Adsorption Behavior of Aqueous Europium on Kaolinite under Various Disposal Conditions

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Abstract−This work investigated the adsorption behavior of europium on kaolinite under various disposal conditions. Batch-wise adsorption and precipitation experiments and equilibrium model calculations were performed over a pH range of 4-10 and CO_2 concentration range of 0%, 0.03%, and 10%. Experimental precipitation behaviors are in agreement with the results of equilibrium model calculations using the geochemical code MINTEQA2. Aqueous species of Eu^{3+} exists mainly at pH 5 or below and solid phases of $Eu(OH)_3(s)$, $Eu(OH)CO_3(s)$, and $Eu_2(CO_3)_3 \cdot 3H_2O(s)$ are formed at higher pH ranges. Adsorption behavior of Eu on kaolinite in the low pH range can be explained by interlayer ion-exchange reaction. The significant increase in adsorbed amount at pH 5-6 is due to the surface complexation at the edge site of kaolinite. In the high pH range, precipitation of Eu contributes mainly to the adsorption quantity. The rapid decrease in adsorbed amount above pH 7 under 10% CO_2 condition occurs by the formation of anionic europium species of $Eu(CO_3)_2^-$. The adsorption of Eu on kaolinite could be well interpreted by the Freundlich adsorption isotherm. The data except for the highest equilibrium concentration ranges were also explained by Langmuir isotherm and the maximum adsorbed quantity of Eu on kaolinite, b, is 1.2 mg/g.

Key words: Europium, Species, Adsorption, Kaolinite

INTRODUCTION

Industrial or radioactive waste has been disposed of at an underground repository. If the hazardous elements in the disposed waste are exposed to groundwater, they can dissolve into water. There is, therefore, a potential for these hazardous elements to migrate and easily enter the terrestrial and aquatic environments. The migration behavior of the hazardous elements in the geosphere greatly depends on the chemical properties of elements as well as their interaction with surrounding solids [Bodek et al., 1988; Lieser, 1995; Park et al., 2002]. One of the chemical properties to retard the migration of the elements is their precipitation. The precipitation occurs by solid-phase formation of elements under the solubility limits in the groundwater. The solid-phase formation and solubility limits determine the species of the element in the aquatic systems [Nitsche, 1991]. The important interaction process to limit the transport of the elements is their adsorption on underground matrices. The species of the elements has an important role in the adsorption behavior. In addition, the adsorption behavior strongly depends on the surface properties of underground solids [Silva and Nitsche, 1995; Stumm, 1992].

Underground matrices are mainly composed of rock-forming materials, together with a smaller amount of oxides and clay minerals. In spite of their smaller quantity, oxide and clay minerals have larger adsorption potentials than rock-forming materials. For this reason, the studies on the adsorption of heavy metals and radionuclides on oxide and clay minerals have been identified as one of the important research works associated with waste disposal. Kaolinite is a typical clay mineral and it is composed of 1-1 type layers, having one silica sheet of tetrahedral structure and one gibbsite sheet of octahedral structure. Kaolinite has a heterogeneous surface. The permanent charge of the layer is generated by an isomorphous substitution. If Si and Al in the basal plane of kaolinite change into Al and Mg, the planes have negative charges and therefore cations become present between two layers. The charge on the crystalline edge is due to the protonation/deprotonation of exposed hydroxyl groups, such as AlOH and SiOH, of edge site [Sposito, 1989; van Olphen, 1977].

Actinide elements in the nuclear waste are of particular concern, since their half-lives are extremely long and in the range of 10^3-10^6 years. In addition, their chemical properties are very complex and not clearly known at present [Silva and Nitsche, 1995]. Investigation of their chemical behavior, therefore, is an essential research work in establishing the conditions that would ensure safe waste disposal. Chemical behaviors of actinides including solid-phase formation and adsorption are greatly influenced by the conditions of natural aquatic systems. These conditions comprise a wide variation of chemical parameters, e.g. pH from 4 to 9, Eh from −200 mV to +600 mV, CO_2 partial pressures from 0.03% to 10% ($10^{-3.5}$ atm. to 10^{-1} atm.), various ionic strength, etc. [Kim, 1986; Stumm and Morgan, 1996].

Many studies have been conducted on the adsorption behavior of actinide with oxide and clay minerals because of its environmental significance [Kohler et al., 1999; Jung et al., 1998; Lee et al., 2000; Kang et al., 2002]. However, the study of actinides has been limited in experimentally possible oxidation states. Generally, thermodynamic data already published are used for the evaluation of adsorption and migration behaviors. The analogue elements having similar chemical properties are also used for the experiments.

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Europium, one of the fission products of uranium, is used as an analogue for other trivalent actinides. The behavior of Eu is very similar to that of Am and Eu is also a toxic element [Choppin and Rizkalla, 1994].

This study investigated the adsorption behavior of Eu on kaolinite under various pHs and CO_2 partial pressures. The calculation of equilibrium Eu species and the experimental solid-phase formation were used to further understand the adsorption mechanism. The adsorption isotherm of Eu on kaolinite was also studied.

METHODS

The stock solution of 1×10^{-2} M europium was first prepared by using $Eu(NO_3)_3 \cdot 4H_2O$ (Aldrich Co., 99.9%). The 1×10^{-6} and 1×10^{-4} M Eu solutions used for adsorption experiments were then made by diluting the stock solution. $HClO_4$ and NaOH solutions of 0.1 M and 1 M concentrations were used for the pH adjustment. $NaClO_4 \cdot H_2O$ (Aldrich Co., 98%) was used for ionic strength adjustment. Source Clay Mineral Repository in U.S.A. supplied kaolinite used in this study. The supplied kaolinite, KGa-1, is well-crystallized kaolinite and has a structure of $(Mg_{0.02}Ca_{0.01}Na_{0.01}K_{0.01})$ $[Al_{3.86}Fe(III)_{0.02}Mn_{tr} Ti_{0.11}]$ $[Si_{3.83}Al_{0.17}]O_{10}(OH)_8$ and a surface area of $10 \, m^2/g$. Batch-wise adsorption experiments were performed at a fixed temperature of 25 °C under three different gaseous conditions of nearly zero%, 0.03%, and 10% CO_2 partial pressures. Experiments under nearly zero% CO_2 partial pressure, which was performed to simulate CO_2-free condition, were carried out in the inert-gas glove box. The glove box was filled with N_2 gas with 99.999% purity. Almost CO_2-free condition was verified by the analysis using GC (DID, HP Agilent). The CO_2 concentration in the glove box did not exceed 3.2 ppm. Atmospheric air condition is 0.03% CO_2 partial pressure. In order to simulate excess CO_2 condition, CO_2 partial pressure was maintained as 10%. 10% CO_2 gas with remainder consisting of N_2 was bubbled continuously through the Eu solution at a flow rate of about 10 ml/min. The concentration of Eu in the solution was fixed to be 1×10^{-6} and 1×10^{-4} M. The ionic strength of the solution was constant to the conditions of 0.01 and 0.1 M $NaClO_4$. Kaolinite was added to the prepared Eu^{3+} solution. The amount of kaolinite added to the 40 ml of Eu^{3+} solution was 0.04 g. It corresponds to the solid to solution ratio of 1 g/L. To investigate the pH effect on adsorption, pHs of solution were varied from 4 to 10. The adsorption time was 3 days. 5 ml of sample was taken from the reaction liquid at equilibrium, and the solutions and the precipitates were separated by 0.45 mm membrane filter (cellulose acetate, Corning 21053-25). Isotherm experiments were performed with varying the initial concentrations of Eu from 1×10^{-6} to 1×10^{-2} M. The amount of kaolinite and ionic strength were fixed as 0.04 g to 40 ml Eu^{3+} solution and 0.01 M $NaClO_4$, respectively. The concentration of Eu in the solution was analyzed by ICP-AES (ICPS-1000III, Shimadzu) and ICP-MS (PQ3, VG Elemental). The adsorbed concentration of Eu on kaolinite was determined from the concentration difference in the solutions before and after adsorption. The pHs of solutions before and after adsorption were measured by a combined glass electrode (Metrohm 6.0233.100). The conditions of precipitation experiments were controlled by the same methods as those of adsorption experiments.

Equilibrium speciation of aqueous and solid-phase europium at various pHs and CO_2 conditions was calculated by the computer

Table 1. Reactions of aqueous and solid europium species and stability constants used for equilibrium model calculations

Reactions	log K
Aqueous species	
$Eu^{3+}+H_2O=EuOH^{2+}+H^+$	−7.8
$Eu^{3+}+2H_2O=Eu(OH)_2^+ +2H^+$	−16.4
$Eu^{3+}+3H_2O=Eu(OH)_3+3H^+$	−25.2
$Eu^{3+}+CO_3^{2-}=EuCO_3^+$	7.9
$Eu^{3+}+2CO_3^{2-}=Eu(CO_3)_2^-$	12.9
$Eu^{3+}+H+CO_3^{2-}=EuHCO_3^{2+}$	12.43
Solid species	
$Eu^{3+}+3e^-=Eu(s)$	−163.2
$Eu^{3+}+3H_2O=Eu(OH)_3(s)+3H^+$	−15.1
$Eu^{3+}+3H_2O=Eu_2O_3(c)+6H^+$	−52.4
$Eu^{3+}+3H_2O=Eu_2O_3(m)+6H^+$	−54
$Eu^{3+}+H_2O+CO_3^{2-}=Eu(OH)CO_3(s)+H^+$	7.8
$2Eu^{3+}+3H_2O+3CO_3^{2-}=Eu_2(CO_3)_3 \cdot 3H_2O(s)$	35

code MINTEQA2 [Allison et al., 1991]. This code includes an extensive thermodynamic database and seven different algorithms for calculating adsorption. This geochemical model can also calculate precipitation-dissolution and solid-phase saturation states of metals. In this study, several complexation reactions of Eu with ligands and the stability constants of each reaction were added to this model. Complexation reactions of Eu with hydroxide, bicarbonate and carbonate ions considered in this calculation are summarized in Table 1. Complexation reactions of Eu with chloride and nitrate were excluded because the parameters of our experimental conditions were pH and carbonate concentration without other ligands. Aqueous species of europium hydroxide, bicarbonate and carbonate were considered in this calculation. Solid species such as europium, europium hydroxide, oxide, hydroxocarbonate and carbonate were also considered in the calculation. The stability constants for the formation of six aqueous species and six solid species are also listed in Table 1. The values of log K used in this calculation were cited from the report written by Spahiu and Bruno [1995].

RESULTS AND DISCUSSION

The adsorbed quantities of europium as a function of pH under 0% CO_2, air, and 10% CO_2 conditions are shown in Fig. 1. In the case of 0% CO_2 condition, the adsorbed Eu amount was over 90% of total Eu in the all pH ranges of 4-10. 90.4% of total Eu was adsorbed at pH 4.0 and the adsorbed quantity increased rapidly as the pH increased. Nearly 100% of total Eu was adsorbed at pH 5.5 or above. When the CO_2 condition was air, the adsorbed quantities were 83-85% at pH 5 or below and nearly 100% at pH 6 or above. The adsorbed quantity is constant in the low pH range. The pH edge was found in the pHs 5-5.5, at which range of pH, the adsorbed Eu increased rapidly as the pH increased. In the case of 10% CO_2 condition, the adsorbed amount of Eu increased as the increment of pH, this amount was almost 100% of total Eu at pH 6.3. The adsorbed amount decreased rapidly in the high pH range.

Typically, two parameters can affect the adsorption characteristics of metals on minerals. One affecting parameter is the specia-
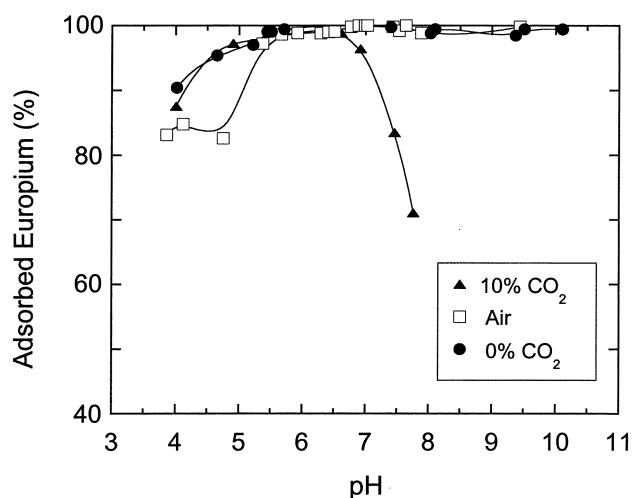
Fig. 1. The amounts of adsorbed Eu as a function of pH in the 1×10^{-6} M solution equilibrated under 0% CO_2, air, and 10% CO_2 conditions.

Fig. 3. Calculated distribution of Eu species in the aqueous and solid phase of 1×10^{-6} M solution equilibrated under air condition.

tion of metal and another parameter is the functional group of mineral surface [Stumn, 1992, 1987]. First, the chemical species of metal varies with pH of solution, Eh of solution, and type of counter ligand. The speciation is generally predicted by model calculation using thermodynamic data of metal reaction. For the prediction of Eu species, the works about the model calculations of aqueous phase Eu were performed [Ledin et al., 1994; Lee at al., 2000]. It is known that Eu exists as its most stable chemical species of Eu^{3+} ion under atmospheric conditions when the pH is not higher than 6. The hydroxide complex or the carbonate complex of Eu is mainly formed in the higher pH ranges. But the Eu speciation involving solid phase has not completely studied up to date.

In this study, the model calculation of Eu species including aqueous and solid phase was carried out under different CO_2 conditions. The results of Eu speciation in the 1×10^{-6} M Eu solution are also shown in Figs. 2, 3, and 4. Fig. 2 shows the distribution of Eu spe-
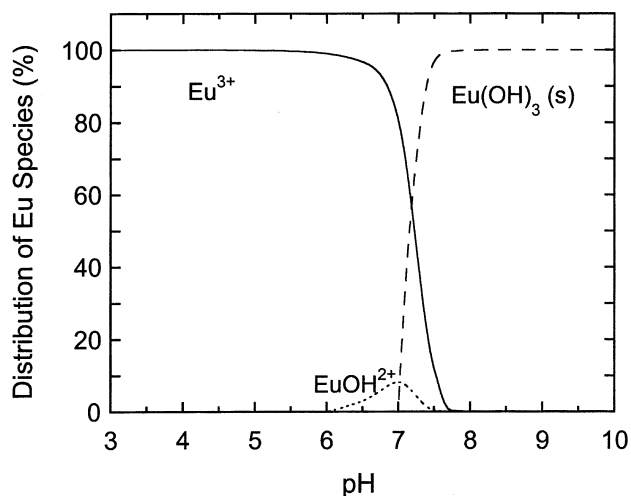

Fig. 4. Calculated distribution of Eu species in the aqueous and solid phase of 1×10^{-6} M solution equilibrated under 10% CO_2 condition.
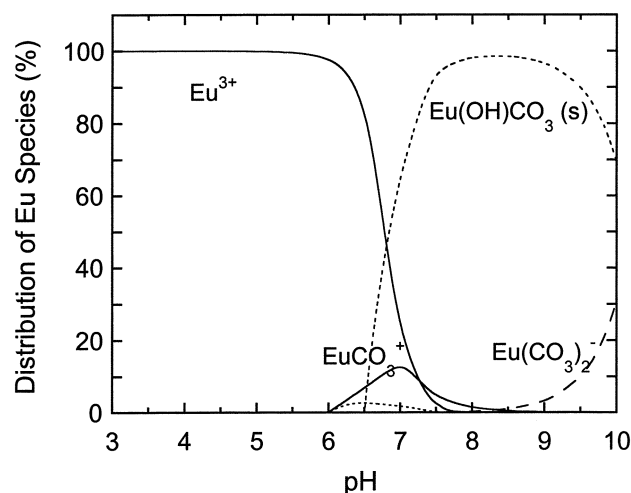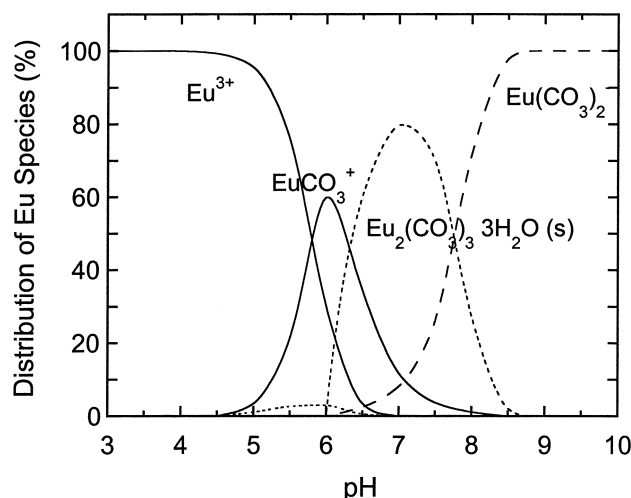
cies under 0% CO_2 condition. Europium exists as Eu^{3+} at pH 7 or below and is precipitated as $Eu(OH)_3(s)$ above pH 7. Fig. 3 is the equilibrium distribution of Eu species under air condition. The solid species of $Eu(OH)CO_3(s)$ is formed at pH>6.5. The amount of aqueous $Eu(CO_3)_2^-$ increases with pH when the pH of the solution is above 9. Fig. 4 is the calculated distribution under saturated CO_2 condition. This result is quite different from those of other CO_2 conditions. Eu exists as Eu^{3+} at pH 5.5 or below, $EuCO_3^+$ in the pH range of 5-7, and $Eu_2(CO_3)_3 \cdot 3H_2O(s)$ in the pH range of 6-8.5. And Eu exists as $Eu(CO_3)_2^-$ at pHs 8 or above. The solid species of $Eu_2(CO_3)_3 \cdot 3H_2O(s)$ is precipitated in the pH range of 6 and 8.5. It was found in our results of Eu speciation that aqueous species of Eu^{3+} exists mainly below pH 5 and solid phase of Eu is formed at a high pH range. Solid phases of Eu have different chemical forms because the carbonate concentration varies with CO_2 partial pressure. From the comparison of the calculated Eu speciation with the experimen-

Fig. 2. Calculated distribution of Eu species in the aqueous and solid phase of 1×10^{-6} M solution equilibrated under 0% CO_2 condition.

tal adsorption in Fig. 1, the dominant contribution of Eu adsorption on kaolinite in the low pH range below 6 is the adsorption by Eu^{3+} species. In the high pH range above 7, the solid phases of Eu hydroxide or carbonate may contribute mainly to the adsorption. In the particular case of 10% CO_2 condition, the adsorbed Eu amount decreased rapidly at pH 6.3 or above. This can be caused by the aqueous species of $Eu(CO_3)_2^-$. $Eu(CO_3)_2^-$ exists dominantly above pH 8 and this species with an anionic charge has no adsorption capacity with mineral surface.

The charge of mineral surface varies with the pH of solution and it also affects the adsorption characteristics. Especially, the adsorption reactions occur at the ion-exchange sites of 1-1 and 2-1 clay minerals such as kaolinite and montmorillonite. The ion-exchange site is due to the permanent charge of the layers of mineral. In the case of kaolinite, the cation exchange capacity (CEC) is around 2.0 meq/100 g and it strongly depends on the particle size and the pH value. However, the CEC of kaolinite entirely corresponds to the charge on edges and basal surfaces. The permanent charge of kaolinite is known to be less than 5% of the total CEC [Ma and Eggleton, 1999]. The hydroxyl groups, which are exposed to the edge of crystalline, contribute largely to the adsorption reaction. These aluminol (AlOH) and silanol (SiOH) groups are formed from the hydration of gibbsite and silica sheets. As the pH of solution increases, the deprotonation of the hydroxy groups is increased and then the charge on the edge site is increased. Therefore, the cation adsorption by the hydroxyl groups depends on the pH of solution and is called the surface complexation [Patrick et al., 1998; Dzombak and Morel, 1990].

It is known that the progress of adsorption reaction at ion-exchanging site is different from that at edge site of SiOH and AlOH structures [Hyun et al., 2000]. As shown in the results of adsorption in Fig. 1, the adsorbed quantity did not vary with pH in the pH range smaller than the pH edge. The adsorbed amounts maintained from 83% to 85% in the pH range of 4-5 under air condition. When the adsorption of metal on the clay mineral occurs by an interlayer ion-exchange reaction, this adsorption has a relation with electro-

static force and resultant adsorbed quantity is relatively small. The cations in the interlayer can be easily exchanged by Eu^{3+} and it is interfered with by the competitive reactions with other cations in the solution, such as Na^+ or Ca^{2+}, as shown in Fig. 5. Fig. 5 shows that adsorbed percentage in the condition of 0.1 M Na^+ is lower than in the condition of 0.01 M Na^+ in both of 1×10^{-6} and 1×10^{-4} M Eu^{3+} adsorption experiments. The adsorbed amounts were constant in the pH range smaller than the pH edge. In addition, the significant increase of adsorbed quantity at pH range of 5-6 is found in the 1×10^{-6} M of Eu initial concentration as shown in Fig. 5. In the case of 1×10^{-4} M Eu concentration, the adsorbed amount increased rapidly in the range of 6-8. This must have resulted from the surface complexation between Eu^{3+} and hydroxyl groups of mineral surface. As the pH of the solution increases, surface complexes are formed by the surfaces of negative charges with chemical species of Eu.

To evaluate the contribution of solid phases of Eu to adsorption behavior, the Eu precipitation at various pHs and CO_2 partial pressures was investigated. The amounts of Eu precipitate in the 1×10^{-6} M Eu solution as a function of pH are shown in Fig. 6. In the case of 0% CO_2 condition, the 20-50% of total Eu was precipitated at pH 7 or below and nearly 100% of total Eu was precipitated at pH 8.5 or above. When the CO_2 condition was air, the precipitated Eu amount was below 10% at pH<5. The precipitated Eu amount increased sharply at pH 5-8 and then reached nearly 100% at pH> 8.5. When the CO_2 partial pressure was 10%, the precipitated Eu amount increased and then decreased rapidly with the increment of pH. The maximum precipitated amount of Eu was 94% of the initial Eu and it was found at pH 6.6. Comparing these experimental results with equilibrium distribution of Eu species in Figs. 2-4, the precipitated Eu amounts agree well with the calculated distribution of solid Eu species such as $Eu(OH)_3(s)$, $Eu(OH)CO_3(s)$, and $Eu_2(CO_3)_3 \cdot 3H_2O(s)$. In the case of air condition, solid Eu species is nearly 0% at pH<6 but is nearly 100% at pH>8. The pH range of rapid formation of solid Eu species is from pH 6 to 8. When the experimental CO_2 condition is 10%, the precipitated Eu increases and then decreases with the increment of pH. The aqueous species of $Eu(CO_3)_2^-$

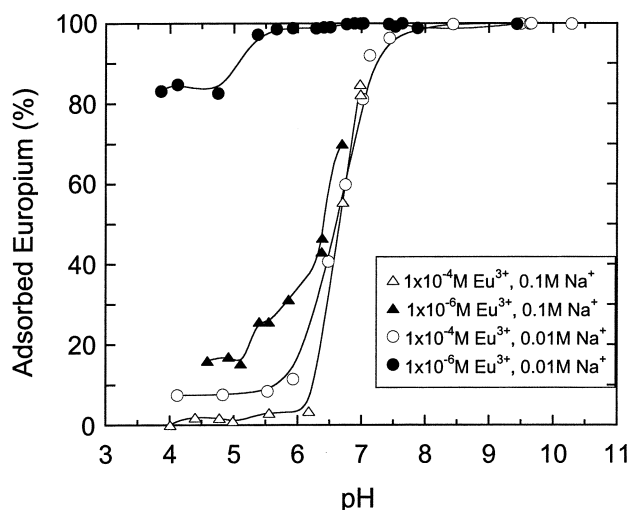**Fig. 5. The amounts of adsorbed Eu as a function of pH in the 1×10^{-6} and 1×10^{-4} M solutions under 0.01 and 0.1 M Na^+ ionic strength.**

Legend:
- △ 1×10^{-4} M Eu^{3+}, 0.1 M Na^+
- ▲ 1×10^{-6} M Eu^{3+}, 0.1 M Na^+
- ○ 1×10^{-4} M Eu^{3+}, 0.01 M Na^+
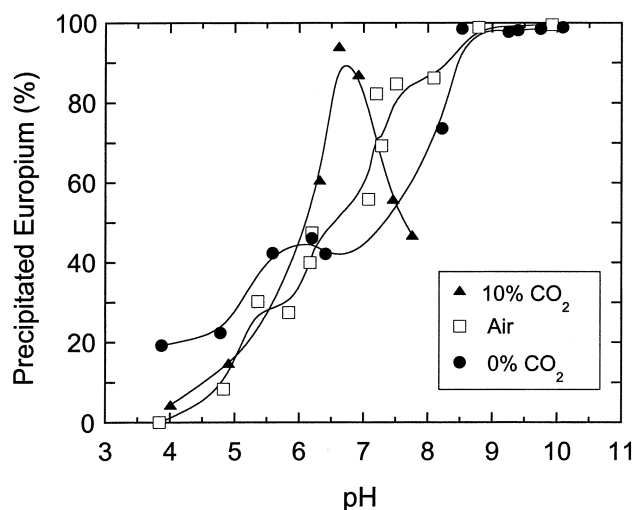- ● 1×10^{-6} M Eu^{3+}, 0.01 M Na^+

Fig. 6. The amounts of precipitated Eu as a function of pH in the 1×10^{-6} M solution equilibrated under 0% CO_2, air, and 10% CO_2 conditions.

Legend:
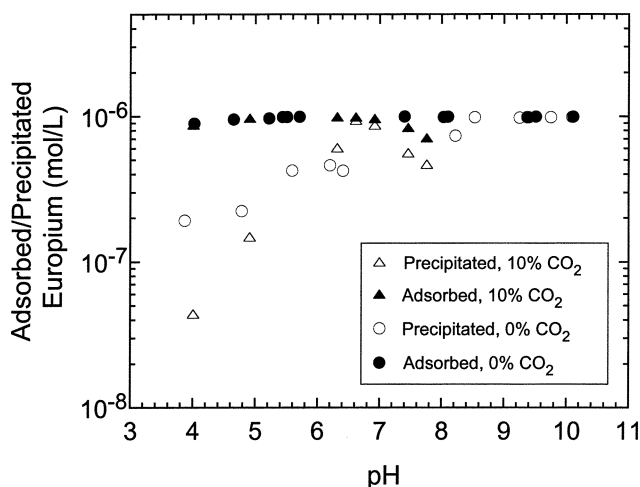- ▲ 10% CO_2
- □ Air
- ● 0% CO_2

Fig. 7. The absolute concentration of adsorbed or precipitated Eu as a function of pH in the 1×10^{-6} M solution equilibrated under 0% and 10% CO_2 conditions.

exists predominantly above pH 8. This result also agrees well with the calculated distribution of solid Eu species in Fig. 4. But the precipitated Eu amount by experiments is relatively higher than its equilibrium distribution in the low pH range under 0% CO_2 condition.

The absolute concentration of adsorbed Eu on kaolinite also was compared with that of precipitated Eu in Fig. 7. The adsorbed concentration is much higher than the precipitated concentration in the low pH range. However, the adsorbed concentration and the precipitated concentration are similar in the high pH range. In the case of 0% CO_2 condition, the excess adsorbed concentration at pH 4-8 is caused by ion-exchange reaction or surface complexation of Eu on kaolinite as mentioned above. In the pH range of 8 to 10, the adsorbed concentration is mainly contributed by the precipitation of europium. In the case of 10% CO_2 condition, the complexation reaction on the surface of kaolinite and precipitation of europium occurs at pH 4-6 and pH 6-8, respectively. The precipitated species are predicted $Eu(OH)_3(s)$ and $Eu_2(CO_3)_3\cdot3H_2O(s)$ in the 0% and 10%

conditions, respectively.

The results of adsorption experiments performed by varying Eu^{3+} concentration at fixed kaolinite concentrations are described in Fig. 8. The Eu concentration adsorbed per 1 g of kaolinite is plotted against the equilibrium concentration of in the Eu solution. The pH of the solution was in the range of 5.5-9.3. As the equilibrium concentration of Eu increased, the adsorbed Eu concentration increased linearly in the low range of equilibrium Eu concentration. In the equilibrium concentration range from 1×10^{-7} mol/L to 5×10^{-4} mol/L, the adsorbed concentration was nearly constant. The linear increase of adsorbed concentration occurs again in the high equilibrium concentration range. All these data from the experiment could be described by Freundlich-type isotherm [Kim et al., 2002]. The equation of Freundlich isotherm at equilibrium is expressed as follows:

$$q=K_FC^n$$

where, q is the adsorbed quantity of Eu onto unit mass of adsorbent (mol/g) and C is the Eu concentration at equilibrium (mol/L). K_F and n are empirical constants, K_F is Freundlich constant and n represents the degree of linearity. The constants K_F and n, are determined to be 5157 and 0.30, respectively, where r^2 of the relative coefficient is 0.967. These experimental adsorption data could be interpreted by Langmuir isotherm [Kim et al., 2002]. This is represented as follows:

$$q=bK_LC/(1+K_LC)$$

where, q is the adsorbed quantity of Eu on adsorbent (mol/g), C is Eu equilibrium concentration (mol/L), K_L is the equilibrium constant of the adsorption reaction, and b is the maximum adsorbed quantity of Eu on adsorbent. The constants K_L and b could be calculated from the linear equation $1/q=1/b+1/K_LbC$ and are determined to be 2.92×10^7 and 7.2×10^{-6}, respectively, where r^2 is 0.973. The maximum concentration of adsorbed Eu corresponds to 7.2×10^{-3} mmol/g. As shown in Fig. 8, all experimental data are well fitted with Freundlich isotherm. In the case of Langmuir isotherm, a deviation occurs at high equilibrium concentration ranges above 9.7×10^{-4} mol/L. This deviation corresponds to the sharp increase of the
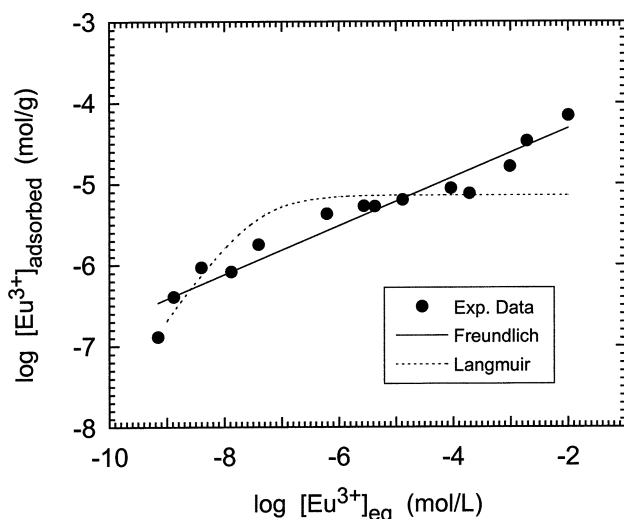

Fig. 8. Adsorption isotherm of 1×10^{-6} M Eu solution on kaolinite under air condition.
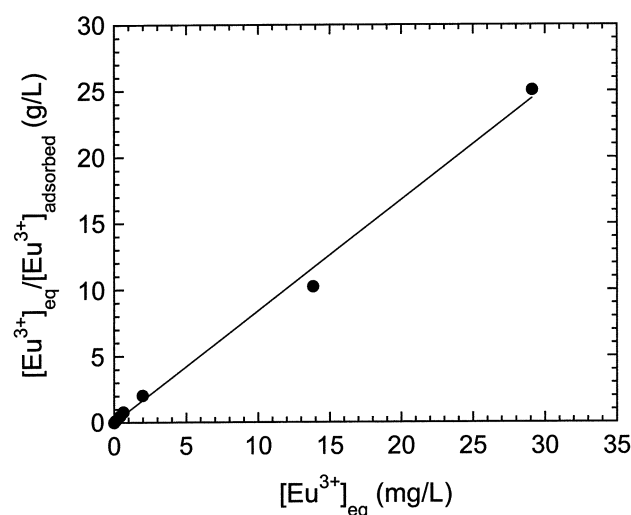

Fig. 9. The linear relationship of equilibrium Eu and equilibrium Eu/adsorbed Eu by Langmuir adsorption isotherm.

adsorbed concentration after zero slope range of adsorbed concentration. This increment is caused by the surface precipitation of metals having a high concentration [Dzombak and Morel, 1990].

In order to compare the adsorption capacity of kaolinite with that of other minerals, the Langmuir equation is converted to the equation $C/q=1/K_Lb+C/b$ and the constants K_L and b could be calculated from the linear relationship as shown in Fig. 9. The determined K_L and b as an mg/g unit are 48.3 and 1.2 mg/g, respectively. From these results, the maximum adsorbed amount of Eu on kaolinite is found to be extremely small compared to that on illite or montmorillonite, which have the maximum adsorbed Eu amounts of 16 mg/g or 48 mg/g, respectively [Lee et al., 2000].

CONCLUSION

It was found from the results of adsorption of europium on kaolinite at various pHs and CO_2 partial pressures that the adsorbed amounts at pH 5 or below were relatively small under 0% CO_2 and air conditions. And then, the adsorbed Eu reached nearly 100% of initial Eu at pH 6 or above. In the case of 10% CO_2 condition, the adsorbed Eu decreased rapidly as the increment of pH in the high pH range above 7. From the results of equilibrium model calculations, Eu exists as Eu^{3+} below pH 5 and it forms solid phase of europium hydroxide or carbonate above pH 7. The estimated solid Eu species are $Eu(OH)_3(s)$, $Eu(OH)CO_3(s)$ and $Eu_2(CO_3)_3 \cdot 3H_2O(s)$ under 0% CO_2, air, 10% saturated CO_2 conditions, respectively. In the case of 10% CO_2 condition, the aqueous phase of $Eu(CO_3)^{2-}$ exists predominantly above pH 8. The results of precipitation experiments are in agreement with equilibrium model calculations. Adsorption behavior of Eu in the low pH ranges can be explained by interlayer ion-exchange reaction. The significant increase of adsorbed amount at pH 5-6 is due to the surface complexation of Eu on the edge site of kaolinite. In the high pH ranges, the precipitation of Eu contributes mainly to the adsorption quantity. The rapid decrease of adsorbed amount at pHs 7 or above in the 10% CO_2 condition occurs by the formation of anionic europium species of $Eu(CO_3)_2^-$. The adsorption of Eu on kaolinite could be well interpreted by the Freundlich adsorption isotherm. The data except for the highest equilibrium concentration ranges were also in agreement with the Langmuir isotherm.

ACKNOWLEDGMENT

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