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## Preliminary results on the Co, Sr and Cs sorption properties of the analcime-containing rock type of the Boda Siltstone Formation

Péter Sipos, Tibor Németh  
*Institute for Geochemical Research,  
Hungarian Academy of Sciences, Budapest*

Zoltán Máthé  
*Mecsekérc Zrt., Pécs*

The search for nuclear waste deposits have been focusing on formations containing large amounts of clay minerals, as they potentially possess significant isolation capacity due to their radionuclide sorption ability. In this study the immobilization processes for Co, Sr and Cs ions on the recently described analcime-containing rock type of the Boda Siltstone Formation were investigated by batch sorption experiments. Our data suggest a high radionuclide immobilization capacity for this rock formation in the following order: Sr > Co > Cs. The dominant immobilization processes were found to be ion exchange and precipitation for Co and Sr, and chemisorption for Cs. The necessity of further sorption analyses on the studied rock formation is also raised.

Key words: radionuclides, sorption capacity, claystone, ion exchange, precipitation, sorption curves, radioactive waste deposition

### *Introduction*

One of the most important requirements for rock bodies used as a natural geologic barrier in nuclear waste deposition is the appropriate isolation capacity. Consequently, the possible highest radionuclide immobilizing capacity is a fundamental requirement for these rock formations. That is why searches for nuclear waste deposits have been focused on rock formations with high clay mineral content. In Hungary, a thick Upper Permian siltstone formation, the Boda Siltstone Formation (BSF) has been selected as a possible repository of high-level nuclear waste (Árkai et al. 2000; Kovács et al. 2000).

Addresses: P. Sipos, T. Németh: H-1112 Budapest, Budaörsi út 45, Hungary  
e-mail: sipos@geochem.hu

Z. Máthé: H-7633 Pécs, Esztergár Lajos u. 19, Hungary

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The Upper Permian sedimentary sequence of the BSF is situated within an area of 150 km<sup>2</sup> in the Western Mecsek Mountains, SW Hungary. Its 15 km<sup>2</sup> surface outcrop can be found in the perianticlinal structure of the Western Mecsek Mts, while in the subsurface it was only encountered in the Ib-4 borehole in the Gorica Block (Fig. 1). Its maximum thickness is 700–900 m, but it is only 350 m in the Gorica block. The BSF was deposited in a shallow-water lacustrine environment (playa mudflat, playa lake), under semi-arid to arid climatic conditions. Its red or reddish-brown color reflects the predominance of the oxidizing nature of the depositional and early diagenetic environments (Jámbor 1964; R. Varga et al. 2005). Five rock types of the BSF can be distinguished based on mineralogical, geochemical and textural considerations: albitic claystone, albitolite, "true" siltstone, dolomite interbeds and sandstone (Barabás and Barabás-Stuhl 1998; Árkai et al. 2000). Their main mineral components are illite-muscovite, chlorite, authigenic albite, quartz, carbonate minerals (calcite and dolomite) and hematite (Máthé 1998; R. Varga et al. 2006). However, the BSF encountered in borehole Ib-4 differs in its mineralogical composition, as it contains abundant analcime (up to 25%) in addition to the above minerals.

The albitic claystone variant of the BSF had already been studied for its radionuclide sorption capacity and ion diffusion characteristics (Mell et al. 2006a, b). However, there are no data on the cationic radionuclide sorption properties of the analcime-containing rock type of the BSF, which was only recently discovered by members of the Institute for Geochemical Research. In this rock type the analcime is a rock-forming zeolite mineral, which can be considered to fairly retain migrating cations and radionuclides (Redkin and Hemley 2000). Diffusion rates of anionic species (<sup>99</sup>TcO<sub>4</sub><sup>-</sup>, H<sup>14</sup>CO<sub>3</sub><sup>-</sup>) and HTO were already determined on ore samples of this rock type by Lázár et al. (2008). They found similar diffusion and retardation parameters for anionic species in their samples to those of the albitic claystone of the BSF.

For a better understanding of the geochemical processes taking place around a nuclear waste deposition site, there is an essential need to have experimental data on the ion sorption properties of the depository rock. Our aim was to study the immobilization processes for Co, Sr and Cs ions in a representative sample of the claystone containing analcime. According to the radionuclide composition of the processed nuclear waste from the nuclear power plant in Paks, Hungary, <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>60</sup>Co are among the most important components in respect of radiation protection (Bárdossy 1998).

### ***Materials and Methods***

Batch sorption experiments were carried out on a representative, fresh, red-colored claystone sample containing analcime from borehole Ib-4 near the village of Ibafa (Fig. 1). Its mineralogical and chemical composition is presented in Table 1. The chemical composition of the rock sample was analyzed by atomic

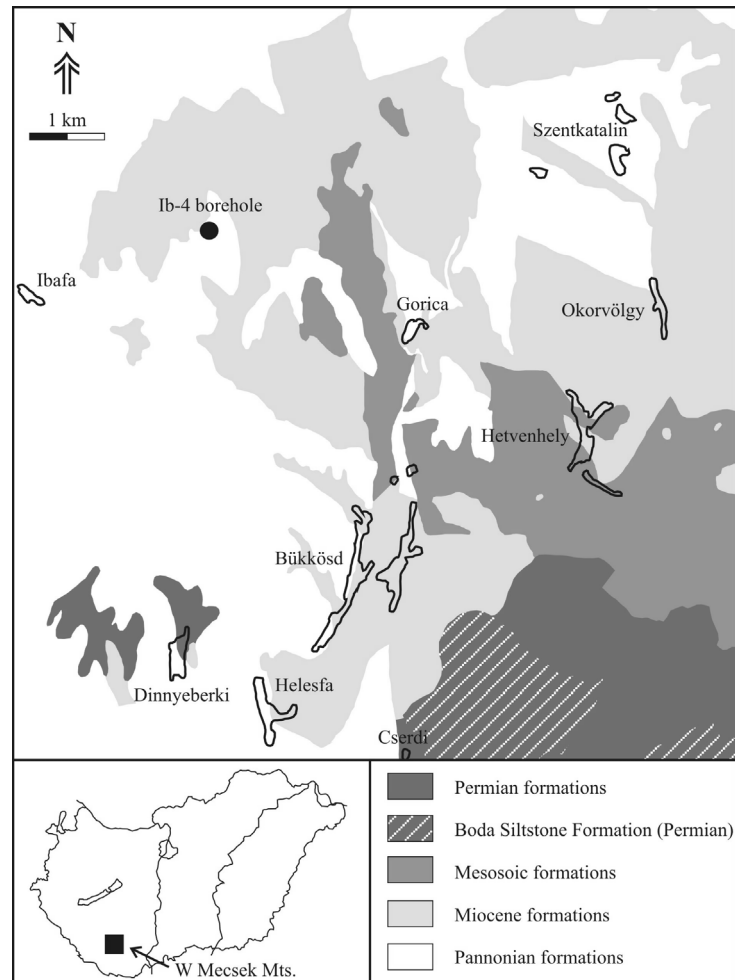


Fig. 1  
Geologic sketch map of the Western Mecsek Mts with the location of borehole Ib-4

absorption spectrometry (Perkin Elmer AAnalyst 300) after Li-metaborate digestion. Mineral composition of the bulk rock was determined on random-powdered samples by semi-quantitative X-ray diffractometric (XRD) (Philips PW 1710) phase analysis after the method of Bárdossy et al. (1980). Clay minerals were identified from the clay fractions of bulk samples by XRD diagrams obtained from parallel-oriented specimens. The following diagnostic treatments were carried out for all of the samples: ethylene glycol solvation at 60 °C overnight, Mg saturation followed by glycerol solvation at 95 °C overnight, K saturation, heating at 350 and 550 °C for 2 hours. Separation of the clay fractions

Table 1  
Chemical and mineralogical composition of the studied sample

Sample	Chemical composition (%)		Semi-quantitative mineralogical composition (%)	
Ib-4/553 m	SiO <sub>2</sub>	51.24 ± 1.14	Illite	35
	Al <sub>2</sub> O <sub>3</sub>	16.28 ± 0.51	Analcime	18
	MgO	3.63 ± 0.17	Quartz	12
	Fe <sub>2</sub> O <sub>3</sub>	7.25 ± 0.02	Albite	12
	K <sub>2</sub> O	4.06 ± 0.04	Calcite	11
	CaO	3.67 ± 0.02	Hematite	6
	Na <sub>2</sub> O	3.08 ± 0.12	Chlorite/Smectite	2
	P <sub>2</sub> O <sub>5</sub>	0.20 ± 0.001	Chlorite	2
	MnO	0.11 ± 0.001	Dolomite	2
	TiO <sub>2</sub>	0.78 ± 0.005	Mineralogy of the clay fraction: Illite >	
	LOI	9.15 ± 0.55	Illite/Smectite > Chlorite/Smectite > Chlorite >>	
	Sum	99.45 ± 2.58	Analcime = Calcite = Hematite > Albite	

was performed by sedimentation in aqueous suspension. The sample was grounded in an agate mortar to a powder with particles smaller than 0.63 mm before the experiments. According to our microscopic observations particles larger than 0.5 mm can be found only sporadically in the studied rock sample.

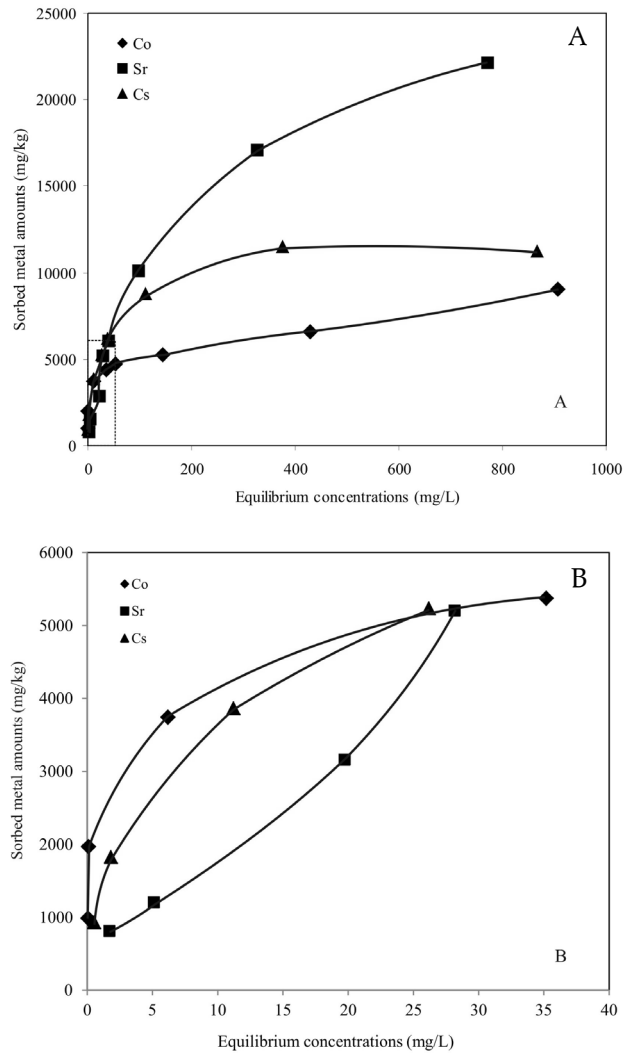
The single element sorption experiments were carried out in polypropylene centrifuge tubes of 50 ml by mixing 400 mg of the studied sample with 40 ml of nitrate solution containing various concentrations (10, 20, 50, 80, 100, 200, 500 and 1000 mg/l) of Co, Sr and Cs in parallel sets. The samples were shaken lengthwise for 48 hours at 25°C, and then they were centrifuged at 3000 rpm for 20 minutes. Element concentrations in the equilibrium solutions were analyzed by atomic absorption spectrometry. Sodium, K and Ca concentrations were also analyzed in the equilibrium solutions to study the role of ion exchange in the sorption process. The average relative standard deviations for the analyzed elements were as follows: 0.31% for Co, 0.86% for Sr, 2.99% for Cs, 6.05% for Na, 7.44% for K and 1.34% for Ca. The classification of the sorption curves was performed after the suggestions of Hinz (2000). The Langmuir and Freundlich curve equations were used to describe the sorption of the metals from the solution onto the studied sample (Limousin et al. 2007). The relative percentage change or "sorption intensity" (SI) was also used to compare the radionuclide sorption capacity of the sample (Xiong et al. 2005), which can be effectively used for comparison of affinities of elements with different initial molar amounts. The mineral changes during the sorption experiments were studied by XRD on oriented specimens.

## Results and Discussion

### Sorption curves

The shape of sorption curves (Fig. 2) indicates the highest affinity toward the studied sample for Co among the studied metals (H type curve) at low initial concentrations, while the L type curve of Cs and Sr also indicates high affinity

Fig. 2  
Sorption curves of Co, Sr and Cs for the studied sample within the entire range of initial metal concentrations (A) and at low initial metal concentrations (B). Broken line in Fig. 2A shows the area enlarged in Fig. 2B



and chemisorption (Giles et al. 1960). This latter fact is supported by the observation by Solovich-Vella and Garnier (2006), who found that among the studied metals the sorption of Cs is strong in a wide pH range (above 4.9). In our case the equilibrium pH values are always above this pH value (>8.84; Table 2). However, others found that the primary immobilization process for Cs is the ion exchange (Ainsworth et al. 2005). In the case of Co the sorbed metal amount slowly increases with increasing equilibrium concentration, also after the breaking point of the curve, suggesting the role of precipitation in the cobalt immobilization process (Stumm 1992). This observation is also true for Sr, however, it is not as characteristic as for Co. The significance of precipitation is also

Table 2  
Summary of the results of sorption experiments

	Type	SI <sub>joint</sub> (%)	Freundlich			Langmuir			pH	
			K <sub>F</sub>	1/n	R <sup>2</sup>	Q <sub>max</sub> (mg/kg)	b	R <sup>2</sup>	initial	equilibrium
Co	H1	19	12.8	0.192	0.94	9031	0.022	0.97	5.85 ± 0.08	8.93 ± 0.77
Sr	L1	34	33.4	0.564	0.98	24 657	0.009	0.98	5.79 ± 0.16	7.90 ± 0.61
Cs	L1	26	13.5	0.347	0.96	11 834	0.050	0.99	5.71 ± 0.28	9.49 ± 0.20

SI<sub>joint</sub> = joint sorption intensity, K<sub>F</sub> = Freundlich adsorption constant, n = Freundlich exponent, Q<sub>max</sub> = maximum sorption capacity, b = Langmuir equilibrium constant

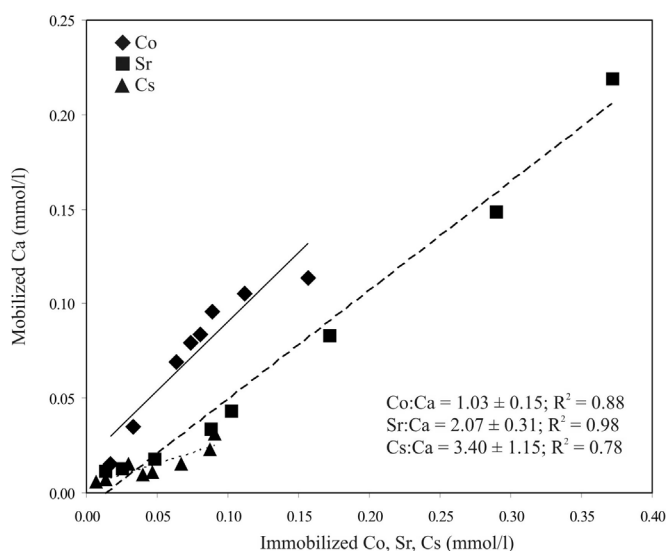
supported by the alkaline pH values (>7.27; Table 2) even at high initial metal concentrations (see further discussion below in more detail). According to Criscenti and Sverjensky (1999) the adsorption of Co at low pH is not significant; however, its immobilization intensely increases with pH and it reaches its maximum between pH 6 and 8.3, depending on the characteristics of the adsorbent. Catalano et al. (2005) found that Co completely precipitates from the solution above pH 8 in forms of carbonate and hydroxide in alkaline sediments. This observation was also supported by our results, since cobalt was almost completely (99% of initial Co) immobilized in samples with equilibrium pH above 8 (Table 2). In spite of the significantly different experimental conditions, Mell et al. (2006a) also found that a significant part of Co and Sr was precipitated during their immobilization by the albitic facies of the BSF.

#### Sorption mechanisms

The amounts of mobilized Na and K do not show any change with increasing sorption of the studied elements. Calcium shows only slight increase with increasing Cs sorption, so the main immobilization process for this element is not ion exchange with Ca, Na or K (Fig. 3). In contrast, Vejsada et al. (2005) found strong competition between Cs and K toward clay surfaces, while this competition was weaker for Ca and Na. According to Mell et al. (2006a) the highest Cs amounts were adsorbed by the albitic claystone samples of BSF with the highest illite contents. In our case Cs could be immobilized by chemisorption on the clay minerals in the sample, which is also suggested by the shape of its sorption curve.

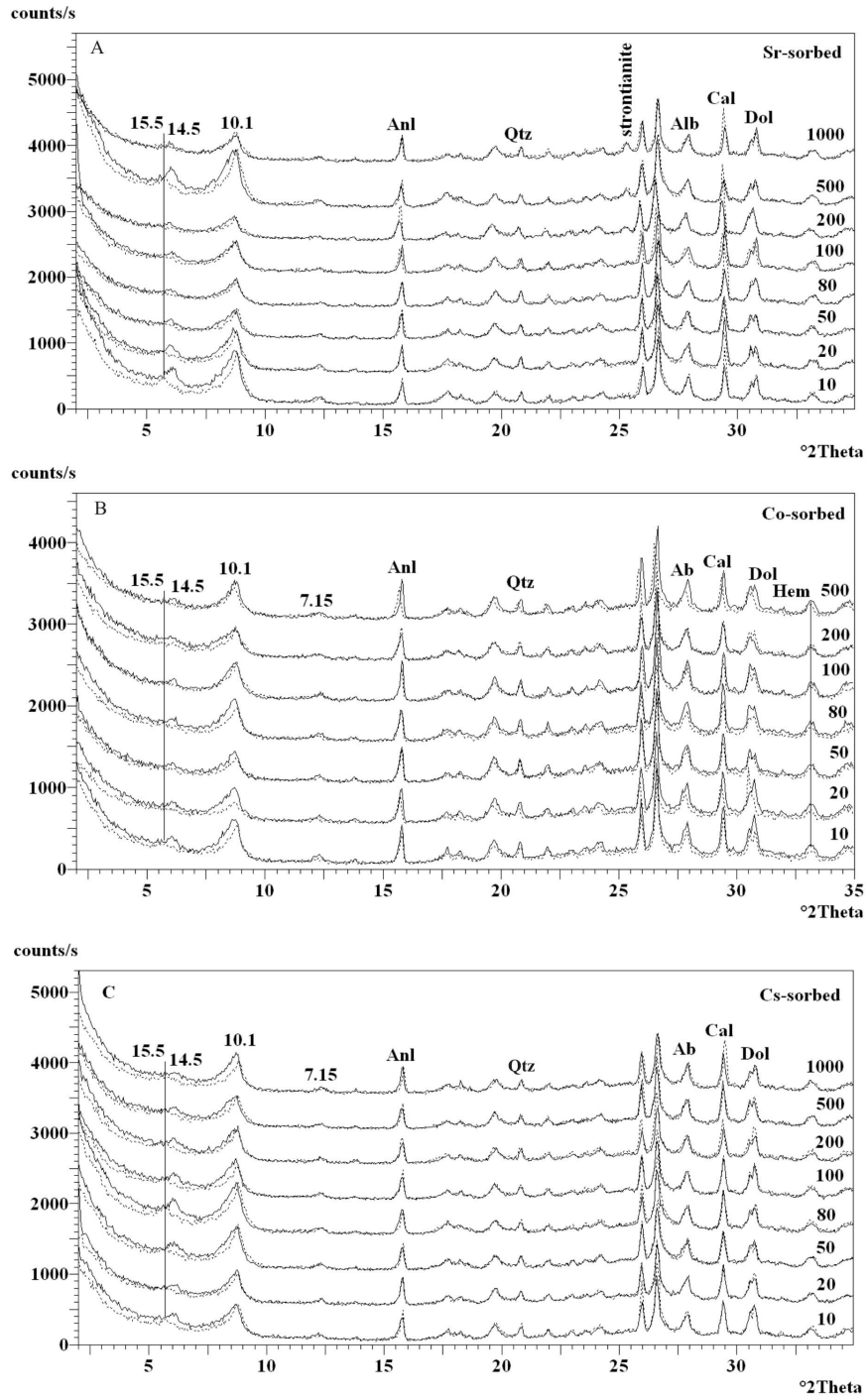
However, Co and Sr show different behavior in our case: the amounts of Ca significantly increased with their increasing sorption. The ratio of adsorbed Co to mobilized Ca is  $1.03 \pm 0.15$ . According to Santillan-Medrano and Jurinak (1975), when the ratio of adsorbed and desorbed element is close to 1 then the sorption is realized through ion exchange between the two elements, in our case between Co and Ca. At the highest initial Co concentration, the Co:Ca ratio significantly exceeds 1, which suggest the appearance of a new immobilization mechanism for this metal at this concentration level. This mechanism could be precipitation, as

Fig. 3  
Amount of mobilized Ca  
from the studied sample due  
to Co, Sr and Cs sorption



was suggested by the shape of its sorption curve. The increase of Ca concentration in the solutions with increasing Sr adsorption is lower than that for Co. The ratio of adsorbed Sr and Ca in solution is  $2.07 \pm 0.31$ . This suggests that there is an other immobilization process beside ion exchange between Ca and Sr. According to Missana et al. (2006) the primary immobilization process for Sr in argillaceous rocks is ion exchange; however, the surface complexity also plays an important role at alkaline conditions (above pH 8). This latter phenomenon was discovered by Chen and Hayes (1999) who observed strong competition between Sr and other ions such as Na and Ca at broad pH range. During the adsorption process partial dissolution of calcite may occur due to the mobilization of  $H^+$  from the mineral surfaces. As the solubility of  $SrCO_3$  is lower than that of calcite, the former will precipitate first due to the interaction of Sr and carbonate ions. Precipitation of strontianite ( $SrCO_3$ ) is confirmed by the presence of the most intense reflection at  $3.52 \text{ \AA}$  on X-ray diffractograms (Fig. 4A). Strontianite will precipitate upon addition of a concentration of 200 mg/l Sr to the rock sample, and above this concentration its amount gradually increases, supporting the increasing importance of precipitation as a mechanism of immobilization.

Beside the appearance of strontianite there are no significant mineralogical changes due to the sorption. There is no unambiguous sign of formation of Co precipitates (carbonate or hydroxide). However, the partial loss of expansion capacity of illite/smectite suggests the sorption of  $Co^{2+}$  in the interlayer space of the smectite component in I/S and Chl/S (Fig. 4B). Németh et al. (2002) demonstrated similar loss of expansion of Co-adsorbed montmorillonite. No change in the diffractograms can be detected for Cs (Fig. 4C). Based on XRD both the swelling clay minerals and analcime remained structurally unchanged





during sorption. Illite/smectite and chlorite/smectite preserved their expansion capacity. This supports our assumption that cesium is sorbed selectively and is fixed at specific sorption sites. These sites are the frayed edge sites which are abundant in illite/smectite (Cornell 1993).

The role of analcime in Cs (and Sr) sorption is a relevant question because of the structural relationship between analcime ( $\text{NaAlSi}_2\text{O}_6 \times \text{H}_2\text{O}$ ) and pollucite ( $\text{CsAlSi}_2\text{O}_6 \times \text{H}_2\text{O}$ ). Since analcime structure is one of the tightest among zeolite structures, cesium exchange in analcime is very slow at low temperatures (Vaughn 1978). However, analcime is fairly well able to incorporate Cs into its framework during crystallization above 200°C (Barrer 1978). Recently, Montagna et al. (2010) demonstrated that Cs can be entrapped in analcime framework-type aluminosilicates by hydrothermal synthesis at 115°C. Incorporation into the solid solutions between analcime and pollucite is suggested to be the major mechanism for cesium retention at 120°C, as suggested by Inagaki et al. (2006).

#### *Sorption capacity*

The maximum sorption capacities of the studied sample for Co, Cs and Sr are 0.153, 0.089 and 0.281 mol/kg, respectively, as calculated from the Langmuir equations (Table 2). They correspond to 9031, 11 834 and 24 657 mg Co, Cs and Sr/kg of analcime-containing claystone, respectively. Taking into account that according to Mell et al. (2006b) the diffusion of metals in the albitic facies of the BSF is very slow (4–6 mm in 210 days), the sorption capacities found can be considered to be significant values. Under the experimental conditions used significantly more sorption sites are available for the metals than in the fresh rock, due to grinding and suspending of the rock sample, so these values are ones of principle; however, they can be effectively used for comparison of affinity of metals for the claystone. Among the studied elements, Sr shows the highest immobilization by analcime-bearing claystone, followed by Co and finally Cs. At low initial metal concentrations the distribution coefficients for Co are higher by one order of magnitude compared to the other studied elements, which can be due to alkaline conditions and the strong precipitation of Co. Several authors (Carroll et al. 1997; Solovich-Vella and Garnier 2006, etc.) found different affinity sequences of the studied elements for claystone; however, different initial concentrations, adsorbents used and experimental conditions strongly influence the results. Mell et al. (2006a) found that among the studied elements the affinity of Sr is the lowest for the albitic claystone facies of the BSF. However, the

← Fig. 4

X-ray diffractograms of Sr (A), Co (B) and Cs (C) sorbed oriented samples before (continuous line) and after (dotted line) ethylene glycol treatments. *d* values are in Å. Anl = analcime, Qtz = quartz, Ab = albite, Cal = calcite, Dol = dolomite, Hem = hematite. Numbers at the right indicate the concentration of added cations in mg/L

significantly different mineralogical characteristics, primarily the presence or absence of large amounts of analcime with high adsorption capacity on the one hand, and different experimental conditions on the other, may result in differences in affinities of a given ion for the different formations.

### **Conclusions**

The affinity of the studied ions for the analcime-bearing claystone sample decreases in the order of  $Sr > Co > Cs$ . The shape of sorption curves suggests high affinity for each studied metal. In the case of Sr and Co, precipitation and ion exchange plays an important role in metal immobilization, while in the case of Cs no proof was found either for precipitation or ion exchange. Cesium seems to be fixed on specific sorption sites in illite/smectite. Our results suggest the high radionuclide immobilization capacity of the studied analcime-containing claystone. Since the role of analcime in the sorption processes in the BSF is not yet clear, further studies are necessary to evaluate the sorption characteristics of this formation. However, it is permissible to state that analcime may readily fix Cs in its structure when recrystallized to Cs-analcime or pollucite at elevated temperatures of the nuclear waste disposal site, and thus, its presence in the BSF is beneficial in retarding radionuclides.

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