EXPERIMENTAL STUDY AND MATHEMATICAL MODELING OF Cs (I) AND Sr (II) SORPTION ON BENTONITE AS BARRIER MATERIAL IN DEEP GEOLOGICAL REPOSITORY

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ABSTRACT
The migration of radionuclides through bentonite barrier in deep geological repository is strongly influenced by sorption processes. In our deep geological repository in granite massif, the canisters made of stainless and carbon steel and compacted bentonite barrier are planed. The experimental investigation on sorption processes in such system and their appropriate mathematical description including observation of a main corrosion product of the canister wall (namely magnetite) were the aim of this work.
Based on own experiences and literature data, we arranged batch experiments in PE vials. The experimental systems consisted of: (i) synthetic granitic water with a given ionic strength ($NaNO_3$) and concentrations of studied radionuclides (10^{-6} mol/l CsCl or $SrCl_2.6H_2O$ spiked with ^{137}Cs or ^{85}Sr); (ii) pre-treated (with the aim to remove carbonates) bentonite suspension, magnetite and their mixtures.
The experimental data were modeled using: (i) surface complexation models for edge sites, namely, two electrostatic – Diffusion Double Layer Model (DLM) and Constant Capacitance Model (CCM) - and Chemical Model (CEM), without an electrostatic correction, (ii) ion exchange model (IExM) for layer sites. The corresponding program package was developed (STAMB-2003). Sorption and protonation parameters were obtained by fitting of appropriate experimental data. The quality of fit was evaluated by means of the WSOS/DF criterion, the values of which should be lower than approx. 20.
In the first modeling step, the interaction of protons and hydroxyl ions with the solid phases was investigated by alkalimetric and acidimetric titrations under exclusion of CO_2 (under N_2 atmosphere) for ionic strengths of background electrolyte 0.1 and 0.01 ($NaNO_3$). On the basis of WSOS/DF criteria values and the meaningful values of the parameters, the CEM model and the ion exchange model (IExM), were chosen and the corresponding values of the protonation constants, total concentration of edge sites, cation exchange constants, total concentration of layer sites were obtained and used for further calculations.
In the second modeling step, the percentage of sorption of Cs(I) and Sr(II) as a function of pH was determined under oxic conditions at room temperature. During 28 - day period (28 days are required for equilibrium establishment) the pH of the system was five times adjusted to the pH value required. Then phases were separated by centrifugation and the equilibrium concentrations in solution were determined. The experimental data of sorption were fitted by CEM+IExM and the corresponding averaged values of sorption constants, including the WSOS/DF, were obtained. These sorption parameters, and parameters mentioned above, were successfully used for simulation calculations of pH-dependences of sorption based on two modeling approaches, namely on so called GC (Generalized Composite) and CA (Component Additivity) approaches.
It has been found that: (i) bentonite is an efficient sorbent for Cs(I) and Sr(II), (ii) the reactions taking place on layer sites of bentonite play the most important role in the sorption process, mainly in the case of Cs, (iii) bentonite is much better sorbent than magnetite, (iiii) the system can be successfully modeled using surface complexation model type of CCM in combination with ion exchange model.

KEYWORDS: barriers, bentonite, magnetite, sorbent, radionuclides, titrations

1. INTRODUCTION

In our deep geological repository the canisters made of stainless and carbon steel and compacted bentonite barrier are planed. The use of bentonite as a backfill barrier in repositories for a nuclear waste is based mainly on its low permeability, swelling properties and capability to significantly retard the migration of most radionuclides. A diffusion strongly influenced by sorption phenomena is a controlling process of radionuclide migration through bentonite barrier in deep geological repository. To be able to

predict the radionuclide fate in repositories, mathematical models have to be developed and for this purpose understanding of each process is necessary. Investigation of sorption characteristics of Cs and Sr in bentonite and corrosion products of magnetite and their mixtures together with their appropriate mathematical description are considered to be the main aims of the research.

The bentonite (clay) surface contains at least two types of surface functional groups. The first type - permanently charged surface functional groups are

created by ionic substitution within the crystal structure. Isomorphic substitution of Al^{3+} for Si^{4+} within the tetrahedral layer creates a permanent negative charge on the mineral surface which is compensated externally by cations. These structure charges of surface sites are denoted as layer sites. On the edges of surface there are sites ($\equiv$SOH) with pH-dependent charge as result of the "adsorption" of H^+ ions (then the so called protonation proceeds: $\equiv$SOH $\rightarrow$ $\equiv$SOH$_2^+$) or "desorption" of H^+ ions (then deprotonation proceeds: $\equiv$SOH $\rightarrow$ $\equiv$SO$^-$) depending on pH of the solution. These variable-charge surface sites are designated as edge sites. These two surface site types are responsible for two types of uptake-processes. The first one, on layer sites, tending to be dominant at low pH and/or high sorbate concentrations is a cation exchange mechanism. The second one, on edge sites, causing dependency of sorption on pH is a surface complexation mechanism. The surface sites of the second type – edge sites were also found for oxide surfaces, so we expected to find them on magnetite.

Titration curve data for bentonite, magnetite and their mixtures and sorption experimental data (sorption dependencies on pH) for Cs and Sr from granitic water were acquired. The resulting data were modeled using:

(i) Surface complexation models for edge sites, namely, two electrostatic – Diffusion Double Layer Model (DLM) and Constant Capacitance Model (CCM) - and Chemical Model (CEM), without an electrostatic correction;

(ii) Ion exchange model (IExM) for layer sites.

The corresponding program package was developed (STAMB-2003). For the modeling of sorption of Cs and Sr on the mixtures mentioned above, two different approaches were tested:

(i) Generalized Composite Approach (GC), where the bentonite-magnetite mixture was considered as compact sorbent characterized by a single set of titration and sorption parameters, which were sought by direct fitting of experimental data.

(ii) Component Additivity Approach (CA), composed of weighted combination of models describing sorption on bentonite and magnetite. Sorption parameters were obtained by fitting appropriate experimental data obtained with individual sorbents. Data calculated by CA Approach for mixture were compared with corresponding experimental data.

Formulation of sorption equations was based on speciation of Cs(I) and Sr(II) in granitic water. In case of cesium, Cs^+ dominates in the system in the whole pH range. For strontium, Sr^{2+}, $SrCO_3^0$, $SrNO_3^+$ and $SrSO_4^0$ are important occurring forms (> 0.1 %); precipitation of $SrCO_3$ occurs approx. at pH > 10. All activity coefficients were calculated using Davis equation. Both formation of sorption complexes on edge sites ($\equiv$SO$^-$, $\equiv$SOH, $\equiv$SOH$_2^+$) and sorption by ion exchange on layer sites ($\equiv$X) were considered for the sorbents being modeled. Thus the protonation and sorption reactions used in the modeling were as follows:

$SO^- + H^+ \leftrightarrow SOH^0$		(KS1)
$SOH^0 + H^+ \leftrightarrow SOH_2^+$		(KS2)
$SOH_2^+ + CO_3^{2-} \leftrightarrow SOH_2CO_3^-$		(KS3)
$SOH_2^+ + HCO_3^- \leftrightarrow SOH_2HCO_3^0$		(KS4)
$XNa + H^+ \leftrightarrow XH + Na^+$		(KS5)
$SO^- + Cs^+ \leftrightarrow SOCs^0$ (KCs1)	$SO^- + Sr^{2+} \leftrightarrow SOSr^+$	(KSr1)
	$2SO^- + Sr^{2+} \leftrightarrow (SO)_2Sr^0$	(KSr2)
	$SO^- + SrNO_3^+ \leftrightarrow SOSrNO_3^0$	(KSr3)
	$SO^- + SrCO_3^0 \leftrightarrow SOSrCO_3^-$	(KSr4)
$XH + Cs^+ \leftrightarrow XCs + H^+$ (KCs2)	$2XH + Sr^{2+} \leftrightarrow X_2Sr + 2H^+$	(KSr5)
	$XH + SrNO_3^+ \leftrightarrow XSrNO_3 + H^+$	(KSr6)

Table 1 Conditions of titration experiments

Solid phase	Ionic strength I [M NaNO$_3$]	Weight m [g]	Volume V [ml]	Liquid to solid ratio V/m [ml/g]
Bentonite (B)	0.1	0.3885	70	206
Bentonite	0.01	0.3885	70	206
Magnetite (M)	0.1	0.6000	120	200
Magnetite	0.01	0.6000	120	200
Mixture B/M = 2/1	0.1	B-0.0389; M-0.0190	70	1226
Mixture B/M = 2/1	0.01	B-0.0389; M-0.0190	70	1226

The equilibrium constants for the protonation and sorption reactions were written in the classical way, e.g. for the constant KSr2:

$$KSr2 = \frac{\left[(SO)_2 Sr^0\right]}{\left[SO^-\right]^2 \cdot \left[Sr^{2+}\right]} \exp(2.\psi.F/(R.T))$$

where F is the Faraday constant, R is the universal gas constant, T is the absolute temperature and Ψ is the electrostatic potential.

The titration and sorption experiments were performed in order to get all data necessary for surface complexation and ion-exchange modeling.

2. EXPERIMENTAL

2.1. MATERIALS

Czech commercial sodium bentonite Sabenil (KERAMOST, Obrnice) was examined. It had following parameters: specific surface area – 44.4 m^2/kg (BET method); mineralogical composition – montmorillonite + nontronite 70 %, quartz 7.9 %, muscovite + illite 6.9 %, goethite 6.6 %, feldspar 5 %, anatase 4% and others; chemical composition – SiO$_2$ 41 %, Fe$_2$O$_3$ 12 %, Al$_2$O$_3$ 11 %, CO$_2$ 1.04 %, TiO$_2$ 3.5 % and others; granulometry – 82.5 % below 0.01 mm. Bentonite was pre-treated (preparation of bentonite suspension) in order to remove carbonate and other impurities. After that it was treated with the procedure described in [1] being modified in the following sequence of operations: about 2.5 g of the bentonite powder was continuously mixed with 40 ml of 1 M NaNO$_3$ + 10^{-3}M HNO$_3$ solution with successive addition of 1M HNO$_3$ solution in 10 minute interval until pH = 3 is reached. Then the suspension was centrifuged and washed with 60 ml 0.1 M NaNO$_3$, centrifuged and supernatant pH was measured. This procedure was repeated until pH had stabilized between pH 5.1 and 5.3. The last two cycles were performed with 0.01 M NaNO$_3$ solution. Finally, the clay was re-dispersed in 0.01 M NaNO$_3$ solution and stored.

In order to confirm the efficacy of the pre-treating procedure, RTG analyses were performed in Institute of Inorganic Chemistry, Academy of Sciences of the Czech Republic. The comparison of powder and suspension showed that impurities had been successfully removed while other minerals stayed intact.

Magnetite was explored as a representative of corrosion products. We used an inorganic pigment Bayferrox 318M, Bayer Chemicals with following characteristics: Fe$_3$O$_4$ – 92 %, SiO$_2$+Al$_2$O$_3$ – 3 %, granulometry 0.2 μm.

The synthetic granitic water was used as liquid phase, composition (M): Na$^+$ 2.3x10^{-3}, K$^+$ 1.1x10^{-4}, Ca^{2+} 3.6x10^{-4}, Mg^{2+} 1.8x10^{-4}, Cl$^-$ 2x10^{-3}, SO$_4^{2-}$1.1x10^{-4}, ΣCO$_3$ 1.8x10^{-3}, pH = 8.2, ionic strength approx. 0.004. The higher ionic strength was adjusted with NaNO$_3$; CsCl or SrCl$_2$.6H$_2$O was added in concentration required and solution was spiked with ^{137}Cs or ^{85}Sr. High purity water and analytical grade chemicals were used for all experiments Milli-Q.

2.2. POTENTIOMETRIC TITRATIONS

The interaction of protons and hydroxyl ions with the bentonite, magnetite and their mixture surface groups were investigated by potentiometric titration under exclusion of CO$_2$ from the system (under N$_2$ atmosphere). Alkalimetric and acidimetric titrations were performed in separate experiments. Two titration curves, namely for ionic strength 0.1 and 0.01, were obtained (overview of titration experiments is summarized in Table 1). pH and values of potential were recorded after 5 and 10 minutes after addition of titre (0.1 M HCl, NaOH). The next titre addition was conditioned by drift in the measured potential less than 2 mV between the readings after 5 and 10 minutes (Wanner, Albinsson, 1994).

2.3. SORPTION EXPERIMENTS

Batch sorption experiments in PE tubes under oxidation conditions at room temperature were designed. All experiments were carried out with duplicate or triplicate samples. Preliminary experiments, with the aim to study sorption on vessel walls (< 3 %), phase separation conditions, time of

Table 2 Conditions of sorption experiments

Solid phase (Sorbent)	Sorbate	Ionic strength I [M NaNO$_3$]	Liquid to solid ratio V/m [ml/g]	pH interval studied
Bentonite	Cs	0.14; 0.04	395; 420	3.5 – 10.5
(B)	Sr	0.14; 0.04	155; 285	3.5 – 9
Magnetite	Cs	0.14; 0.04	40; 100	3.5 – 10.5
(M)	Sr	0.14; 0.04	50; 25	3.5 – 9
Mixture	Cs	0.14; 0.04	B/M = 2/1; 10/1; 1/25; 1/50	3.5 – 10.5
(B+M)	Sr	0.14; 0.04	B/M = 1/25; 1/50	3.5 – 9

Table 3 Parameters characterizing solid phase (sorbent)

Syst.	Model	Edge-sites (CEM)			Layer-sites (IExM)		Sorption of CO_3^{2-} and HCO_3^-	
		log KS1 (I = 0)	log KS2 (I = 0)	log ΣSOH_{TOT} [mol/kg]	log KS5 (I = 0)	log ΣX_{TOT} [mol/kg]	log KS3 (I = 0)	log KS4 (I = 0)
B	CEM+IExM	9.81 ± 0.11	7.79 ± 0.29	-0.96 ± 0.02	2.03 $^=$	-1.40 ± 0.04	2.95*	6.38*
M	CEM+IExM	9.70 ± 0.02	6.07 ± 0.17	-1.42 ± 0.01	3.29 $^=$	-1.25 ± 0.13	4.83+	4.89+
B+M	CEM+IExM	6.58	7.59	-0.86	2.84	-1.62	3.58x	5.88x

* Štamberk et al. (1998)
+ Štamberk et al. (2002)
x values obtained as a weighted average of logarithms of bentonite and magnetite constants (based on mixture composition B/M = 2/1)
$^=$ values obtained by calibration method.

equilibrium (28 days are required for equilibrium establishment) and sorption isotherms were performed. The sorption edges, i.e. the percentage of sorption as a function of pH, were determined under given conditions (see Table 2). The suspension of bentonite, magnetite powder or their mixture was mixed with required amount of liquid phase (synthetic granitic water, ionic strength NaNO$_3$, 10^{-6} mol/l CsCl or SrCl$_2$.6H$_2$O labelled with ^{137}Cs or ^{85}Sr). During 28 - day period the pH of the system was five times adjusted to the pH value required. Then phases were separated by centrifugation and the equilibrium Cs or Sr concentrations were determined in solution by γ-spectroscopy.

3. RESULTS

3.1. EVALUATION OF TITRATION CURVES

In the first modeling step, the protonation constants (KS1, KS2; re-counted for $I = 0$), total concentration of edge sites (ΣSOH_{TOT} in mol/kg), cation exchange constants (KS5; re-counted for $I = 0$), total concentration of layer sites (ΣX_{TOT} in mol/kg) for bentonite, magnetite and their mixture and Helmholz capacitance G for CCM were obtained via fitting titration data. These values (see Table 3) formed the input data set for sorption modeling. The goodness of fit was evaluated by means of the WSOS/DF criterion (Herbelin, Westall, 1996). The good fit is indicated when the criterion value is less then 20) and on the basis of these criteria values and the meaningfullness of the parameter values, the CEM model combined with the ion exchange model (IExM) were chosen for further calculation. The experimental and modeling results for one selected system are depicted in Fig. 1.

3.2. GC APPROACH – SORPTION ON MIXTURE OF BENTONITE (B) + MAGNETITE (M)

The sorption data under conditions given in Table 2 were finalized using CEM+IExM and the constants characterizing the sorption of Cs(I) on the mixture of B+M (B/M=2/1) were obtained (input parameters were taken from Table 3). The resulting average values of sorption constants are shown in Table 4 and calculated pH dependences of Exp. 1 (Cs(I); B/M=2/1; I=0.14; V/m=363 ml/g. WSOS/DF = 1.34) and Exp. 2 (Cs(I); B/M=2/1; I=0.04; V/m=363 ml/g. WSOS/DF = 0.97) are shown in Fig. 2. It is evident that the coincidence of both experiments is very good.
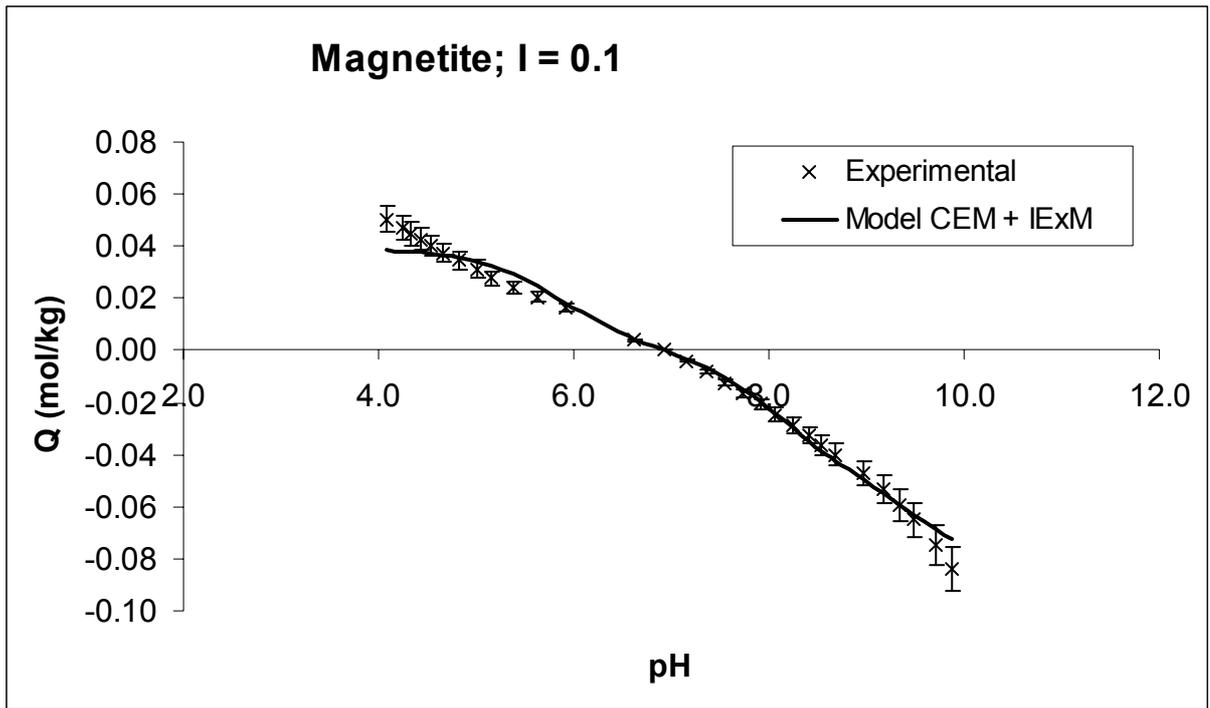
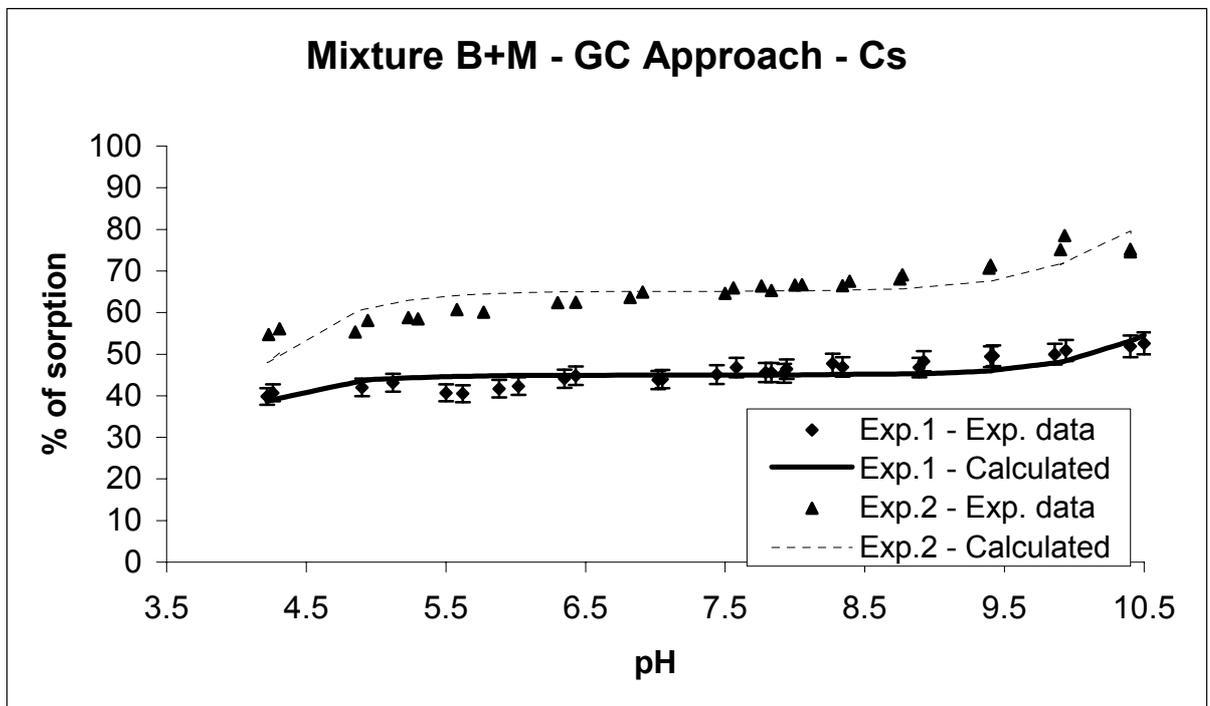**Fig. 1**   Experimental data and mathematical modeling for titration curve of magnetite I = 0.1

Fig. 2   Experimental data and modeling results for mixture B+M - Exp. 1 (Cs(I); B/M = 2/1; I = 0.14; V/m = 363 *ml*/*g*. WSOS/DF = 1.34) and Exp. 2 (Cs(I); B/M = 2/1; I = 0.04; V/m = 363 *ml*/*g*. WSOS/DF = 0.97)

Table 4 Average values of sorption constants

Solid phase	Sorption constants ($I = 0$)					
	log KCs1 ($\equiv$SOCs)			log KCs2 ($\equiv$XCs)		
Bentonite	5.81 ± 0.34			1.08 ± 0.13		
Magnetite	3.41 ± 0.38			-1.56 ± 0.18		
Mixture (B/M=2/1)	4.67 ± 0.69			0.41 ± 0.20		
	Sorption constants ($I = 0$)					
	log KSr1 ($\equiv$SOSr$^+$)	log KSr2 ($\equiv$(SO)$_2$Sr)	log KSr3 ($\equiv$SOSrNO$_3$)	log KSr4 ($\equiv$SOSrCO$_3^-$)	log KSr5 ($\equiv$X$_2$Sr)	log KSr6 ($\equiv$XSrNO$_3$)
Bentonite	7.32±0.61	9.27±1.31	7.90±1.05	7.59±1.68	-0.54±0.78	1.30±1.44
Magnetite	5.39±0.62	6.38±0.42	5.68±0.89	5.40±1.05	-8.34±0.27	-1.40±0.14
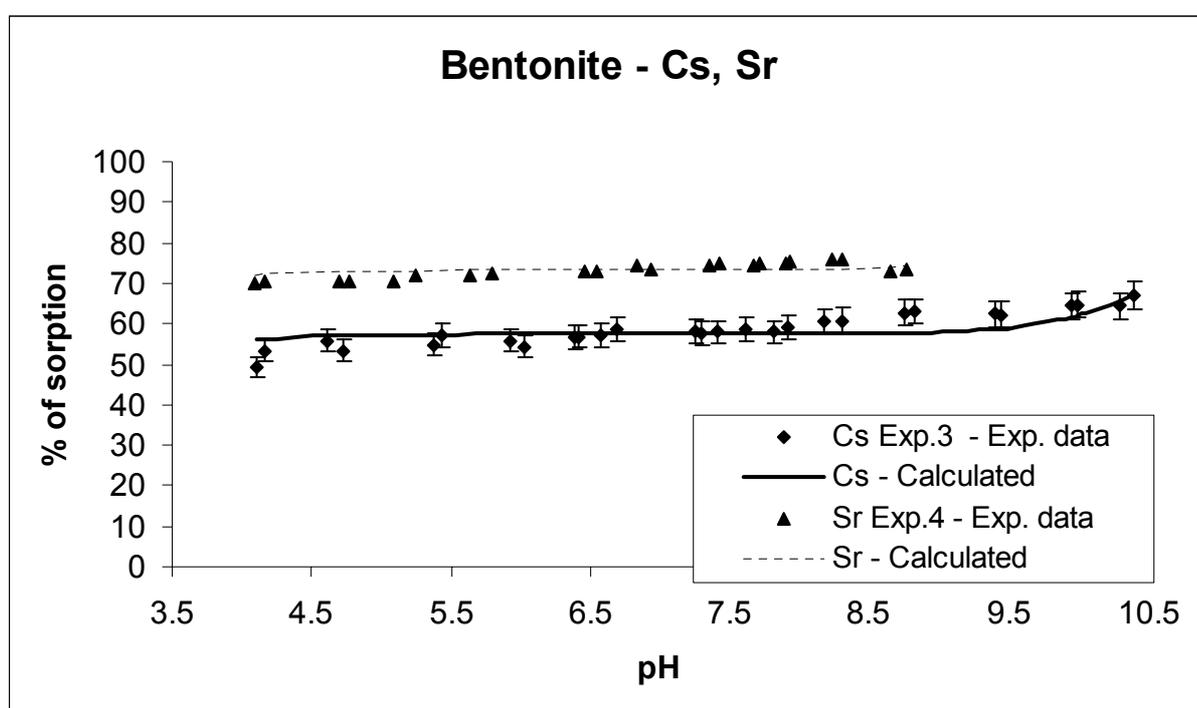
Fig. 3 Experimental data and modeling results for bentonite Exp. 3 (Cs(I); <u>B</u>; $I = 0.14$; *V/m* = 395 *ml/g*. WSOS/DF = 1.39) and Exp. 4 (Sr(II); <u>B</u>; $I = 0.14$; *V/m* = 285 *ml/g*. WSOS/DF = 0.26)

CA APPROACH

1. *SORPTION ON COMPONENTS OF BENTONITE (B) + MAGNETITE (M) MIXTURE*

The experimental data of sorption of Cs(I) or Sr(II) separately on bentonite and separately on magnetite (the conditions see Table 2) were fitted by CEM+IExM in the same manner as in previous case. The corresponding average values of sorption constants are given in Table 4. The calculated pH dependences of Exp. 3 (Cs(I); <u>B</u>; I=0.14; *V/m*=395 *ml/g*. WSOS/DF=1.39) and Exp. 4 (Sr(II); <u>B</u>; I=0.14; *V/m*=285 *ml/g*. WSOS/DF=0.26) are presented in the form of graphs in Fig. 3; the calculated dependences of Exp. 5 (Cs(I); <u>M</u>; I=0.14; *V/m*=40*ml/g*.

WSOS/DF=7.34) and Exp. 6 (Sr(II); <u>M</u>; I=0.14; *V/m*=25 *ml/g*. WSOS/DF=33.1) are shown in Fig. 4. It is evident that the sorption of both metals on magnetite depends more on pH comparing to their sorption on bentonite, but the values of WSOS/DF point to have the better goodness-of-fit of sorption on bentonite. In addition, as found the reactions taking place on layer sites play the most important role in the sorption process mainly in case of bentonite. Although we didn't expect it, we had to use the combination of surface complexation and ion exchange model also in case of magnetite because only surface complexation one did not satisfactorily describe sorption dependencies obtained for magnetite.
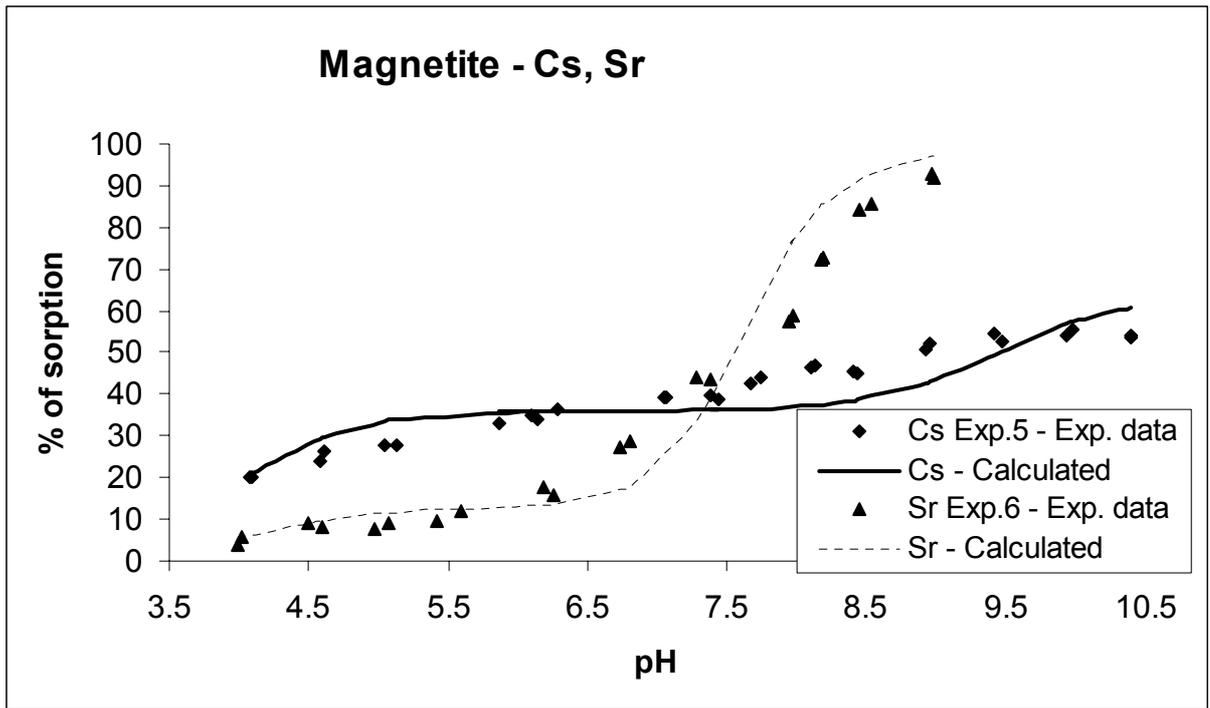
Fig. 4 Experimental data and modeling results for magnetite Exp. 5 (Cs(I); <u>M</u>; *I*=0.14; *V/m*=40 *ml/g*. WSOS/DF=7.34) and Exp. 6 (Sr(II); <u>M</u>; *I*=0.14; *V/m*=25 *ml/g*. WSOS/DF=33.1)
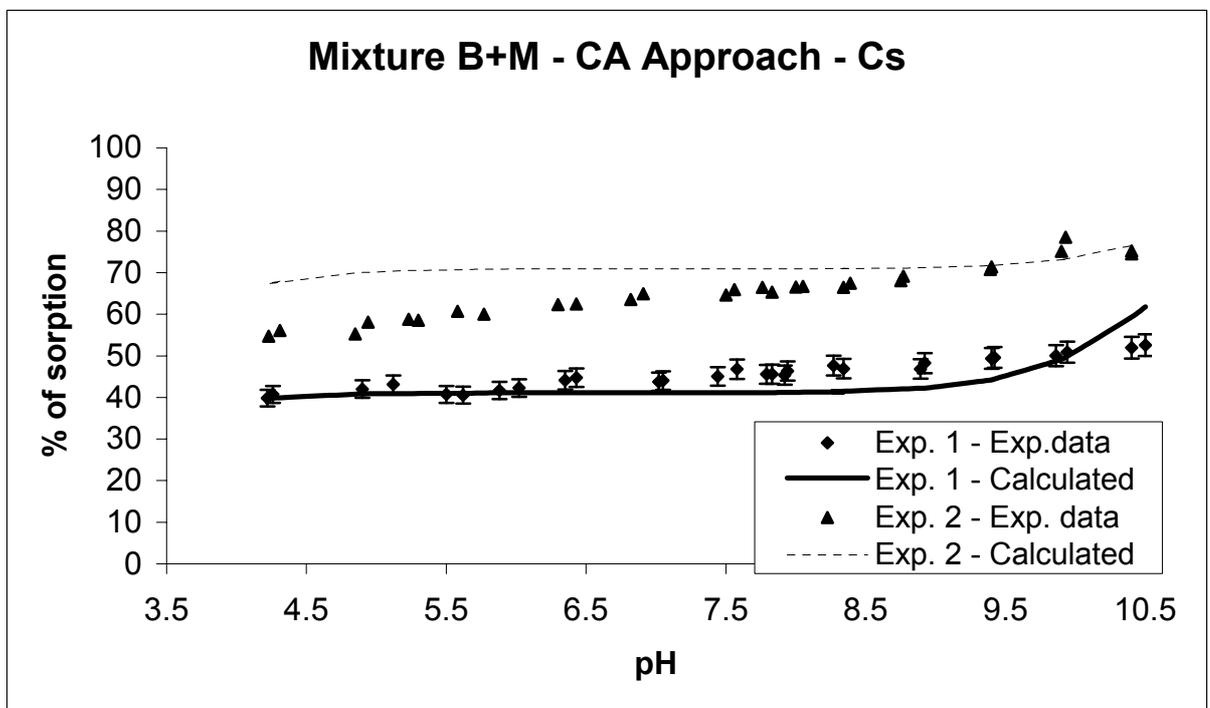

Fig. 5 Experimental data and modeling results for mixture B+M - Exp. 1 (Cs(I); B/M=2/1; *I*=0.14; *V/m*=363 *ml/g*. WSOS/DF = 3.11) and Exp. 2 (Cs(I); B/M=2/1; *I*=0.04; *V/m*=363 *ml/g*. WSOS/DF = 6.61)

2. PREDICTION OF CS(I) AND SR(II) SORPTION ON MIXTURE OF BENTONITE + MAGNETITE

CEM + IExM models for the sorption of cesium or strontium by bentonite and magnetite were incorporated into CA Approach code by means of which the sorption of Cs(I) and Sr(II) under different conditions were calculated with input data from Table 3 and 4. The calculated dependences for Exp. 1 and 2 (see above) are depicted in Fig. 5. These results were compared with GC approach modeling results.

Hereafter the sorption behaviour of mixtures of different composition (ratio B/M = Cs: 10/1; 2/1; 1/25; 1/50; Sr: 1/25; 1/50) was modeled via CA approach. The results of simulations were satisfying, mainly in the region of pH: 7-9.5, which is the region important from the perspective of conditions in deep geological repositories.

3.3. COMPARISON OF GC AND CA APPROACHES

Two sets of experimental data for cesium sorption by mixture of bentonite and magnetite were modeled. In order to enable quantitative comparison, calculation of values of WSOS/DF was incorporated into CA Approach code. The following order of suitability of CA and GC approaches using CEM + IExM models predicting the sorption of cesium on the mixture of bentonite and magnetite was found:

Cs + mixture (B/M=2/1) Exp. 1 (I = 0.14) - WSOS/DF: GC(1.34) > CA(3.11)

Cs + mixture (B/M=2/1) Exp. 2 (I = 0.04) - WSOS/DF: GC(0.97) > CA(6.61).

It seems to be evident that both approaches are well comparable and suitable but GC Approach seems to be more accurate.

4. CONCLUSIONS

The sorption of selected fissionable products, Cs(I) and Sr(II), on bentonite and magnetite (as representatives of corrosion products) including their mixtures was studied by batch radiotracer method. Titration curves and sorption edges were acquired and modeled via three types of surface complexation models for edge sites and ion exchange model for layer sites.

On the basis of experimental results it can be concluded that sorption on magnetite is much more pH dependent then sorption on bentonite for both, Cs and Sr. From the experimental data course it is evident that sorption of cationic forms of these nuclides runs via surface complexation on deprotonized forms of edge sites in basic pH region and via ion exchange mechanism on layer sites in acidic pH region. This was observed mainly in case of bentonite but also experimental data and results of mathematical modeling in case of magnetite showed these two mechanisms though in less extent. Up to now, the layer sites on magnetite were not described in literature and that is why we did not find the satisfying explanation for ion exchange mechanism sites on this material. But on the other hand it is interesting that the structure of magnetite is alike structure of bentonite, i.e., that it is built of octahedral and tetrahedral layers, too. It implies that the explanation for ion exchange mechanism mentioned above may be found just with structural features of both minerals being similar.

During the evaluation of titration data sets, the most appropriate surface complexation model – chemical model (CEM) combined with ion exchange model (IExM) - were chosen on the bases of WSOS/DF criterion and physical meaningfullness of constant values. The constants characterizing bentonite, magnetite and mixture B/M=2/1 (protonation constants, total concentration of edge sites, cation exchange constants, total concentration of layer sites) were evaluated and used for sorption modeling.

From the sorption experimental and modeling data it is evident that:

- sorption of Cs is more selective than sorption of Sr

- bentonite is stronger sorbent than magnetite

- sorption on layer sites asserts more remarkably in case of Cs.

Even the behaviour of mixture B/M=2/1 is closer to the behaviour of pure bentonite phase. The growing influence of magnetite on sorption character with a raising content of magnetite in the mixture is evident.

The CA Approach was compared with GC Approach in case of Cs sorption on mixture of B/M=2/1 composition. In case of GC Approach titration curve of the mixture was evaluated and these parameters were used for sorption modeling. Both approaches are well comparable and suitable, but strictly judged GC Approach seems to be more accurate.

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