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Sorption of Np, Pu, Am, Sr, Cs on the Mineral Phases of the Rocks of the Nizhnekansky Granitoid Massif under Conditions of Underground Repositories for Radioactive Wastes

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Abstract—The sorption properties of mineral phases that constitute the rocks of the Yeniseysky area of the Nizhnekansky granitoid massif toward Np, Pu, Am, Sr, and Cs were studied under conditions of underground radioactive waste disposal. As a result of sorption experiments, sorption kinetic curves were obtained, and the distribution coefficients ($\underline{K}_d$) of radionuclides on mineral phases were determined. The resulting data made it possible to evaluate the differences in the sorption properties of minerals towards the studied radionuclides. For each radionuclide, with the exception of americium, it was possible to establish the most effective mineral phases in terms of sorption: for Np(V)—hydroxyapatite and calcite, for Pu(V)—hydroxyapatite and magnetite, for Sr(II)—biotite and hydroxyapatite, for Cs(I)—biotite and muscovite. Americium was sorbed quantitatively on all mineral phases, except for biotite and quartz.

Keywords: sorption, radionuclides, mineral phases, distribution coefficients, cesium, strontium, americium, plutonium, neptunium, exocontact of the Nizhnekansky granitoid massif.

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INTRODUCTION

At present, in Russia, as in most countries of the world, the problem of disposal of high-level and intermediate-level waste containing long-lived radionuclides into deep geological formations is being solved [1–3]. The prospect of radioactive waste disposal in geological structures represented by crystalline rocks has been considered by Russian scientists for 30 years already [4–6]. At the same time, the main attention was focused on the study of large granitoid bodies and sequences of Precambrian metamorphic rocks of the South Yenisei Ridge in the Krasnoyarsk Territory. According to the results of preliminary surveys, among the three areas under consideration: Itatsky, Kamenny, Yeniseysky, the Yeniseysky area, located in the exocontact zone of the Nizhnekansky granitoid massif (NGM), was chosen as the most promising area for the repository. To justify the safety of the repository, the first step is the creation of an underground research laboratory, where, along with geofiltration studies of rock properties, it is planned to study the migration of radionuclides under conditions of underground burial [7, 8]. The key processes that control the migration of radionuclides are the processes of sorption/desorption on host rocks. The rocks of the NGM of the Yeniseysky area are plagio- and granite-gneisses, which are intruded by dike complexes of dolerites and gabbro-diabases, and are also cut by quartz and carbonate veinlets, which indicates the heterogeneity of these rocks [9, 10].

The study of the sorption properties of multimineral crystalline rocks, which are also characterized by an inhomogeneous surface topography, is a rather difficult task. In this regard, in recent years, researchers

Table 1. Literature data on the sorption of Np, Pu, Am, Sr, Cs in the concentration range 10^{-6}–10^{-11} M on various mineral phases under conditions of aqueous solutions with pH 7–8, $I = 0.01$–0.1

Minerals	log K_d [mL/g]					Sources
	Np(IV/V)	Pu(IV/V)	Am(III)	Sr(II)	Cs(I)	
Tectosilicates						
L/S	25–5000	40–2500	70–2000	20–2000	20–2000	
Quartz	0.5–1	3	2.7–3	1.2	0–2	[17–23]
K-feldspar	1.5	–	3.8	–	0.7–2.3	[22–25]
Na-feldspar	0–1.3	–	3.6	–	1.4–2.3	[18, 22–24, 26]
Zeolite (synthetic)	–	2–3	4–5	3.8–4.5	5.3	[27–30]
Clinoptilolite	1	3	–	3.8	4	[17, 27, 29, 31]
Chabazite	–	–	–	2.9	–	[32]
Phyllosilicates						
Biotite	1.3–1.9	5	4.3	4	2.8–3.8	[18, 22, 23, 26, 33]
Vermiculite	–	–	–	4.3	5.3	[30]
Chlorite	1–2	–	4.3	–	2–3	[18, 23, 34]
Illite	2–3	5.4	5–5.5	2.2	2.6–3.5	[21, 23, 35–40]
Phosphates						
Apatite/hydroxyapatite (HAp)	3–4	5.5	4	1.6	2	[18, 30, 41, 42]
Carbonates						
Calcite	2.5–2.7	1.3–3[a]	4	1.3	1	[18, 23, 43–45]
Fe-bearing minerals						
Magnetite	0–2.8	4–5	4	1.5	1.5	[18, 46–49]

[a] K_d values vary depending on the plutonium oxidation state.

have increasingly used to an integrated approach to studying the behavior of radionuclides in a medium of crystalline rocks, which makes it possible to establish the contribution of mineral grains to the retention of radionuclides in view of surface inhomogeneity [11, 12].

The sorption properties of the crystalline rocks of the Yeniseysky area were studied in [13–16]. In addition to determining the main sorption parameters (distribution coefficients, sorption degrees) obtained from bulk sorption experiments, the authors studied the microdistribution of radionuclides on the surface of solid rock samples using digital radiography. As a result of the studies, the authors note that the sorption of radionuclides is extremely uneven and the mineral composition has a significant effect on the nature of the distribution of radionuclides on the surface of rock samples. For example, layered aluminosilicates (biotite, muscovite, etc.) make a significant contribution to the sorption of cesium [15]. The least efficient sorbents for Cs, Ra, Am, and Pu are quartz and feldspars [13]. Biotite has the highest efficiency toward Am sorption, which was quantitatively confirmed by crushing a whole sample and measuring each mineral fraction by γ-spectrometry [5]. Thus, the sorption of a radionuclide can be controlled by a certain mineral or a group of minerals of the same class present in the rock.

The sorption properties of various minerals toward radionuclides have been considered in many studies. Table 1 shows the distribution coefficients (K_d) for minerals that can be included in the composition of the NGM rocks of the Yeniseysky area.

However, it is known that the K_d values are strongly affected by the experimental conditions, including the pH values of the solutions, the concentration of the element in the solution, the liquid-to-solid (L/S) phase ratio, the specific surface area of the sorbents, and their structure. Thus, based only on the K_d values obtained from various overall sorption experiments, it is difficult to analyze the

Table 2. Specific surface area of samples of mineral phases used for sorption experiments (grain size <0.1 mm)

Classification	Mineral name	Formula	Surface area, m^2/g
Framework silicates	Quartz	SiO_2	0.4
Layered silicates	Muscovite	$KAl_2[AlSi_3O_{10}](OH)_2$	12.3
		$K(Mg,Fe)_3[Si_3AlO_{10}][OH,F]_2$	4.9
Carbonates	Biotite	$CaCO_3$	0.2
Phosphates	Calcite	$Ca_{10}(PO_4)_6(OH)_2$	67.1
Oxides (Fe-containing minerals)	Hydroxyapatite (HAp)	$FeO \cdot Fe_2O_3$	1.3
	Magnetite		

competitive sorption capacity of mineral phases under specific underground disposal conditions. For solving this problem, a method was proposed for determining the quantitative parameter of the relative sorption efficiency (RSE) of radionuclides on the mineral phases of solid rock samples [50, 51]. The contribution of each mineral of the heterogeneous system to the retention of U, Np, Pu, Am, Ra, Cs was determined based on RSE data, and the patterns of radionuclide sorption on certain mineral phases of various samples of the Yeniseysky area were revealed. To confirm the derived patterns of radionuclide sorption in heterogeneous systems and to obtain the K_d parameters for further modeling of the migration behavior of radionuclides, it is necessary to consider monomineral systems under the conditions of previously analyzed whole rock samples.

This work is aimed at studying the sorption properties of mineral phases that are part of the rocks of the NGM exocontact zone of the Yeniseysky area towards Np, Pu, Am, Sr, Cs in order to obtain K_d parameters and establish patterns of radionuclide sorption on the considered minerals.

EXPERIMENTAL

For conducting overall sorption experiments, we considered mineral phases that are part of the rocks of the Yeniseysky area of the NGM exocontact zone and previously established in [10, 13]. Since the isolation of mineral phases directly from the studied rock samples of the Yeniseysky area is technically difficult, we have chosen analogs of the mineral phases that compose the rocks. In total, six mineral phases were considered, some of them were synthesized (quartz, hydroxyapatite (HAp), magnetite), the rest (muscovite, biotite, calcite) were of natural origin. To fabricate powders, the selected minerals were ground in a ball mill, and a fraction with a grain size of less than 0.1 mm was isolated. Characteristics of mineral phases in accordance with their classification, as well as the values of the specific surface area of powders of mineral phases, determined by the BET method, are given in Table 2.

Sorption experiments were carried out at room temperature (22 ± 2°C) in a plastic dish in an air atmosphere. A model groundwater solution was utilized as the liquid phase. The solution was prepared by adding bentonite to distilled water in a ratio of 1 g/L (bentonite from the 10th Khutor deposit, Khakassia, Russia). Equilibrium was reached within a week, then the water was separated from the sediment by centrifugation (15 000 rpm) during 20 min. The concentration of the main components in the model solution (mg/L) was as follows: Ca^{2+} 2.4, Mg^{2+} 1.9, Na^{+} 16.9, Al^{3+} 6.8; Si^{4+} 19.5. In accordance with these concentrations, the ionic strength of the solution was 0.01 M. Then, the radionuclides $^{237,239}Np$, $^{239,240}Pu$, ^{241}Am, $^{90}Sr/^{90}Y$, and ^{137}Cs were added separately to the prepared model solutions with an initial concentration in the solution of 10^{-9} M. The initial specific activity of solutions (A, Bq/L) labeled with ^{239}Np, ^{240}Pu, ^{241}Am, $^{90}Sr/^{90}Y$, and ^{137}Cs was 1.3×10^6, 2.2×10^3, 3.3×10^4, 4.5×10^5, and 3×10^5, respectively.

The pH value of the solutions was maintained in the range of 7–8 by adding NaOH and HCl solutions. The experiments were carried out at L/S ratios (volume of the liquid phase of the solution/weight of the solid phase of the sorbent) of 100 and 5000 mL/g. The attainment of equilibrium was monitored by periodic sampling of 100–500 μL aliquots from the solutions. In the case of Np, aliquots were taken within a week because of the short half-life of ^{239}Np. In other cases, the time of sampling varied from a day to a month/three months as equilibrium was reached. The activity of $^{90}Sr/^{90}Y$

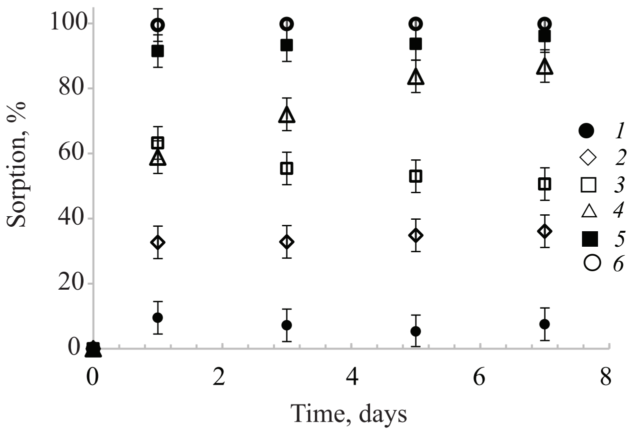

Fig. 1. Sorption of Np(IV/V) the mineral phases: (*1*) quartz, (*2*) muscovite, (*3*) biotite, (*4*) magnetite, (*5*) calcite, (*6*) hydroxyapatite, L/S (mL/g) = 100, model groundwater solution, pH 7–8, $I = 0.01$ M.

aliquots was measured after 20 days to establish an equilibrium between ^{90}Sr and ^{90}Y. The content of radionuclides in solutions was determined by liquid scintillation spectrometry (Tri-Carb 2810 TR, Canberra and Quantulus 1220, PerkinElmer).

The recovery (R, %) of radionuclides was calculated based on the following ratio:

$$R = \frac{I_0 - I_\mathrm{t}}{I_0},$$

where I_0 is the count rate of the radionuclide in the initial solution (pulses/min); I_t, is the count rate of the radionuclide in the solution at time t (pulses/min).

For each mineral phase, the K_d (mL/g) of the studied radionuclides were determined based on the following ratio:

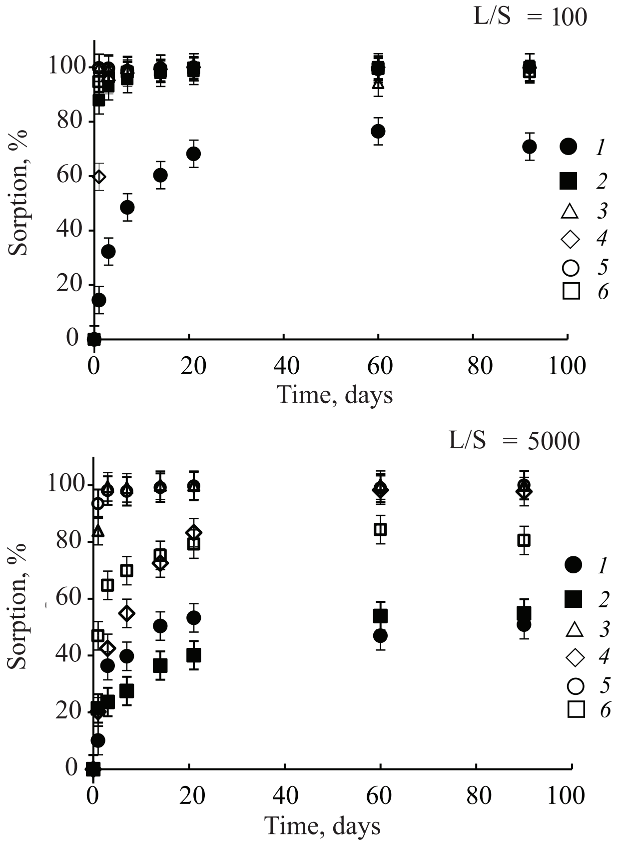

Fig. 2. Sorption of Pu(IV/V) at various L/S ratios on mineral phases: (*1*) quartz, (*2*) calcite, (*3*) magnetite, (*4*) muscovite, (*5*) hydroxyapatite, (*6*) biotite. Model groundwater solution, pH 7–8, $I = 0.01$ M.

$$K_\mathrm{d} = \frac{I_0 - I_\infty}{I_\infty} \times \frac{V}{m},$$

where I_0 is the count rate of the radionuclide in the initial solution (pulses/min); I_∞ radionuclide count rate at the

Table 3. Distribution coefficients of radionuclides on mineral phases in model groundwater solutions with pH 7–8, $I = 0.01$ M, radionuclide concentration 10^{-9} M

Minerals	log K_d [mL/g]								
	L/S = 100					L/S = 5000			
	Np	Pu	Am	Sr	Cs	Pu	Am	Sr	Cs
Quartz	0.9	2.3	3.3	1.6	1.9	3	3.1	3.4	4
Muscovite	1.7	>6	6	3.8	3.4	5	4.7	3.7	4.6
Biotite	2.1	4.5	6	2.2	4	4.1	4	3.5	5.3
Calcite	3.1	3.8	6	1.2	0	3.1	5	2.8	4
Hydroxyapatite	3.7	>6	5.1	3.8	0.8	6	5.2	3.8	4
Magnetite	2.4	>6	6	2	2.1	6	5	3.4	3.8

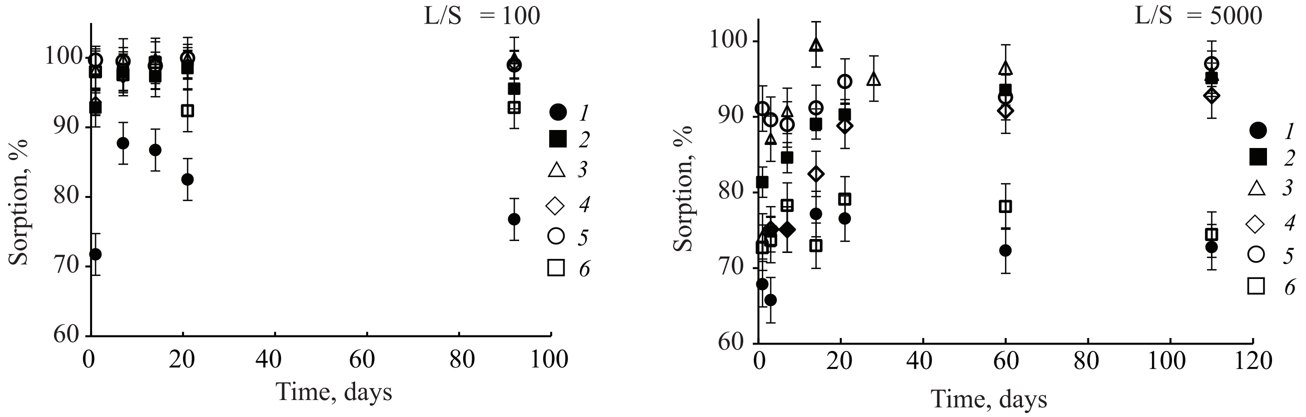

Fig. 3. Sorption of Am(III) at various L/S ratios on mineral phases: (*1*) quartz, (*2*) calcite, (*3*) magnetite, (*4*) muscovite, (*5*) hydroxyapatite, (*6*) biotite. Model groundwater solution, pH 7–8, $I = 0.01$ M.

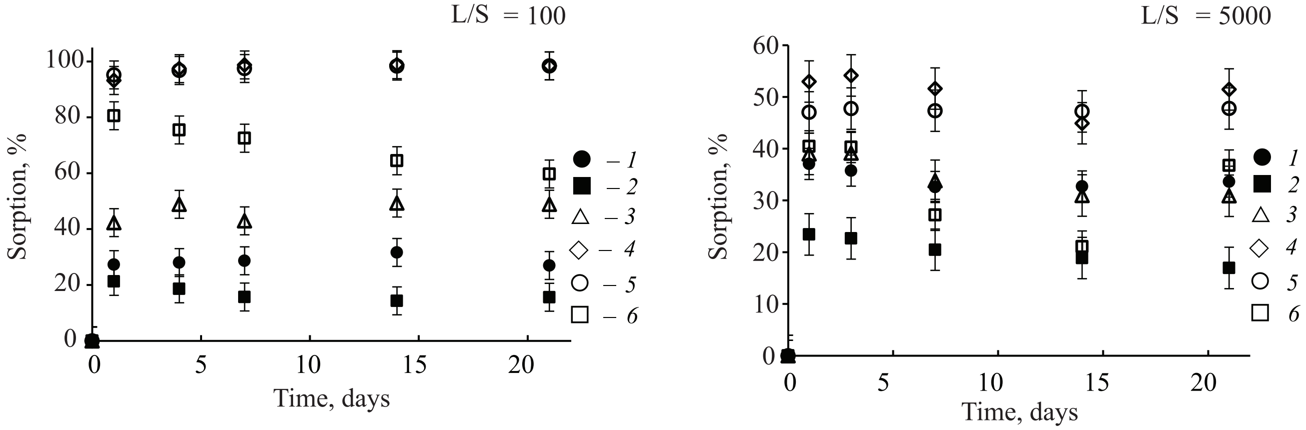

Fig. 4. Sorption of Sr(II) at various L/S ratios on mineral phases: (*1*) quartz, (*2*) calcite, (*3*) magnetite, (*4*) muscovite, (*5*) hydroxyapatite, (*6*) biotite. Model groundwater solution, pH 7–8, $I = 0.01$ M.

system equilibrium (pulses/min); V is the volume of the solution (mL); m is the weight of the mineral sample in contact with the solution (g).

RESULTS AND DISCUSSION

Kinetic curves of sorption of Np(IV/V), Pu(IV/V), Am(III), Sr(II), and Cs(I) radionuclides were plotted according to data of the sorption experiment on mineral powders (Figs. 1–5). The resulting distribution coefficients are listed in Table 3. The distribution coefficients of Np(IV/V), Pu(IV/V), Am(III), Sr(II), Cs(I) on mineral phases based on the literature data (Table 1) are compared with the values resulted in sorption experiments, if possible at close L/S ratios.

At studying the sorption kinetics of neptunium, for which the characteristic oxidation state under preset conditions is +5, it was noted that for almost all minerals, equilibrium is reached within a week, with the exception of magnetite and biotite. This behavior may be due to the slow reduction of neptunium in the presence of Fe(II)-bearing mineral phases [52]. At the same time, in [53], the authors point out that Np(V) is reduced not in the liquid phase, but on the surface of the mineral. Based on the data of neptunium sorption kinetic curves (Fig. 1), different sorption efficiency of mineral phases was established. According to the neptunium sorption efficiency, the mineral phases are arranged in the following order: hydroxyapatite > calcite > magnetite > biotite > muscovite > quartz with the corresponding K_d values, mL/g: 5.4×10^4, 1.4×10^3, 258, 125, 49, 8. High sorption capacity of minerals of the phosphate (HAp) and carbonate (calcite) groups toward neptunium is explained by the formation of complexes of neptunyl ions (NpO_2^+) with PO_4^{3-} and CO_3^{2-} anions on the surface of these minerals [41, 54]. When comparing the K_d values of neptunium on minerals from sorption ex-

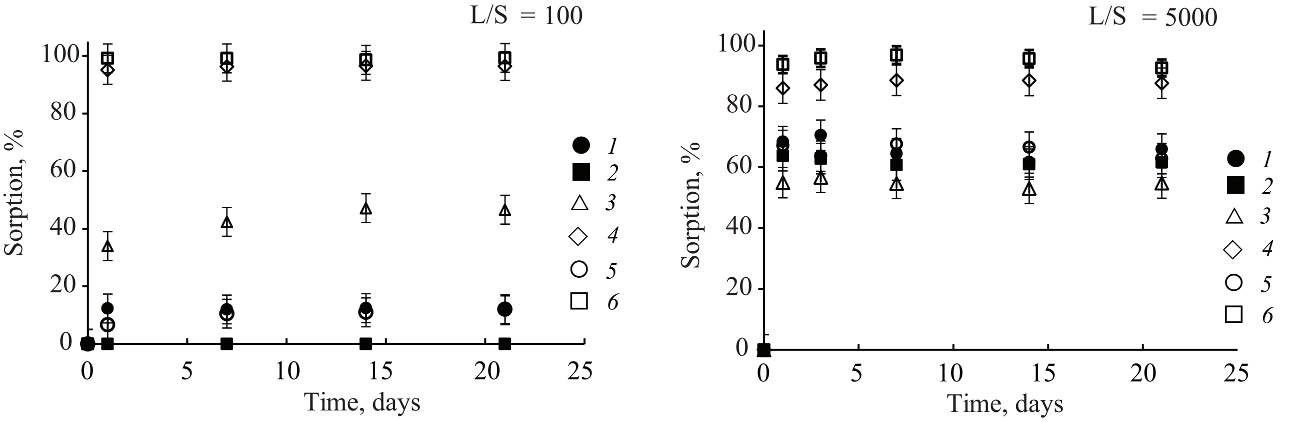

Fig. 5. Sorption of Cs(I) at various L/S ratios on mineral phases: (*1*) quartz, (*2*) calcite, (*3*) magnetite, (*4*) muscovite, (*5*) hydroxyapatite, (*6*) biotite. Model groundwater solution, pH 7–8, $I = 0.01$ M.

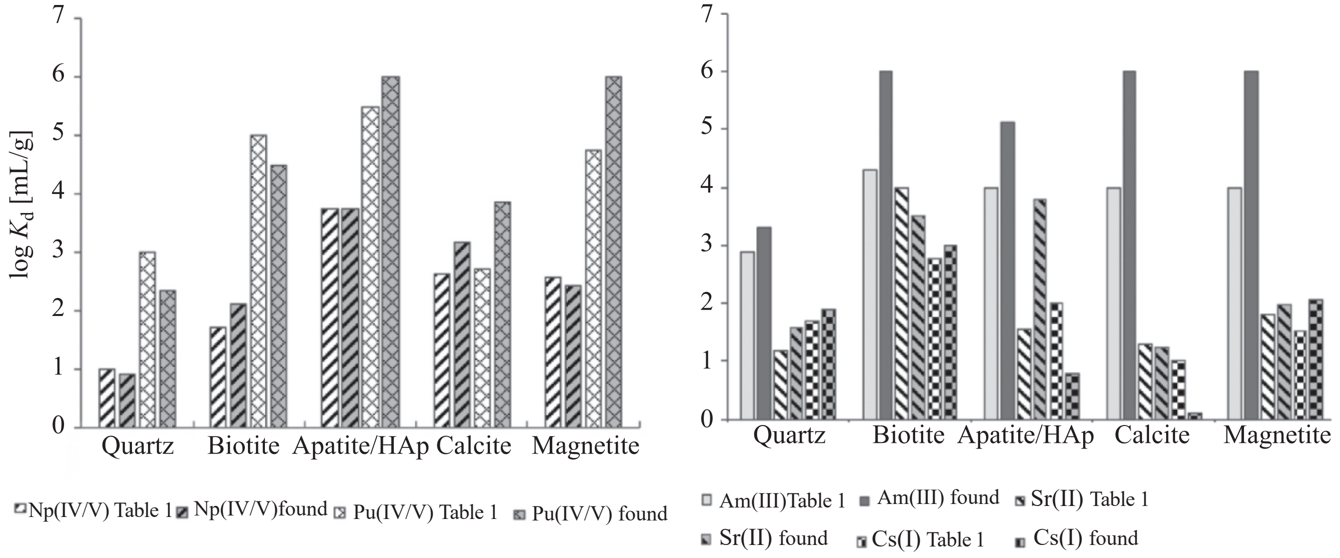

Fig. 6. Comparison of the obtained distribution coefficients of Np(IV/V), Pu(IV/V), Am(III), Sr(II), Cs(I) on mineral phases (Table 3) at a L/S ratio of 100 with the literature values (Table 1).

periments with the K_d values from the literature data (Fig. 6), the convergence of the values within one order of magnitude for quartz and magnetite is noted; in the case of apatite, the values practically coincide. It is also worth emphasizing that the obtained data on K_d from monomineral sorption experiments are consistent with the previously obtained data on the relative sorption efficiency (RSE) of neptunium on minerals of whole samples from [51], where the phosphate group mineral monazite is the most effective phase toward neptunium (RSE = 2.3), and biotite has average sorption properties with a RSE value of 1.4.

In the case of Pu and Am (Figs. 2, 3), at a L/S ratio of 100, rapid and quantitative sorption was observed for almost all minerals. An exception is the quartz phase, which was less efficient toward these nuclides, while in the system with Am, sorption equilibrium was not reached even for 90 days. Thus, based on the data obtained at a L/S ratio of 100, it is difficult to draw a conclusion about the difference in sorption properties. An increase in the L/S ratio to 5000 made it possible to find out some differences in the behavior of Pu and Am toward various minerals.

At an L/S ratio of 5000 (Fig. 2), a difference was noted in the Pu sorption kinetics on layered aluminosilicates, biotite and muscovite. It follows from the data obtained that, at first, sorption on biotite is faster than on muscovite, then, as equilibrium is reached, the muscovite sorption capacity toward Pu becomes higher than that of biotite, with K_d values equal to 10^5 and 10^4 mL/g, respectively (Table 3). Hydroxyapatite and magnetite were the predominant phases of Pu sorption, for which K_d values of about 10^6 mL/g were detected. Quartz and calcite have the lowest sorption capacity ($K_d = 10^3$ mL/g). It should be noted that initially, during preparing aqueous solutions, plutonium was in the +5 oxidation state. However, it is known that interaction of Pu(V) with minerals results in its reduction to Pu(IV) on the surface of solid phases [55, 56]. Thus, Pu can have two oxidation states in the system. On the whole, the values of K_d for Pu obtained from monomineral experiments agree both with the literature data (Fig. 6) and with the RSE data from [57], where it was shown that the most effective phases toward Pu are Fe-bearing minerals (magnetite, ilmenite) and phosphates (apatite). In the case of layered minerals, a slight difference was noted; according to RSE data [57], muscovite and biotite have average sorption properties (RSE = 1.4–1.5) toward Pu, while the K_d values for biotite in monomineral experiments (Fig. 6) can reach 10^4–10^5 mL/g, which indicates a high sorption efficiency, comparable with the efficiencies for phosphates (apatite/HAp) and the Fe-bearing mineral magnetite.

For Am, an increase in the L/S ratio to 5000 is not indicative (Fig. 3), since in this case, the quantitative sorption ($K_d \sim 10^5$) of americium is characteristic of all mineral phases, with the exception of biotite and quartz with K_d of 10^4 and 10^3 mL/g, respectively (Table 3). Based on the literature data (Table 1), it is also difficult to draw a conclusion about the most effective mineral phases that absorb Am, since this radionuclide is equally retained on almost all mineral groups, with the exception of framework silicates (quartz, feldspars). At the same time, when studying the microdistribution of Am over mineral phases, it was found that the predominant phases that retain Am are magnetite and apatite with RSE of 2 [51, 57].

When studying the sorption of Sr in systems with different L/S ratios, discrepancies in the sorption properties of minerals are observed (Fig. 4). The Sr sorption on quartz at the L/S ratio of 5000 increases sharply compared to sorption at the L/S ratio of 100. It should be noted that for the most sorption-efficient mineral phases, biotite and hydroxyapatite, the K_d values do not change vs. the L/S ratio and are about 10^3 mL/g (Table 3). The high sorption efficiency of these minerals is explained by sorption in accordance with the ion exchange mechanism characteristic of Sr, although we do not rule out the possibility of complex formation on the surface [58, 59]. For the rest of the mineral phases, the K_d values increased significantly at the L/S 5000 compared to the L/S 100, which led to similar K_d values in the case of biotite, quartz, and magnetite (Table 3). Thus, in the case of Sr, the difference in the sorption properties of mineral phases is more clearly observed at Lthe /S ratio of 100. Also, when comparing the K_d for Sr values obtained from monomineral experiments at the L/S ratio of 100 with the literature data (Fig. 6), the K_d values practically for all considered mineral phases coincide within one order of magnitude, with the exception of phosphate (apatite/HAp).

For Cs (Fig. 5), as for Sr, there is a difference in the sorption properties of mineral phases at the L/S ratios of 100 and 5000. It can be seen that all values of the sorption at the L/S 5000 increased, especially for quartz, hydroxyapatite, calcite, and magnetite, which at the L/S ratio of 100 demonstrated low sorption. However, despite the difference in the results at different L/S ratios, for cesium, layered aluminosilicates muscovite and biotite with K_d values of 10^3–10^4 mL/g (Table 3) are the predominant sorption phases in both cases (Table 3), which also is confirmed by the data on RSE values [51]. These minerals have a crystallographic layered structure with ion-exchange centers that are directly involved in cesium sorption [60]. The fairly rapid attainment of equilibrium in the system also indicates the ion exchange mechanism (Fig. 5). As in the case with Sr, for Cs at the L/S ratio of 100, the K_d values from monomineral experiments converge with the literature values (Fig. 6) for almost all considered mineral phases. The exception is the phosphate phase, apatite/HAp, for which the K_d values differ by an order of magnitude, which is more likely due to the different nature of the studied phosphates. In our studies, we examined synthetic HAp, while in the literature a natural mineral is considered [30].

CONCLUSIONS

Comparative analysis of the sorption properties of minerals that are part of the rocks of the Yeniseysky area of NGM under underground disposal conditions was carried out based on the obtained data on the recovery and distribution coefficients of Np, Pu, Am, Sr, Cs on mineral phases. For each radionuclide, with the exception of Am, it was possible to find out the most effective mineral phases: for Np(V)—hydroxyapatite and calcite, for Pu(V) hydroxyapatite and magnetite, for Sr(II)—biotite and hydroxyapatite, for Cs(I)—biotite and muscovite. Americium was sorbed quantitatively on all mineral phases at various L/S ratios (100 and 5000 mL/g), with the exception of biotite and quartz; therefore, it was not possible to establish the predominant phases in this case. For Pu, an increase in the L/T ratio to 5000 made it possible to establish a difference in the sorption properties of minerals, which may be due to the different sorption capacity of the investigated mineral phases. The distribution coefficients of the studied radionuclides we obtained are also in good agreement with the previously obtained data on the RSE of mineral of whole rock samples.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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