filled with the calculated volume of distilled water. Leaching tests were carried out at (25±2.5) °C for 2, 5, 17 h, four 24-hour intervals followed by 14, 28, and 43, 60, and 90-day intervals. Finally, the samples were removed from the solutions after 1 year of testing. After that, the surface layer (~1 mm thick) of the samples that was in contact with the solution was analyzed for phase composition (XRD) and structure (SEM). As can be seen from Fig. 1 all samples after leaching in water for 1 year retained their integrity and shape.

a *b*

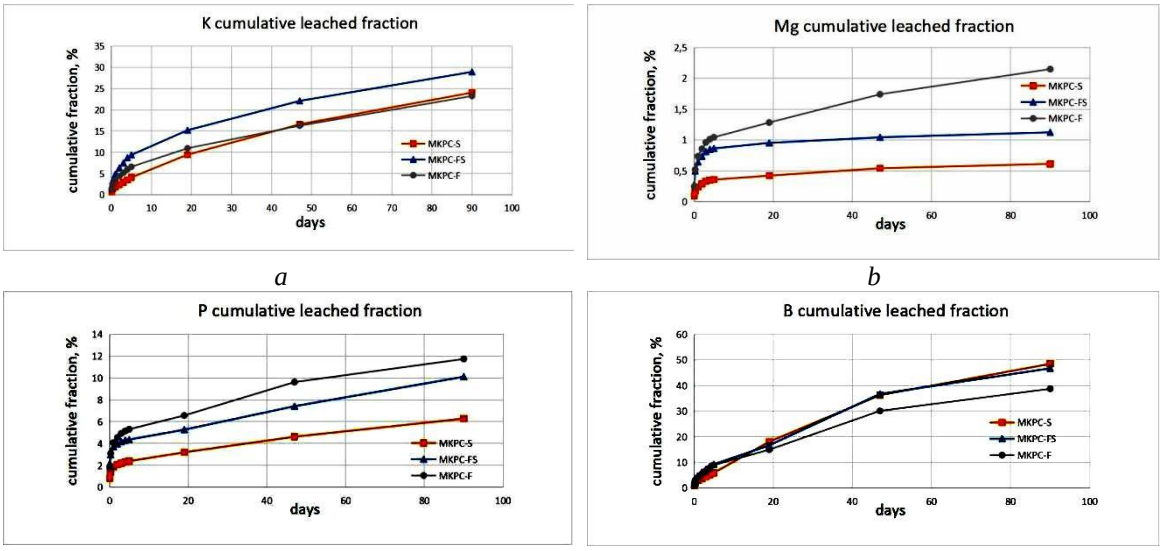
Fig. 1. Samples MKPC-F, MKPC-S, and MKPC-FS (from left to right): a – before, b – after leaching for 1 year

At the end of each leaching interval, 20 ml aliquots of the leachate were taken and visual observations of the samples were made. Aliquots were analyzed using ICP-MS. Based on the measured values for the concentration of elements, the cumulative fractions CFL, effective diffusion coefficients D and leaching indices LI of the main elements of the MKPC matrix: K, Mg, and P, as well as B, Si, Al, and Ca were calculated. These data are needed to evaluate the behavior of the containment waste form under permanent water saturation conditions, which mimic the most conservative disposal scenario.

The cumulative leaching fractions of the MKPC matrix elements are shown in Fig. 2. For all samples, K has the highest values of the cumulative fraction (> 20 %) compared to other structure-forming elements (Mg and P). The character of the CLF of K curves from MKPC-F, MKPC-S, and MKPC-FS depending on time is similar (see Fig. 2,a). The highest CLF of K values are observed in MKPC-FS, despite its lower initial content compared to MKPC-S. The higher content of K in the original MKPC-S is provided by the presence of K in the blast-furnace slag as a filler for MKPC-S (see Table).

The value of CLF of P is lower than CLF of K and is about 6…12% on the 90th day of leaching (see Fig. 2,c). The largest values of CLF of P are observed in MKPC-F, the smaller ones in MKPC-FS, and the smallest ones in MKPC-S. Similarly, the CLF of Mg decreases in MKPC-F, MKPC-FS, and MKPC-S (see Fig. 2,b). CLF of Mg has the lowest values compared to CLF of P and CLF of K.

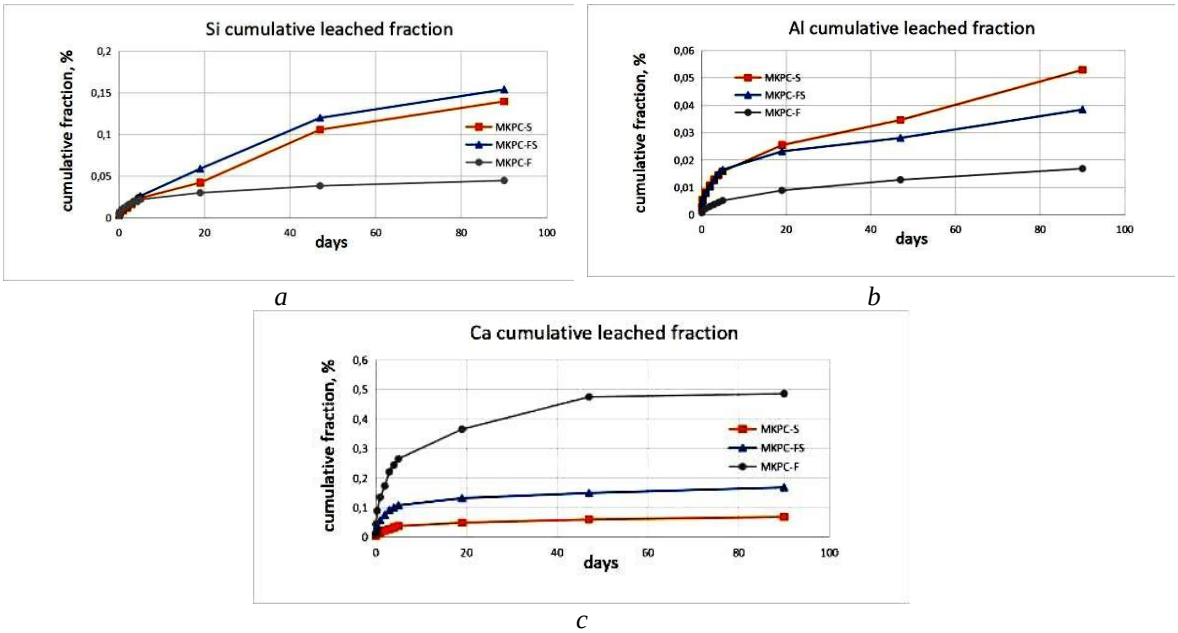
Compared to the CLF of the structural elements, the CLF of B values approach 50% in MKPC-S and MKPC-FS, and in MKPC-F they approach 40% (see Fig. 2,d).



a *b*

c *d*

Fig. 2. Cumulative leaching fractions of elements: a – K; b – Mg; c – P, and d – B



a *b*

c

Fig. 3. Cumulative leaching fractions of elements: a – Si; b – Al, and c – Ca

The values of the cumulative fractions of Si, Al, and Ca, the main filler elements: sand, fly ash, and blast-furnace slag, are much lower than the CLF of structure-forming elements and B. The CLF of Si in MKPC-S and MKPC-FS are close (~ 0.15%) and significantly higher, than in MKPC-F (Fig. 3,a).

CLF of Al show the lowest degree of Al leachability compared to other considered elements. Higher CLF of Al values are observed in MKPC-S and MKPC-FS than in MKPC-F despite the lower initial Al content (see Fig. 3,b).

The similar situation for CLF of Ca was observed. CLF of Ca has the highest values in MKPC-F (see Fig. 3,c). At the same time, the initial Ca content in MKPC-F is the lowest compared to MKPC-S and MKPC-FS, due to the high content of Ca in blast-furnace slag (see Table).

In the case analysis of leaching of monolithic samples, three main mechanisms are considered that usually control this process:

– surface wash-off, when the most soluble phases are removed from the surface of the monolith;

– diffusion transport of dissolved particles in the aqueous phase inside the monolith;

– surface dissolution of a monolith in contact with its surrounding aqueous medium [18]. In this case, the dissolution of the material from the surface occurs faster than diffusion through the pore space of the monolith.

Various published studies [5, 7, 18, 19] show that the determination of the controlling mechanism of leaching can be realized based on the slope of the linear dependence of the logarithm of CLF versus the logarithm of time. If the slope is less than 0.35, the leaching control mechanism will be surface wash-off (or depletion if found in the middle or end of the test), for slope values between 0.35 and 0.65, diffusion will be the control mechanism, and slope values above 0.65 represent the dissolution mechanism.

Plots of log(CLF) versus log(time) for K, Mg, P, and B are shown in Fig. 4. The slope value for K in MKPC-S during the first 5 days of leaching is close to 0.5, indicating that diffusion is the controlling mechanism for K leaching (see Fig. 4,a). As the leaching time increases, the value of the slope increases to 0.65, up to the value of the slope from which the mechanism of dissolution begins to be realized. In this case, dissolution of K occurs both from K-struvite $MgKPO_4 \cdot 6H_2O$ and from other potassium-containing phases of the MKPC-S material, which are formed upon the addition of reactive particles of blast-furnace slag [17, 20]. The process of K leaching from MKPC-F and MKPC-FS during the entire leaching period is controlled by diffusion (slope = 0.4). The effective diffusion coefficients K differ insignificantly and amount to $1.81 \cdot 10^{-11}$ cm^2/s in MKPC-S on the 90th day of leaching, $1.52 \cdot 10^{-11}$ cm^2/s in MKPC-FS and $1.59 \cdot 10^{-11}$ cm^2/s in MKPC-F sample.

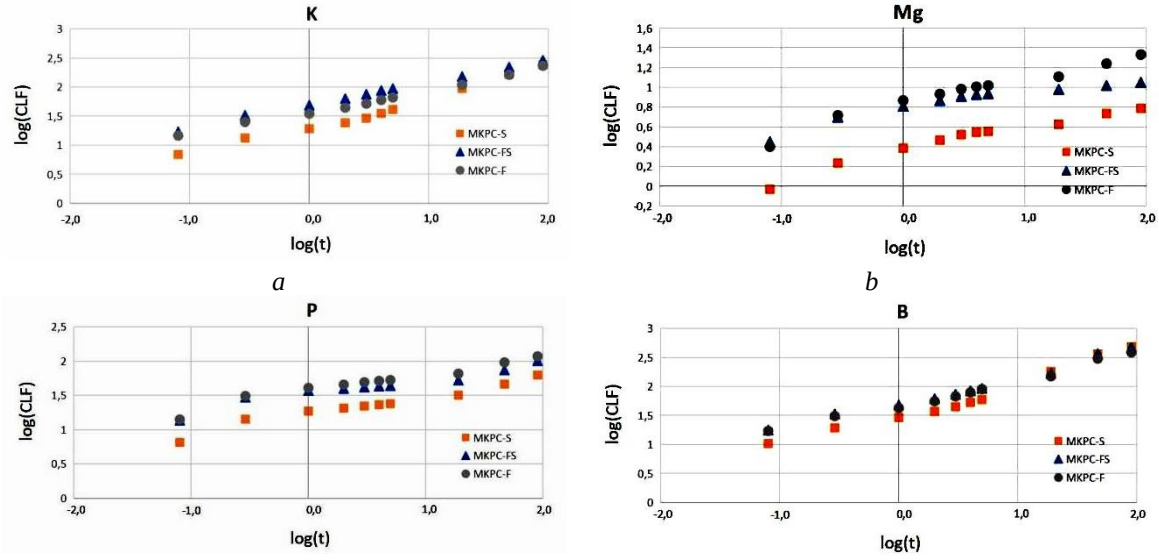
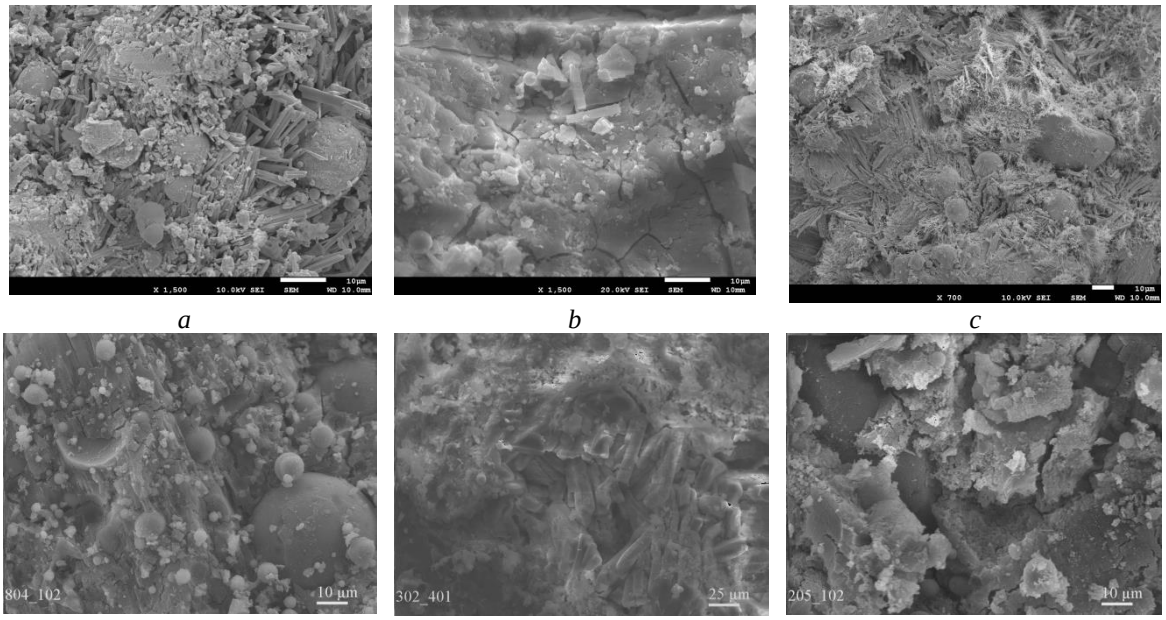


Fig. 4. Log-log plot of cumulative release of: a – K; b – Mg; c – P, and d – B

The dissolution of K-struvite crystals is quite clearly visible on the surface of the MKPC-S sample after leaching for 1 year. Fig. 5 shows the cleavage microstructure of MKPC samples before leaching and the surface microstructure of MKPC samples after leaching tests. Before leaching, K-struvite crystals have a well-cut prism shape (see Fig. 5,a,b). After leaching, K-struvite crystals with rounded faces are visible on the surface of the MKPC-S sample, which is caused by their partial dissolution (see Fig. 5,e). Changes in the surface of K-struvite crystals in an aqueous medium were also observed by Linlin Chong et al. [21].



a *b* *c*

d *e* *f*

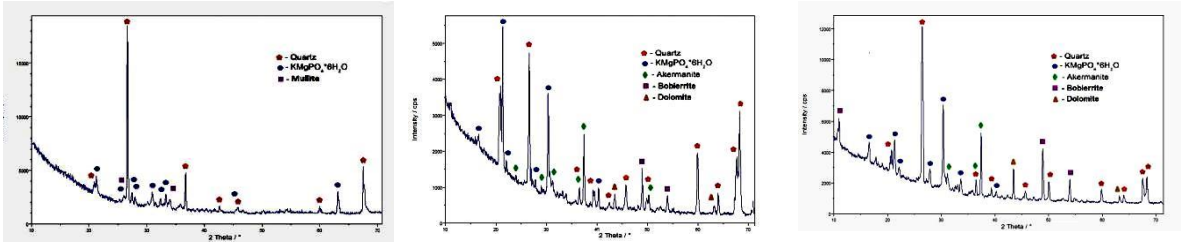
Fig. 5. SEM image of MKPC structure before: a – MKPC-F; b – MKPC-S; c – MKPC-FS, and after leaching: d – MKPC-F; e – MKPC-S; f – MKPC-FS

The authors [7] detected that K leaching occurs mainly by a diffusion-controlled mechanism. This is explained by the fact that part of K did not enter into the reaction of formation of K-struvite and dissolved in the pore water. At the beginning of leaching, K enters the solution due to diffusive transfer (slope = 0.36). Then K dissolves and enters the leachate from the surface of the monolith, mainly from K-struvite. And at the end of the leaching, depletion occurs (slope = 0.09) [7].

Similarly to K, the controlling mechanism for Mg at the beginning of leaching in all studied samples is diffusion (slope = 0.37). Then, over time, surface depletion becomes the main leaching mechanism (see Fig. 4,b).

P shows a slope that appears to be consistent with diffusion (slope ~ 0.4) during the first day of leaching and subsequent depletion until the end of the test (slope close to 0.3). This behavior is typical for all three samples: MKPC-S, MKPC-FS and MKPC-F (see Fig. 4,c).

As X-ray phase analysis data show, on the surface of the MKPC-F sample after leaching, along with K-struvite $MgKPO_4 \cdot 6H_2O$, less soluble quartz SiO_2 and mullite $Al_{(4+2x)}Si_{(2-2x)}O_{(10-x)}$ are present (Fig. 6,a), and on the surface of MKPC-S, MKPC-FS samples – ackermanite $Ca_2MgSi_2O_7$, dolomite $CaMg(CO_3)_2$, and deposited bobierrite $Mg_3(PO_4)_2 \cdot 8H_2O$ (see Fig. 6,b,c). The presence and formation of new poorly soluble phases on the surface of MKPC samples leads to an increase in the water resistance of MKPC.



a *b* *c*

Fig. 6. Diffraction patterns of the sample surface after leaching: a – MKPC-F, b – MKPC-S, c – MKPC-FS

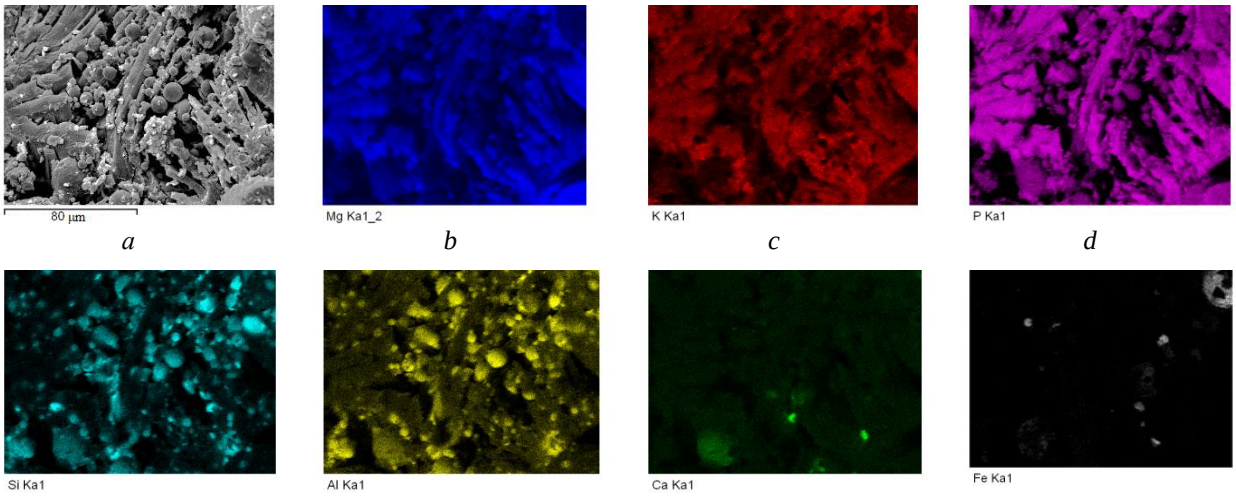
B shows the same leaching mechanisms as K. The slope of the curves for B in all samples during the first 5 days of leaching is close to 0.4; diffusion is the controlling leaching mechanism (see Fig. 4,d). This is followed by dissolution (slope ~ 0.7).

Thus, K and B are the most leachable elements of MKPC materials, the CLF values of which are 30 and 50%, respectively (see Fig. 2,a,d). The increased leaching of these elements is due, firstly, to the presence of some of them in an undissolved form. This part of K and B remains in the pore water and enters the solution through diffusion. Second, the dissolution of the surface of MKPC samples apparently occurs due to the dissolution of readily soluble phases, including boron, whose content is too low to be detected by X-ray diffraction [17].

Si, Al, and Ca are rather low concentrations in the leachate compared to K, Mg, P, and B. Si, Al, and Ca are present in the MKPC material both in the structure of K-struvite and in the composition of unreacted particles of fly ash and blast-furnace slag (Fig. 7). It was shown in [17] that during the preparation of MKPC, a more significant dissolution of BFS particles occurs compared to FA. In this case, some of elements such as

Al, Si, and Ca are entered in the crystal structure of K-struvite, as indicated by the expansion of its crystal lattice parameters.

According to EDS data, in MKPC with the addition of 30% fly ash, the K-struvite reaction product contains a low concentration of Al and Si [22]. A study by Gardner et al [20] states that during the formation of the MKPC binder, the aluminosilicate glassy fractions of fly ash and ground blast furnace slag were dissolved to form a potassium aluminosilicate phase that improved overall compaction and potentially increased the durability of the material.



a *b* *c* *d*

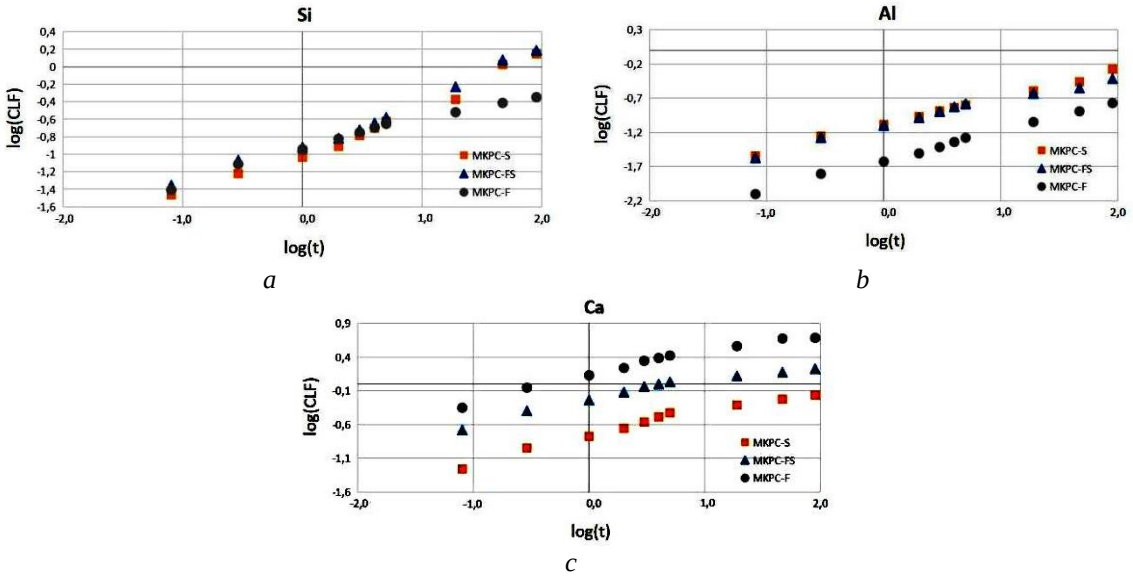
e *f* *g* *h*

Fig. 7. Distribution maps of elements MKPC-F

As can be seen from Fig. 8,a, the slope of the curves for Si is close to 0.5, which confirms the diffusion-controlled leaching process. Si leaching from MKPC-F at a later date corresponds to depletion (slope ~ 0.2).

Al demonstrates the diffusive nature of leaching (slope ~ 0.4) throughout the test in all studied samples (see Fig. 8,b).

At the beginning of leaching, Ca is characterized by a release into solution through diffusion transfer (slope ~ 0.5), and then depletion occurs (slope ~ 0.2) in all MKPC samples (see Fig. 8,c). Possible depletion of Ca at the end of the test procedure is clearly seen in the analysis of curves of cumulative Ca leaching fractions (see Fig. 3,c).



a

b

c

Fig. 8. Log-log plot of cumulative release of: a – Si; b – Al, and c – Ca

Fig. 9 shows the results of calculating the leaching indices for K, Mg, P, and B for samples MKPC-S, MKPC-FS, and MKPC-F. For all samples, the minimum LI values are observed for K and B, and the maximum for Mg. The smallest LI value for K obtained for the MKPC-FS sample is 10.52, for B it is 10.37 (see Fig. 9,b). In reference [7], devoted to the study of the effectiveness of the use of magnesium-potassium phosphate cements for the solidification of nickel-containing wastes, the leaching index for K calculated according to the ANS 16.1 test is slightly lower and amounts to 9.9. As is known, FA and GBFS have a significant impact on the properties of MKPC. Their presence in MKPC paste leads to a decrease in heat generation and a slowdown in cement solidification. FA increases fresh fluidity of the paste, reduces drying shrinkage, improves mechanical strength and water resistance [23].

The highest LI values for all main matrix elements: Mg, K, and P, as well as for B, are observed on the MKPC-S sample. For MKPC-S, the leaching index and effective diffusion coefficient were for: K – 11.00 and $1.8 \cdot 10^{-11}$, Mg – 13.78 and $1.65 \cdot 10^{-15}$, P – 11.99 and $6.89 \cdot 10^{-13}$ cm^2/s, respectively.

The results demonstrate that the use of blast-furnace slag instead of fly ash as a filler makes it possible to improve the corrosion properties of the MKPC matrix, which is expressed in an increase in the value of LI for K by 2 %, for Mg by 8.2 % and for P by 5.6 %. The improvement in corrosion properties is associated with the formation of a denser structure due to the high reactivity of blast-furnace slag particles.

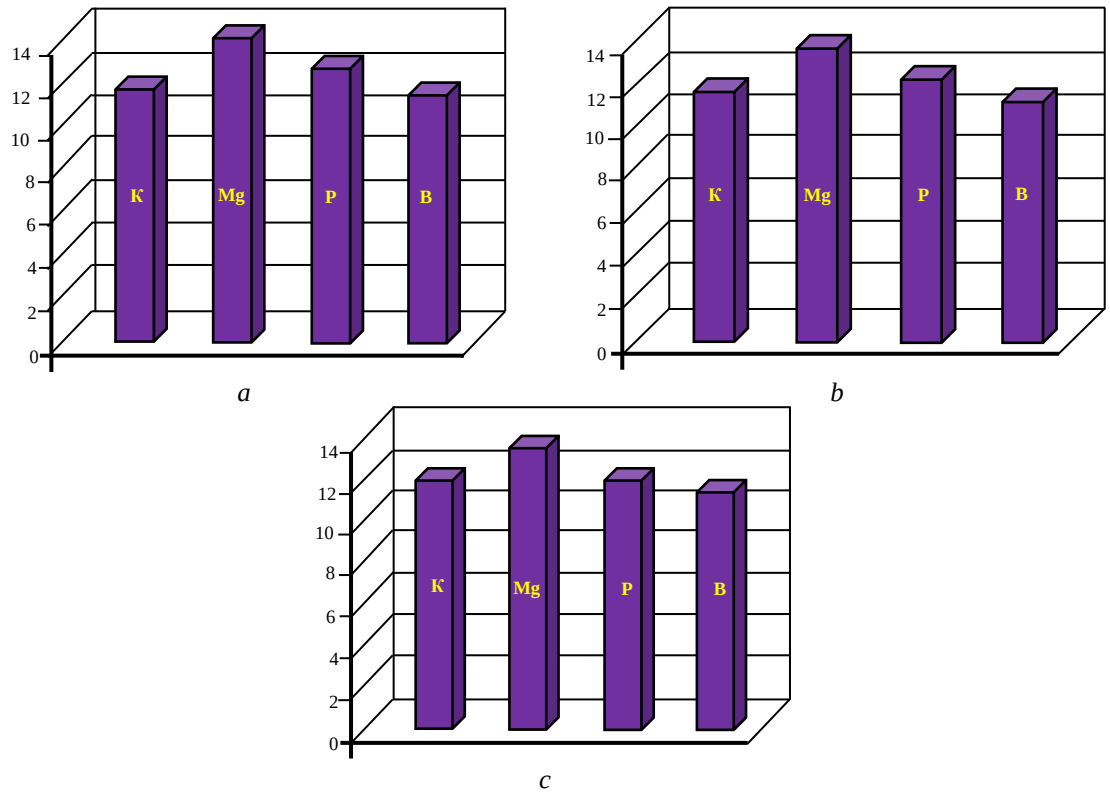


Fig. 9. LI for K, Mg, P and B samples: a – MKPC-S; b – MKPC-FS, and c – MKPC-F

Even higher LI values are observed for all samples for Si, Ca, and Al (Fig. 10). The highest LI value for Si is 15.95, and for Al, 16.98 were obtained on the MKPC-F sample, while the highest values for K, Mg, and P are characteristic of the MKPC-FS sample. This indicates that silicon and aluminum, which are mostly contained in unreacted spherical particles of fly ash, are in a more stable form in relation to leaching. Fig.7 clearly shows the presence of such spherical fly ash particles in the MCPC-F material.

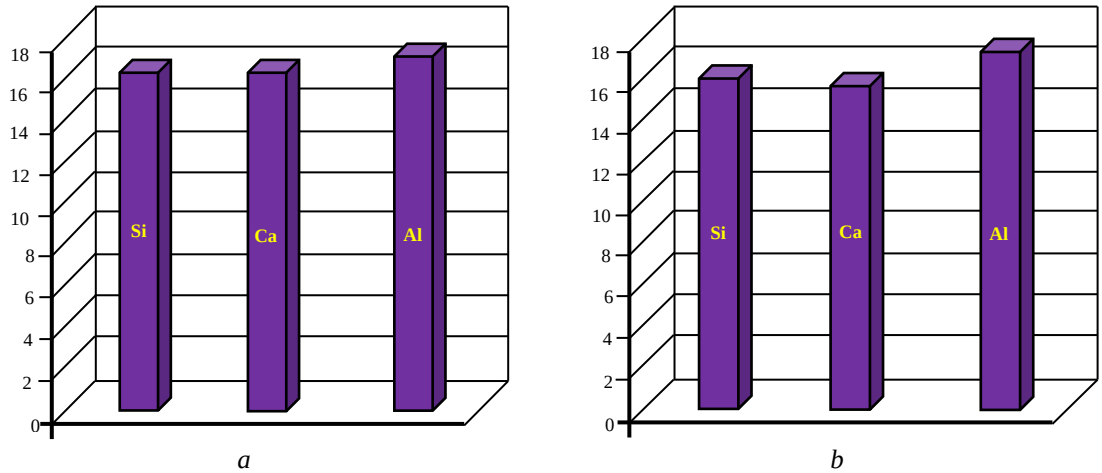
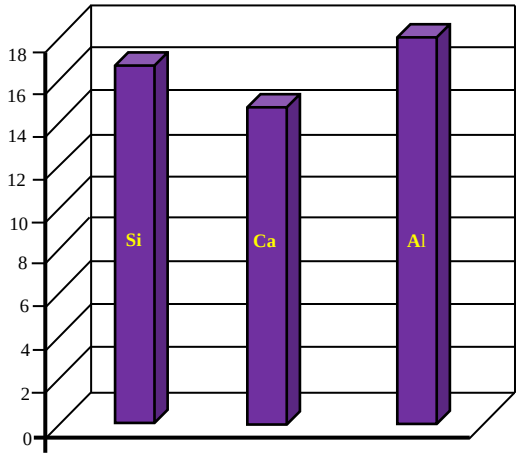


Fig. 10. LI for Si, Ca, and Al samples: a – MKPC-S, b – MKPC-FS



c

Fig. 10. LI for Si, Ca, and Al samples: c – MKPC-F

For Ca, the LI values are slightly lower and amount to 15.54 for the MPKC-S sample, 14.76 for the MKPC-FS sample, and 13.97 for the MKPC-F sample. Apparently, the high LI values for calcium on the MKPC-S and MKPC-FS samples can be explained by the fact that it is part of the more stable phases of ackermanite and dolomite. The presence of akermanite and dolomite, along with K-struvite, quartz and bobierrite, is observed in the surface layer of sample MKPC-S and MKPC-FS after leaching for 1 year (see Fig. 6,b,c).

Thus, as a result of the leaching tests of samples of magnesium-potassium-phosphate cements with various fillers, it was found that the leaching index for all constituent elements is above 10 with a requirement of more than 6. This means that MKPC materials meet the criteria for using them as a matrix for nuclear waste storage at controlled sites.

CONCLUSIONS

Semi-dynamic leaching tests according to ANSI/ANS 16.1 test were carried out on samples of magnesium potassium phosphate cement with fillers such as fly ash and blast furnace slag. Based on the test results, the cumulative leaching fractions, effective diffusion coefficients and leachability indices of the main elements of the MKPC matrix: K, Mg, and P, as well as B, Si, Al, and Ca were calculated. The highest index values for all the main matrix elements: Mg, K, and P, as well as for B, are observed on the MKPC sample with the addition of blast-furnace slag. Leaching mechanisms for various elements were analyzed. Diffusion has been found to be the predominant controlling mechanism. Signs of solubility and depletion were also found. After leaching, changes in the phase composition of the surface layer of the MKPC samples were detected. Despite the resulting high values of leaching indices (> 10) obtained in standardized tests for semi-dynamic leaching in deionized water, further studies are needed on the stability of MKPC materials in leaching processes in various media: surface water, ground water, cement pore water, etc.

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ПОВЕДІНКА МАГНІЙ-КАЛІЙ-ФОСФАТНИХ ЦЕМЕНТІВ ПІД ЧАС ВИЛУГОВУВАННЯ

С.Ю. Саєнко, В.А. Шкуропатенко, Є.О. Світличний, С.О. Карсім, Д.В. Кутній, А.В. Зикова, К.В. Лобач

Ефективність застосування магній-калій-фосфатних цементів (МКФЦ) як захисних матриць радіоактивних відходів була досліджена у даній роботі з точки зору вилуговування основних елементів. Як наповнювачі МКФЦ використовували золу виносу та доменний шлак. Випробування на вилуговування проводили на монолітних МКФЦ-зразках у відповідності до тесту ANSI/ANS 16.1. На підставі отриманих даних визначені кумулятивні фракції вилуговування, механізми вилуговування, ефективні коефіцієнти дифузії та індекси вилуговування K, Mg, P, B, Si, Al та Ca. Значення розрахованих індексів вилуговування (> 10) складових елементів вказують на те, що характеристики досліджуваної МКФЦ-матриці прийнятні для іммобілізації радіоактивних відходів.