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Effect of pH and Ionic Strength on ^{152}Eu Sorption to Granitic Rocks and Component Minerals

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Abstract

The basic objective of radioactive waste management is to prevent unacceptable radiological risk for people today, and for future generations. The immobilisation of radionuclides in groundwater environments cannot be explained solely by the empirical models. The health and safety approach to the storage of radioactive waste in different geochemical media depends solely on the ability of rocks to retard through sorption any leach radionuclides from the near field. Sorption studies of ^{152}Eu as a representative member of trivalent actinides were carried out in the lab at varying pH and different saline environment using different granitic rocks and minerals. To investigate the effect of ionic strength as well as pH, two sets of experiments, in the presence of 0.1 and 0.05 mol·dm^{-3} NaCl, were used with a control having no NaCl present. The effect of surface area has been taken into consideration in the surface complexation studies using the JChess geochemical code. NaCl is used to create a low concentration brine/seawater environment. With the possible disposal/storage of nuclear waste in deep geological repository. Sample separation was performed as described above and counting was performed for Eu counting was performed based on its gamma emitting properties using Cobra II Auto Gamma. Varying pH had no effect on sorption in 0.05 and 0.1 mol·dm^{-3} NaCl systems however, net sorption for all Eu species in solution was higher than for NaCl free system. Sorption varied with pH below pH 7 for sorption in NaCl free system. Saturation was attained after pH 8. Sorption in varying pH showed similar sorption profiles in both NaCl systems. Eu sorption is enhanced in the presence of NaCl. Surface complexation constants, Log K values for experimental data have been determined by fitting modelled data to experimental data. The modelling ap-

proach can be included efficiently in transport simulations having chemical gradients in space and time.

Keywords

Surface Complexation, Sorption Edge, Saline Environment, Immobilisation of Radionuclides

1. Introduction

The basic objective of radioactive waste management is to prevent unacceptable radiological risk for people today, and for future generations. Risk is defined here as the probability of fatal cancer for an individual, or of a serious deleterious/hereditary effect for his/her descendants. As toxic elements can be associated with radioactive nuclides during conditioning processes or decay, a chemical risk assessment must be considered. This can be appreciated most by considering the concentrations of the elements in water or soils with regard to limiting concentrations (Aksoyoglu, 2008). ^{152}Eu, ^{154}Eu, and ^{155}Eu are produced primarily as fission products from fissile nuclides such as ^{235}U, ^{152}Eu can also be produced by neutron activation of nuclear reactor control rods.

$^{151}\text{Eu} + {}^{1}\text{n} \rightarrow {}^{152}\text{Eu}$ neutron capture process

The immobilisation of radionuclides in groundwater environments cannot be explained solely by the empirical models discussed earlier by Ebong et al. (Ebong et al. 2026). These empirical models are not sensitive to the varying conditions that are found in nature. Sorption has been shown to be very sensitive to pH, Eh and ionic strength of the solution in which the radionuclides are found. Using mass action laws, it is possible to describe sorption to heterogeneous samples such as granites by use of the surface complexation models. Surface complexation modelling approaches are generally more robust in their application over varying geochemical conditions than empirical models because they adopt a more mechanistic approach to sorption. This flexibility is often gained at the expense of simplicity, and SCMs may require a larger number of parameters to accommodate their increasing complexity. Surface complexation models use mass action laws analogous to aqueous phase reactions to describe adsorption, thus, accounting for changes in chemical speciation, competitive adsorption, and other multisolute interactive chemical effects.

1.1. Surface Complexation Modelling

An empirical model can be defined as a mathematical description of the experimental data without any particular theoretical basis (USEPA, 1999). For example, the K_d (The partition (or distribution) coefficient, K_d, is a measure of sorption of contaminants to geomedia, and is defined as the ratio of the quantity of the adsorbate adsorbed per unit mass of solid to the amount of the adsorbate remaining

in solution at equilibrium). K_d values are thermodynamically determined at stated equilibrium conditions, Freundlich isotherm, and Langmuir isotherm are considered empirical models by this definition (USEPA, 1999). Mechanistic models refer to models based on thermodynamic concepts, such as reactions described by mass action laws and material balance equations. Four of the most commonly used mechanistic models include the Helmholtz, Gouy-Chapman, Stern, and Triple Layer models (Sposito 1984). The empirical models are often mathematically simpler than mechanistic models and are suitable for characterising sets of experimental data with a few adjustable parameters, or for interpolating between data points. On the other hand, mechanistic models contribute to an understanding of the chemistry at the interface and are often useful for describing data from complex multi-component systems. Cation adsorption occurs on both the σo (solid-metal ion interface) and σβ (metal ion water interface) which are termed inner and outer sphere complexes, respectively (Tanaka et al., 2004). Adsorption on the β-plane is more affected by variations in both background electrolyte and concentration of trace metal concentration than that which occurs on the o-plane. The formation of inner and outer sphere complexes depends upon the nature of oxide surfaces as well as on the adsorbate cation, ionic radius, degree of hydration and or complexation with the anion of the background electrolyte. It is also common that some of the metal adsorbed as hydrolysed species (Kanungo, 1994). Surface complexation reactions include both protonation and deprotonation reactions as shown below. Surface protonation/deprotonation leads to a non-permanent pH-dependent surface charge (+ or −) (Elzinga & Sparks, 2001). Reactions responsible for surface charge are such as:

1) $\equiv SOH^{2+} \rightarrow \equiv SOH^{0} + H^{+}$

$$K_{a1} = \frac{\left[\equiv SOH^{0}\right]H^{+}}{\left[\equiv SOH_{2}^{+}\right]} \tag{1}$$

2) $\equiv SOH^{0} \rightarrow SO^{-} + H^{+}$

$$K_{a2} = \frac{\left[\equiv SO^{-}\right]H^{+}}{\left[\equiv SOH^{0}\right]} \tag{2}$$

For very low activities of metal species and those of its complexes with surface species, the equilibrium constant may be expressed as (Kanungo, 1994; Tripathy et al., 2006)

$$\log\left[\frac{M_{adsorbed}^{n+}}{M_{soln}^{n+}}\right] = \log K_e + x\left(\mathrm{pH} + \log\left[\equiv SOH\right]\right) \tag{3}$$

In modelling sorption of radionuclides to surfaces at varying pH, the surfaces of the mineral are considered too complex to be quantified in terms of the contributions of individual phases to adsorption. Instead, it is assumed that adsorption can be described by SCM equilibria written for "generic" surface functional groups, with the stoichiometry and formation constants for each SCM mass law evaluated on the basis of simplicity and goodness-of-fit (Goldberg et al., 2007). The generic surface sites represent average properties of the granite type rather than specific

minerals.

The equilibrium constant K_e describes the distribution of a given constituent among its possible chemical forms if complex formation and dissociation reactions are at equilibrium. The equilibrium constant is affected by a number of factors, including the ionic strength of the aqueous phase, presence of competing reactions, and temperature (Hayes & Katz, 1996). A plot of the left-hand side as a function of {pH + log[≡SOH]} yields a linear relationship, the slope of which gives the stoichiometry of the reaction. This relationship is applicable in the region of steep rise in adsorption with pH (Kanungo, 1994). Values of "x" (x is a dimensionless constant) greater than unity suggest a mixed reaction type. If the adsorption is smeared, the proton stoichiometry in the region of lower pH value is even lower than 0.05 an indication that adsorption takes place in the β-plane (Kanungo, 1994). Main assumptions of SCM include:

1) Sorption occurs through interaction with the hydroxyl groups,

2) Constituent minerals are uncoated and do not interact with each other.

This model has been applied in calculating the proton stoichiometry (PS) at the point of steep rise in sorption with pH (Kanungo, 1994). This plot defines the narrowest region in the adsorption spectrum of a pH range and also gives the Log Ke (equilibrium constants) values. Because Rd values are very sensitive to pH, as we shall see in the sections that follow, the Rd concept will have limited application in describing sorption processes that take place over a range of pHs. However, an indication of the Rd will be shown on plots against pH for ease of comparison.

Ionic strength is an important factor controlling the behaviour of metal cations, such as their hydrolysis, and the distribution coefficients for their adsorption on mineral surfaces (Yoshida & Suzuki, 2006). Most field and laboratory experiments are conducted at low ionic strengths. However, only limited knowledge exists on the effects of ionic strength on the migration of cations in porous media (Yoshida & Suzuki, 2006). Apart from ionic strength, the ability of geologic materials to exhibit high sorption at low pH values will depend very much on the position of the point of zero charge (pH at which the net surface charge is zero) for the surfaces. Geologic materials such as granite and its minerals exhibit relatively low points of zero charge (USEPA, 1999). This work investigates the effect of pH on sorption at constant metal concentration, using and ^{152}Eu. The choice of ^{152}Eu has been discussed in earlier work by Ebong et al. (2026). Different granitic rocks and granitic minerals have been used in the sorption experiments in order to understand the sorption profile, and the sorption edge (pH at which there is a sharp rise in sorption with pH). Sorption parameters to quantify the sorption processes have been calculated, from the sorption data. Surface complexation modelling using the JChess geochemical code has been performed.

1.2. XRD for the Different Granitic Rocks

Quantitative powder XRD analyses are summarised in **Table 1**. Powder X-ray diffraction analysis indicated that the three granites had approximately similar min-

eralogies and were predominantly composed of quartz (mean *ca.*33%), plagioclase feldspar (mean *ca.*31%) and K-feldspar (mean *ca.*31%) together with small/trace amounts of "mica" (undifferentiated mica species possibly including muscovite, biotite, illite, illite/smectite, etc.). Small amounts of amphibole were also identified in the samples "Biotite granite" and "Rapakivi granite". Traces of chlorite, kaolinite and smectite were also identified in some of the samples.

Table 1. Summary of quantitative whole-rock XRD analysis.

	Mineralogical percentage composition							
Sample	**amphibole**	**smectite**	**chlorite**	**kaolinite**	**K-feldspar**	**"mica"**	**plagioclase**	**quartz**
Graphic Granite	nd	nd	nd	<0.5	49.4	0.5	21. 6	28.3
Granite Adamellite	nd	<0.5	<0.5	nd	32.9	3.1	25.7	38.1
Biotite Granite	2.9	<0.5	<0.5	nd	17.2	7.4	40.0	28.1
Grey Granite	nd	nd	<0.5	nd	22.6	4.3	34.4	38.6
Rapakivi Granite	3.5	<0.5	<0.5	<0.5	32.1	1.6	29.2	33.3

nd = not detected, "mica" = undifferentiated mica species including muscovite, biotite, illite and illite/smectite etc. (Wagner & Kemp, 2011).

1.3. Europium Speciation in Water from Low to High pH

JChess geochemical code (speciation programme) by Ecole de Mines de Paris France provided by the Department of Chemistry was used to study the variation of Eu species in solution in the absence of any complexing agents. Figure 1 shows the main species in solution between pH of 4 and 11. The pH range does not affect the net amount of Eu being sorbed but can change the mechanism of the sorption.

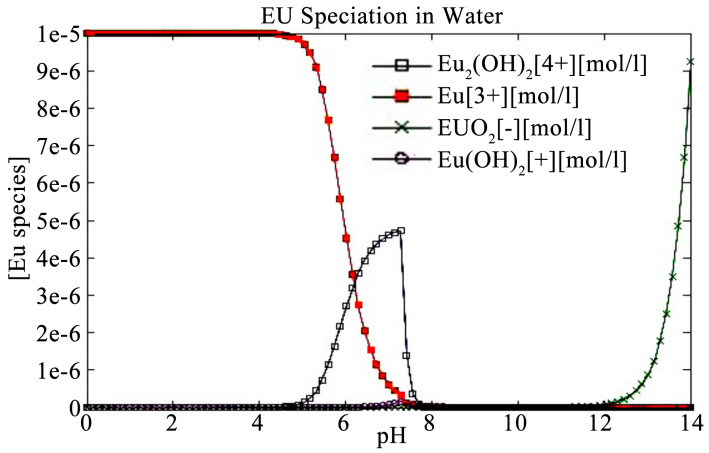

Figure 1. Eu speciation in DI water as calculated using JChess, geochemical code using the default JChess database, taking a cut-off concentration of 1×10^{-14} mol·dm^{-3} for species to be considered (van der Lee, 2003).

2. Experimental

To investigate the effect of ionic strength as well as pH, two sets of experiments, in the presence of 0.1 and 0.05 mol·dm^{-3} NaCl, were used with a control having no NaCl present. The effect of surface area has been taken into consideration in the surface complexation studies using the JChess geochemical code. NaCl is used to create a low concentration brine/seawater environment. With the possible disposal/storage of nuclear waste in deep geological repository, and with the potential rise in sea levels due to climate change, there is growing concern regarding the possible intrusion of brine into repository vaults. Thus, it is important to understand the effect of sea water on sorption of radionuclides in the far-field. For pH dependent sorption, 1×10^{-5} mol·dm^{-3} solutions of $EuCl_3$ were prepared, and the pH was adjusted from 4 to 11, with intervals of 0.6 to 0.8 on the pH scale using NaOH (aq) and HCl (aq). Different concentrations of pH adjusting solutions were made from very weak to very strong acid or base. This was important, so as to be able to vary the pH without significantly altering the concentration of the Eu solution. To move the pH to a lower pH value of about 4, a strong acid was preferred since very little volume of the acid was required thus, minimising the impact on the overall concentration of the solution. 0.1 g of every sample were weighed into a 20 cm^3 vial and 20 cm^3 of pH adjusted solutions added. 0.1 cm^3 of spike solution was added. After shaking the batch was allowed to equilibrate over a period of 5 to 7 days. The effect of ionic strength was investigated by the addition of different amounts of NaCl in the bulk $EuCl_3$ solutions to give ionic strengths of 0.05 and 0.1 mol·dm^{-3} NaCl solutions. Sample separation was performed as described above and counting was performed for Eu counting was performed based on its gamma emitting properties using Cobra II Auto Gamma. All experiment samples were carried out in triplicates and mean values used in processing the data used in the isotherm. The counts per vial for each sample did not vary by more than 1 percent.

3. Results and Discussion

3.1. Effect of pH on Sorption

Sorption reactions at solid-water interfaces decrease solute mobility and often control the fate, bioavailability and transport of radionuclides. Adsorption of metals in solution generally becomes more specific as the pH increases, i.e. formation of inner sphere complexes is favoured at elevated pH. Based on sorption experiments, the pH dependent sorption on mineral surfaces is usually modelled assuming two types of sites (Scheidegger et al., 1996):

1) Ion exchange, or nonspecific adsorption, sites that exchange background electrolyte cations with weakly bound hydrated metal ions (outer sphere complexes).

2) Specific adsorption at amphoteric surface hydroxyl sites, such (Al-OH, Si-OH) in which the surface sites hydrolyse and then bond directly to surface O or OH groups and are not easily displaced by electrolyte (inner sphere complexes)

(Scheidegger et al., 1996). This usually occurs at low sorbate concentrations. With increasing pH or sorbate concentrations, precipitation can occur. When a precipitate contains chemical species derived from both the solution and the dissolution of the sorbent mineral, it is referred to as a coprecipitate (Dähn et al., 2003).

3.2. ^{152}Eu Sorption to Granitic Rocks

To investigate Eu sorption to granitic rocks, varying pH sorption experiments were set up in different NaCl concentrations as described in the experimental section. Three sets of experiments were performed, to study the effect of pH and ionic strength on Graphic Granite (GG), Granite Adamellite (GA) and Rapakivi Granite (RG). Quantitative whole-rock XRD analysis of the rocks as in **Table 1** showed similarity in the major components of the rocks**.** Results have been plotted as concentration of metal bound vs. pH. Results for the three rocks are shown in **Figure 2**. The results show:

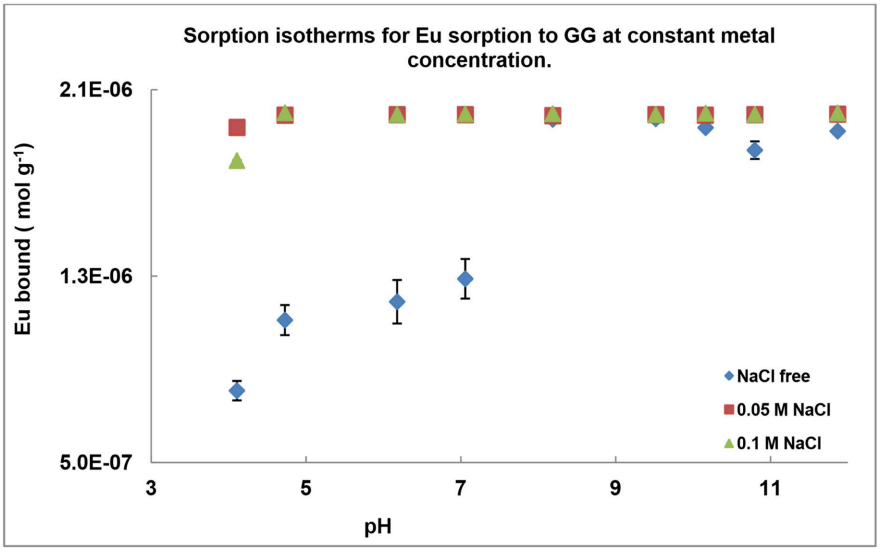

Figure 2. Varying pH sorption of Graphic Granite at constant metal concentration of Eu. Figure showing sorption profile of Eu in different ionic strength environments, equilibrating *ca.* 8 days with a liquid-solid ratio of 200:1, at rtp.

1) Varying pH had no effect on sorption in 0.05 and 0.1 mol·dm^{-3} NaCl (For composite rock samples) systems, however, net sorption for all Eu species in solution was higher than for NaCl free system as seen in **Figure 2**.

2) Sorption varied with pH below pH 7 for sorption in NaCl free system. Saturation was attained after pH 8.

3) Sorption in varying pH showed similar sorption profiles in both NaCl systems.

Because of the chemically heterogeneous nature of granite, the sorption behaviour is assumed to be the sum of the properties of the constituent minerals, following the generalised composite model (It assumes that a mineral assemblage is too complex to be described as a superposition of the individual phases). In this

approach, sorption is described using generic sites (notation: ≡SOH) and the values of the site densities and formation constants are obtained by fitting the experimental data (Wagner & Kemp, 2011; Tertre et al., 2008). Based on this assumption, it is possible to determine the proton stoichiometry for the entire granitic sample. Thus, the mass action law was applied to the sorption data. Sorption parameters are shown and the sorption edge.

From the results presented in **Figure 2**, it is evident that there was no region of significant rise in sorption with pH in the entire pH range studied. As such proton stoichiometry is not calculated for Eu sorption in 0.05 and 0.1 mol·dm^{-3} NaCl. However, for sorption in NaCl free solution, the proton stoichiometry is defined in the region of significant rise in sorption with pH as shown in **Figure 3**. Sorption parameters are shown in **Table 2**. It can be said that the sorption of Eu^{3+} on granite is complex with multiple surface species showing different behaviour (Ishida et al., 2009). Work by Li et al. (Li et al., 2017). has shown that sorption of Eu to granite was governed by sorption of Eu to feldspar present in the granite, and sorption was by inner and outer sphere complexation. However, based on the observation in **Figure 2**, Na^+ ions in solution enhanced the sorption of Eu below pH 7. One reason for this observation can be due to the modification of sorption sites. This modification of sites can lead to increased sorption at lower pH. Eu strongly hydrolysed at pH levels above 6, the strong hydrolysis makes the Eu fully hydrolysed at pH 8 to 9, and therefore exhibits a strong sorption on mineral surfaces (Baes & Mesmer, 1976). Due to the heterogenous nature of granitic rocks, different sorption mechanisms such as precipitation and co-precipitation reactions are usually considered the most important for mitigating radionuclide release from near-field. Sorption can be loosely defined as any process which results in the removal of a solute from solution by attachment of that solute to the surface of a solid phase. It includes processes such as: ion exchange, surface complexation, specific adsorption and physical processes (De Windt et al., 2004) precipitation, diffusion into dead-end pores, mineralisation, molecular filtration and isomorphous transition mineralisation. Sorption can be loosely defined as any process which results in the removal of a solute from solution by attachment of that solute to the surface of a solid phase this has been verified my work done by Ebong et al. (2026). **Table 2** shows Log K values derived after modelling the sorption using JChess.

Table 2. Eu sorption to granitic rocks at constant metal concentration and the application of the mass action law. LogK_e values are derived from the slope, at the region of steep rise in sorption with pH. Table shows that the Kurbatov model was not applicable as no sorption edge was defined.

Varying pH sorption of Eu to granitic rocks			
Sample	NaCl free (mol·dm^{-3})		
	SE pH	PS	LogK_e
Graphic Granite (GG)	7 - 8	1.3	−9.7
Granite Adamellite (GA)	7 - 8	1.58	−8.8
Rapakivi granite (RG)	7 - 8	1.2	−6.01

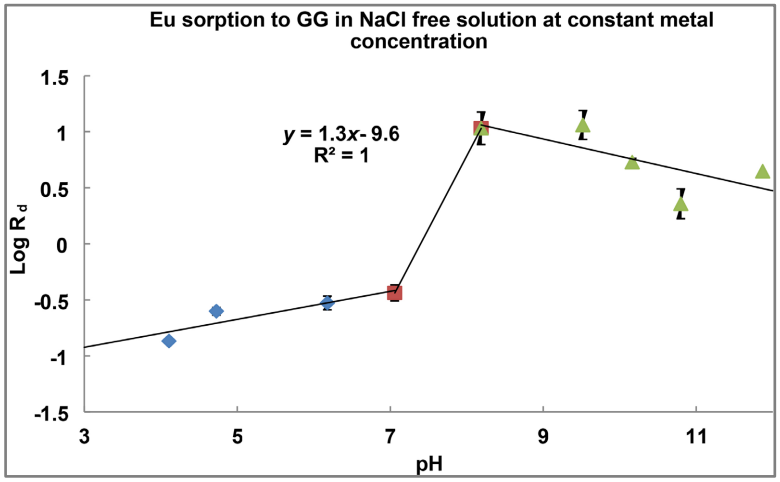

Figure 3. $LogR_d$-pH plot for Eu sorption to Graphic Granite at constant metal concentration in NaCl free solution. Figure shows region of steep rise in sorption within a narrow pH range (sorption edge), and region of gentle increase in sorption with pH. Equilibrating *ca.* 8 days with liquid solid ratio of 200:1, at rtp.

3.3. Eu sorption to Rose Quartz, Orthoclase Feldspar and Muscovite Mica

The interaction of quartz with Eu is of importance, particularly from the view point of deep geological storage/disposal. It is suggested that the Eu species in solution may sorb onto silica-water interfaces by inner sphere bidentate complexation (Hu et al., 1942). The importance of Eu sorption to mica cannot be overstated. Results for Am (usually substituted by Eu in the study of actinide retardation in the geomedia) sorption to granite showed that sorption was controlled by sorption to Biotite Mica (Ishida et al., 2009). The same studies showed that Eu sorption to granitic rocks was dominated by feldspar. To investigate the effect of ionic strength as well as pH, two sets of experiments, in the presence of 0.1 and 0.05 $mol{\cdot}dm^{-3}$ NaCl, were used with a control having no NaCl present.

JChess Geochemical code had various input data that is obtained by carrying out a set of other experiments to determine parameters such as the point of zero charge, BET surface area for each of the mineral phase studied input data can include volume of solution, pH of solution and Temperature. However, the complete application of the Code is beyond the work presented here. Figure 4 shows the comparison between the sorption profiles of modelled and experimental data for Eu sorption to Rose Quartz in NaCl free solution. Assuming mono and bidentate binding (for modelled data), and exchange capacity of quartz (0.06 $\mu mol{\cdot}m^{-2}$) determined from titrimetric experiments, Log K value (−2.5) for the sorption of Eu was described as shown the equations in Table 3.

Table 3. Non stochiometric equations of sorption sites and Log*K* values for Eu sorption to granitic minerals as modelled using the JChess Geochemical code.

Non stochiometric reactions of Eu and sorption sites	Log*K*	[NaCl]
$\equiv QuartzOH + Eu^{3+} \rightleftharpoons \equiv QuartzOEu^{2+}$	$LogK = -2.5$	none

Continued

≡ QuartzOH + Eu^{3+} ⇌ (quartz-O)$_2Eu^+$	Log$K$ = −2.5	none
≡FeldsparOH + Eu^{3+} ⇌ ≡FeldsparOEu^{2+}	Log$K$ = 4.5	none
≡FeldsparOH + Eu^{3+} ⇌ (≡Feldspar-O) $_2$ Eu^+	Log$K$ = 5	none
≡MuscoviteOH + Eu^{3+} ⇌ ≡MuscoviteOEu^{2+}	Log$K$ = 5.5	none
≡Muscovite OH + Eu^{3+} ⇌ (≡Muscovite-O)$_2$Eu	Log$K$ = 5.5	none

Based on Figure 4, it can be seen that the modelled data fitted to the experimental for the determined Log K values of −2.5 for mono and bi dentate binding. Application of the mass action law (Equation (3)) to the experimental data gave LogK_e value of −0.8. The difference between the equilibrium constants derived from the mass action law and the JChess, modelling can be as a result of experimental errors and purity of sample. Above pH 9 disagreements between the modelled data and experimental data were observed. Above pH of 9, Eu sorption decreased to 3.0×10^{-8} mol·g^{-1} for the model, while experimental data showed sorption to be high (1.5×10^{-6} mol·g^{-1}). The difference in the sorption could be as a result of precipitation at pH values above 9. Above pH 9, lanthanides (Eu) are fully hydrolysed, and they therefore exhibit a strong sorption on mineral surfaces (Baes, & Mesmer, 1976). The effect of NaCl on Eu sorption is shown in Figure 4, it can be concluded that Eu sorption is enhanced at pH < 9, in the presence of NaCl. Nordén (1994) used Eu as a tracer during the studies quartz and alumina. In the systems studied, K_d reached a value > 3 m^3/kg with quartz and alumina as solid phases (Nordén, 1994), highlighting strong Eu sorption at high pH values as also shown in this section.

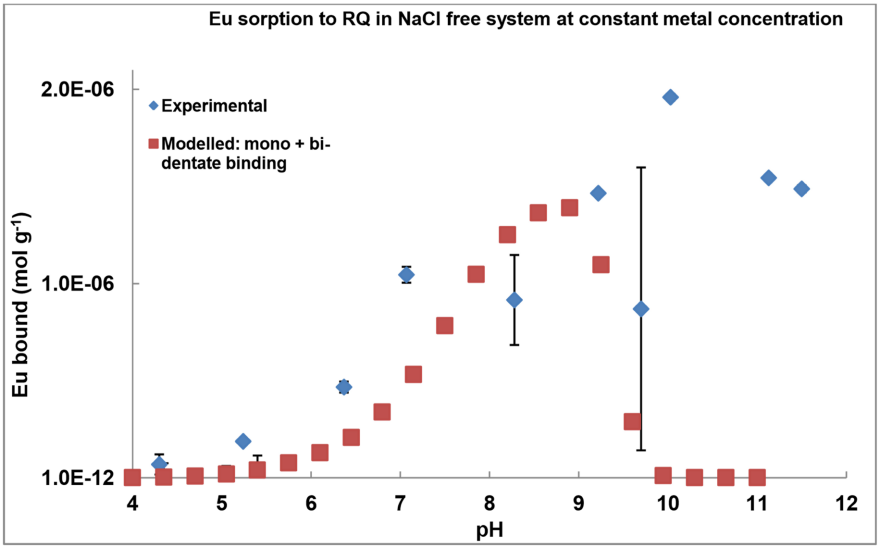

Figure 4. Varying pH sorption isotherms for Eu sorption to Rose Quartz. Figure showing data from experimental data and data obtained from JChess Code, using the same experimental conditions, modelling done assuming mono and bidentate complex formation, equilibrating *ca.* 8 days with a liquid-solid ratio of 200:1, at rtp.

Results for Eu sorption to feldspar are presented in the following section. As was the case with Eu^{3+} sorption to quartz, Eu sorption to feldspar is also enhanced in the presence of NaCl at pH < 7. Proton stoichiometry derived from the gradients of Log R_d pH plots gave values < 1 for Eu sorption in the three systems. PS values < 1 (pH < 7) show that no region of steep rise in sorption is defined. Mean R_d value (sorption at pH < 7) was determined as 574 $cm^3 \cdot g^{-1}$ for NaCl free system. However, for Eu sorption in the presence of NaCl, R_d values were calculated as 2.9 $cm^3 \cdot g^{-1}$ for 0.05 $mol \cdot dm^{-3}$ NaCl and 2.5 $cm^3 \cdot g^{-1}$ for 0.1 $mol \cdot dm^{-3}$ NaCl systems. The difference in R_d for NaCl free and NaCl systems is thus, an indication that sorption is affected by the presence of NaCl in the system. By fitting experimental data to that obtained from JChess modelling, it was possible to allocate Log K values for the binding of Eu to the mineral surfaces as shown below. Surface complexation, based on experimentally measured JChess input parameters showed that the modelled data fitted to the experimental data at pH < 9 for Eu sorption to feldspar in 0.1 $Mol \cdot dm^{-3}$ NaCl system (Figure 5).

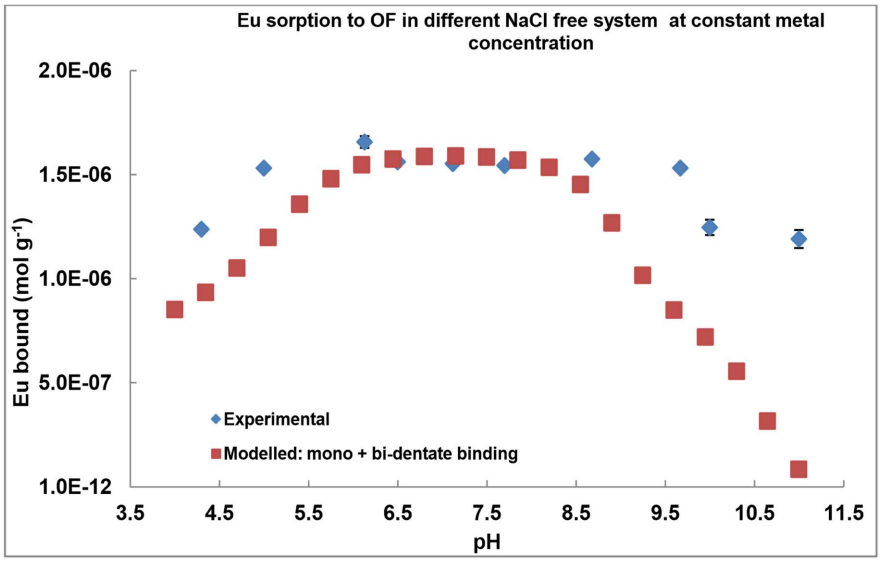

Figure 5. Varying pH sorption isotherms for Eu sorption to Orthoclase Feldspar. Figure showing data from experimental data and data obtained from JChess Code, using the same experimental conditions, modelling done assuming mono and bidentate complex formation equilibrating *ca.* 8 days with a liquid-solid ratio of 200:1, at rtp.

At pH > 9, results show that modelled data and experimental data did not agree. At pH > 9 experimental data showed sorption to be high (1.5×10^{-6} $mol \cdot g^{-1}$) as compared to 9.0×10^{-8} $mol \cdot g^{-1}$ for sorption in 0.1 $mol \cdot dm^{-3}$ NaCl. The reason for the observed difference between the two models can, as was the case for Eu sorption to quartz, attributed to purity of the experimental sample. The low fit between the experimental and modelled data can also be due to purity of sample and other sorption mechanisms that can be present such as intraparticle diffusion and ion exchange. Modelling the sorption process with JChess considered sorption to take place by surface complexation (double layer model) without taking into consider-

ation precipitation, diffusion into dead end pores, and cationic exchange. Fitting modelled data for Eu sorption in NaCl free and 0.05 mol·dm^{-3} NaCl to experimental data gave poor fits. Figure 6 shows that sorption in the presence of NaCl is constant and enhanced, as compared to that in NaCl free solution. One reason for the constancy in the sorption capacity from low to high pH can be due to the position on the point of zero charge (PZC pH at which the net surface charge is zero) on the pH scale.

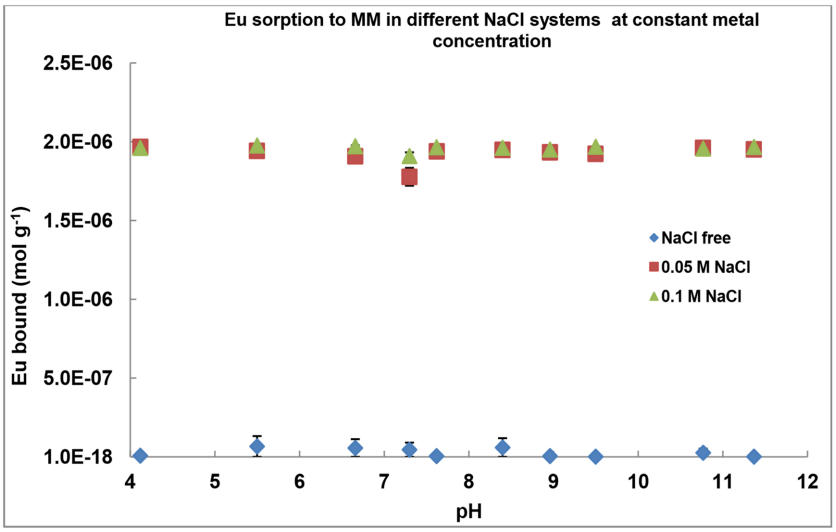

Figure 6. Varying pH sorption of Eu to Muscovite Mica at constant metal concentration of Eu. Figure showing sorption profile of Eu in different ionic strength environments, equilibrating *ca.* 8 days with a liquid-solid ratio of 200:1, at rtp.

Figure 7 depicts the effect of NaCl on Eu sorption. Eu sorption did not vary with the pH for Muscovite Mica, however, in NaCl free solution, sorption is reduced. Because no region of significant rise in sorption is defined from the Log Rd-pH plots as shown in the example shown below, the proton stoichiometry for the sorption process is not calculated. Mean R_d for Eu sorption in NaCl free solution was <900 cm^3·g^{-1} (880 cm^3·g^{-1} NaCl free system) while for Eu sorption on NaCl systems, the R_d was determined to be >10 cm^3·g^{-1}. Modelling the sorption process in a similar manner to the other granitic minerals showed that Eu sorption (in 0.1 mol·dm^{-3} NaCl) could be described thermodynamically for mono and bidentate binding as described below. At pH > 9 sorption capacity decreased as was observed for Eu sorption to quartz and feldspar. Modelled data obtained, for Eu sorption in NaCl free and 0.05 mol·dm^{-3} NaCl did not fit to the experimental data.

Experimental results obtained for Eu sorption to granitic rocks and minerals in different NaCl systems, showed that sorption was enhanced, in the presence of NaCl. Taking into consideration the three systems studied (NaCl free, 0.05 and 0.1 mol·dm^{-3}); theoretical models using Muscovite Mica, with exchange capacity of 70.4 μmol·m^{-2}, calculated particle radius of 2.8 nm, and Log K values of 5.5 for mono and bidentate complexation, particle radius were produced as shown in Figure 8. Results from the JChess models, show that sorption is enhanced in the

presence of NaCl. The reason for the enhanced sorption of Eu in the presence of NaCl is not very clear, however, Na^+ ions modify the sorption sites for more Eu to sorb. The perceived enhancement of Eu sorption as the concentration of NaCl increase can not only be attributed to the hydrolysis of the silonal sites, Because of the complexity of the ionic speciation the formation of not stoichiometric complexes between the ionic species and SO sites is possible leading to not just mono or bidentate complexes but also to metal complex deposition on the mineral surfaces.

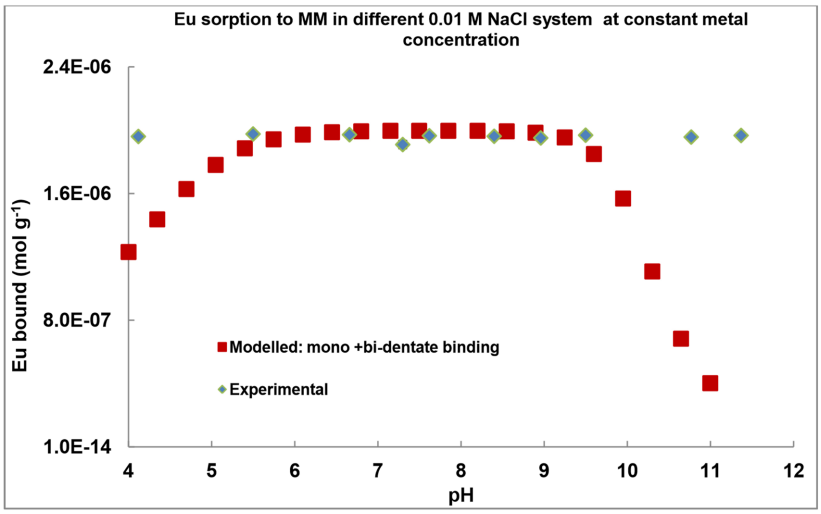

Figure 7. Varying pH sorption isotherms for Eu sorption to Muscovite Mica. Figure showing data from experimental data and data obtained from JChess Code, using the same experimental conditions, modelling done assuming mono and bidentate complex formation, equilibrating *ca.* 8 days with a liquid-solid ratio of 200:1, at rtp.

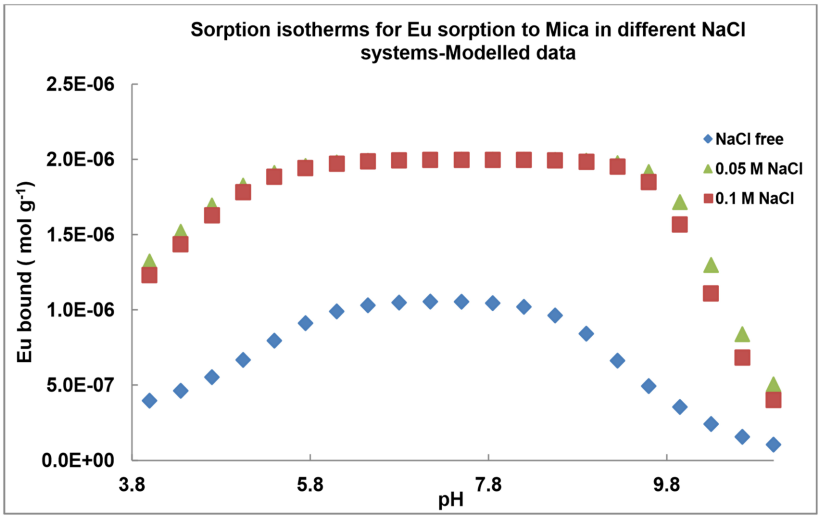

Figure 8. Modelled data for Eu sorption to Muscovite Mica in three different environments. Figure shows NaCl enhances the sorption of Eu from granitic materials studied using the same experimental, equilibrating *ca.* 8 days with a liquid-solid ratio of 200:1, at rtp. Exchange capacity of 70.4 μmol·m^{-2}, calculated particle radius of 2.8 nm, and Log K values of 7.1 and 8.1 for mono and bi dentate.

Work done by Bradbury, working on Sorption of Eu on Na- and Ca-montmorillonites showed that all of the measured sorption edge data could be modelled using cation exchange and the monodentate surface species, $\equiv SSOEu^{2+}$, $\equiv SSOEuOH^{+}$ and $\equiv SSOEu(OH)^{3-}$, on the strong site type. However, an additional modelling study showed that the same data were almost equally well described by considering bidentate surface complexes, $(\equiv SSO)_2Eu^{+}$ and $(\equiv SSO)_2Eu(OH)^{2-}$, and cation exchange (Bradbury & Baeyens, 2002) results obtained can fit into the conclusions based on the fact that Na based rocks are used and most sorption sites are the same.

4. Conclusion

The following conclusions can be made after studying varying pH sorption profiles of selected granitic rocks and minerals in different NaCl concentrations, using different models. Eu sorption is enhanced in the presence of NaCl. Surface complexation constants for experimental data have been determined by fitting modelled data to experimental data. The modelling approach can be included efficiently in transport simulations having chemical gradients in space and time (Goldberg et al., 2007).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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