

Sorption and desorption of radiocobalt on montmorillonite—Effects of pH, ionic strength and fulvic acid

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Abstract

Humic substances and clay minerals have been studied extensively in radioactive waste management. In our research, the sorption and desorption of radiocobalt on montmorillonite in the presence and absence of fulvic acid as a function of pH and ionic strength were investigated under ambient conditions by using batch techniques. The results indicate that the sorption of cobalt is strongly dependent on pH values and independent of ionic strength. Surface complexation rather than cation exchange is considered as the main mechanism of cobalt sorption to montmorillonite. The presence of fulvic acid enhances cobalt sorption obviously at pH values < 8. The desorption behavior changes of surface-sorbed cobalt from montmorillonite were studied by decreasing pH values and the solution cobalt concentrations independently. The results indicated that the sorption of cobalt on montmorillonite is irreversible.

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Keywords: Montmorillonite; Radiocobalt; Sorption; Desorption

1. Introduction

The important radionuclides ⁶⁰Co and ⁵⁸Co are present in liquid wastes released from pressurized water nuclear power reactors, and ⁶⁰Co is also widely used in research and medical applications (Xu et al., 2006a; Yu et al., 2006). The sorption of radiocobalt on a mineral surface changes the physiochemical properties of radiocobalt and thereby controls the migration and diffusion of radiocobalt in the natural environment. The sorption of radiocobalt by clay minerals or oxides has been studied extensively (Tewari et al., 1972; Tewari and Lee, 1975; Eriksen et al., 1999; Nagy and Kónya, 1998). Tewari et al. (1972) and Tewari and Lee (1975) found that the sorption of Co²⁺ on Fe₃O₄, Al₂O₃, MnO₂, TiO₂ and ZrO₂ increases markedly with the solution pH increase between 5 and 8. Eriksen et al. (1999) studied the sorption of Co²⁺ on bentonite and found that Co²⁺ forms outer-sphere complexes with the permanent layer sites. Nagy and Kónya (1998) studied the sorption of cobalt on montmorillonite and found that Co²⁺ forms

strong complexes of Co–EDTA. Yu et al. (2006) found that sorption of cobalt on bentonite is strongly dependent on pH values and weakly dependent on ionic strength. Little effect of FA on cobalt sorption was found at pH < 6; a positive effect of FA at pH 6–8; and a negative effect of FA at pH > 8.

It is well known that humic substances (HSs) influence the sorption and complexation of metal ions onto the surfaces of clays and oxides (Wang et al., 2004a, 2006; Xu et al., 2006b; Chen and Wang, 2007a, b; Tan et al., 2007a). Generally, the presence of humic substances enhances the sorption of metal ions at low pH range and then decreases the sorption at high pH range, as compared with the sorption in the absence of HS. The increase of sorption is interpreted by the sorption of HS onto the mineral surface, followed by the interaction of metal ions with surface-adsorbed HS, whereas the reduction of sorption is explained by the formation of soluble M–HS complexes, which stabilize the metal ions in the aqueous solution (Montavon et al., 2002; Wang et al., 2005a; Takahashi et al., 1999).

Based on the results mentioned above, one can see that the sorption mechanisms of Co(II) on montmorillonite are

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not yet sufficiently conclusive. Herein, the sorption and desorption of Co(II) on bare and fulvic acid-bound montmorillonite were studied as a function of FA, pH and ionic strength. The sorption mechanism is discussed in detail.

2. Experimental

2.1. Materials

All chemicals were purchased as analytically pure and used without any purification. The sample of montmorillonite was derived from Lin'an county (Zhejiang, China) and was well characterized. The main components of the montmorillonite are listed in Table 1. The water content of 9.5% was determined. The N₂-BET surface area is 38.5 m²/g. The cation exchange capacity is 0.89 meq/g, and the point of zero charge (pH_{pzc}) is 7.8 ± 0.1.

Fulvic acid was extracted from weather coal of Gongxian (Henan Province, China) and had been applied in sorption experiments by other authors (Wang et al., 2003, 2004b).

⁶⁰Co(II) was used as radiotracer and the concentration of ⁶⁰Co(II) was analyzed by liquid scintillation counting (Packard 3100 TR/AB Liquid Scintillation analyzer, Perkin–Elmer) with an ULTIMA GOLD AB™ (Packard) scintillation cocktail. All the experimental data were the averages of duplicate or triplicate determinations.

2.2. Procedures

All the sorption and desorption measurements were carried out at $T = 20 \pm 2$ °C under ambient conditions using batch technique. The stock solution of montmorillonite, KNO₃ solution, cobalt stock solution, HNO₃ or KOH and FA stock solution were added in the polyethylene test tubes to achieve the desired background electrolyte concentration, FA concentration and pH of the aqueous solutions. The pH values were adjusted with negligible amounts of 0.1 or 0.01 M HNO₃ or KOH.

For desorption experiments, two methods were used to investigate the desorption of Co(II) from the solid to liquid phase:

- Method 1: The suspension of montmorillonite was centrifuged (7500 rpm, 20 min; the same method to separate the solid from liquid phases as in the sorption experiments) at the end of the sorption experiments; half of the supernatant was pipetted out and an equal volume of background electrolyte solution with the

same pH value was added. Then the mixture was shaken and centrifugation was done under the same conditions as in the sorption experiments.

- Method 2: The sorption experiments were carried out at high pH values (herein the pH was 7.7 ± 0.1); then the pH of the suspension was adjusted to low pH values (herein the pH was 7.3 ± 0.1) (it is necessary to note that to each tube was added the same negligible amount of 0.01 M HNO₃ and the pH of the suspension in each tube decreased to 7.3 ± 0.1). The desorption isotherm was then compared with the sorption isotherm at the same pH values (i.e., 7.3 ± 0.1 ; the sorption at pH 7.3 was carried out after the desorption tests (Method 2) to compare the desorption of Co(II) from pH 7.7 to 7.3).

3. Results and discussion

3.1. Characterization of montmorillonite

Fig. 1 shows the XRD pattern of montmorillonite sample. The d_{001} peaks of the sample are in the range of $2\theta < 10^\circ$. The basal spacing value of the sample is 14.9 Å. The XRD analysis shows that the sample is composed of sodium aluminum silicate calcium potassium hydroxide hydrates (JCPD standards: $2\theta = 7.13^\circ, 7.43^\circ, 18.68^\circ, 21.57^\circ, 26.21^\circ$ and 28.80°).

The FTIR spectra are shown in Fig. 2. The broad band at 3628 cm^{-1} are due to the O–H stretching vibration on the surface. The spectral band at 1636 cm^{-1} reflects the bending HO–H bond of water molecules, which is retained in the silica matrix. The strong band at 1038 cm^{-1} represents the Si–O–Si groups of the tetrahedral sheet. The spectral band at 915 cm^{-1} reflects the stretching vibration of Al–O(OH)–Al. The bands at 522 and 468 cm^{-1} are due to the deformation and bending modes of the Si–O bond. The FTIR spectra also suggest that

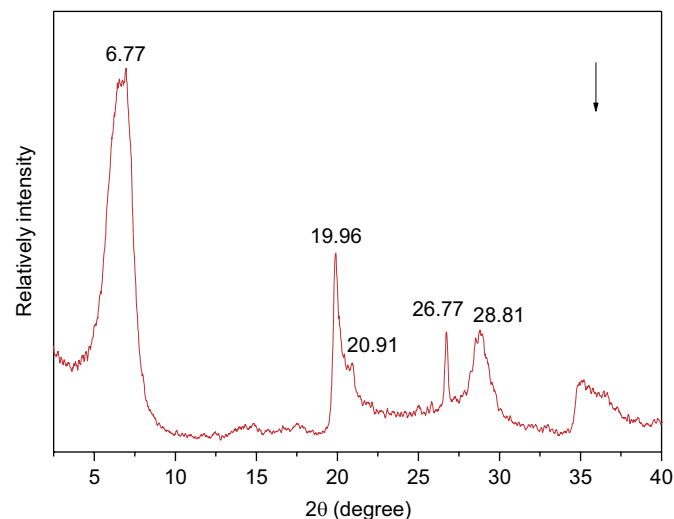


Fig. 1. XRD spectra of montmorillonite sample.

Table 1
The main chemical component of the montmorillonite

| Component | Al ₂ O ₃ | SiO ₂ | MgO | Fe ₂ O ₃ | K ₂ O | Na ₂ O | CaO |
|-----------|--------------------------------|------------------|-----|--------------------------------|------------------|-------------------|-----|
| % | 15 | 69 | 3.3 | 2.0 | 0.9 | 0.2 | 0.1 |

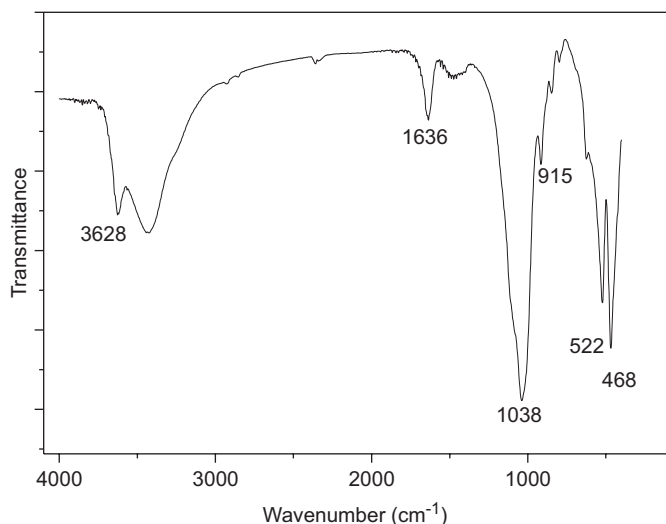


Fig. 2. FTIR spectra of montmorillonite sample.

aluminum silicate and water molecules are the components of montmorillonite.

3.2. Effect of contact time

The time dependence of sorption kinetics of Co^{2+} on montmorillonite is shown in Fig. 3. The sorption of cobalt on montmorillonite mainly occurred in the first 10–15 h of contact time and then a steady value of sorption was achieved. Compared with the sorption of Co^{2+} to bentonite (Yu et al., 2006), the sorption equilibration was very similar to that on bentonite. However, Dong et al. (2000) found that sorption of $\text{Co}(\text{II})$ on kaolinite was rather slow and an abnormal sorption shape was found. However, Dong et al. (2000) did not give any interpretation for the abnormal sorption shape. Herein, 48 h of equilibration time was selected in the following experiments according to the time-dependent sorption.

3.3. Effect of pH

Fig. 4 shows the pH dependence of cobalt sorption on montmorillonite in 0.01 M KNO_3 solutions. A steep increase of sorption on bare montmorillonite was observed at pH values ranging from 6 to 9. The step increase in sorption is a typical behavior for the sorption of hydrolysable transition metal ions on montmorillonite surface (Davis, 1982). CoOH^+ is formed and the adsorption process is rapidly completed by interaction with the negative charge on montmorillonite, but montmorillonite also absorbs cobalt ions in the sheets of hydroxyl groups (Yu et al., 2006; Yavuz et al., 2003):

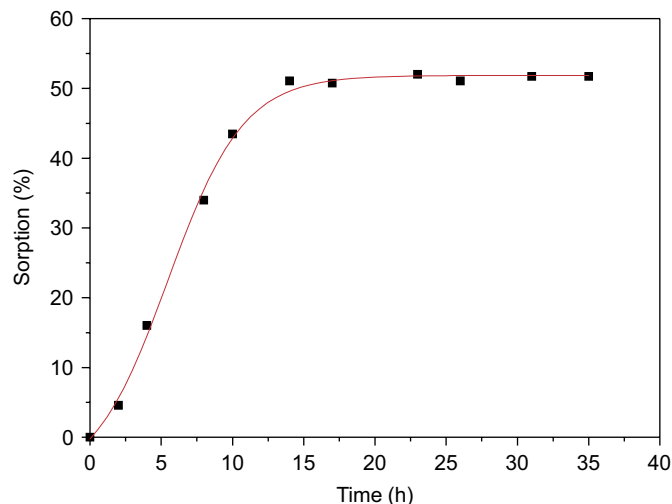
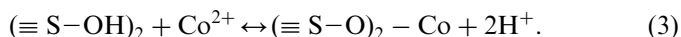
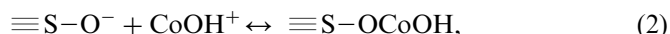
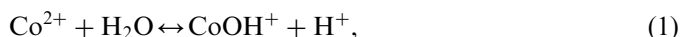


Fig. 3. Time-dependent sorption of $\text{Co}(\text{II})$ on montmorillonite. pH 7.6, $m/V = 1.6 \text{ g/L}$, $3.3 \times 10^{-4} \text{ mol/L}$, $C(\text{KNO}_3) = 0.01 \text{ mol/L}$.

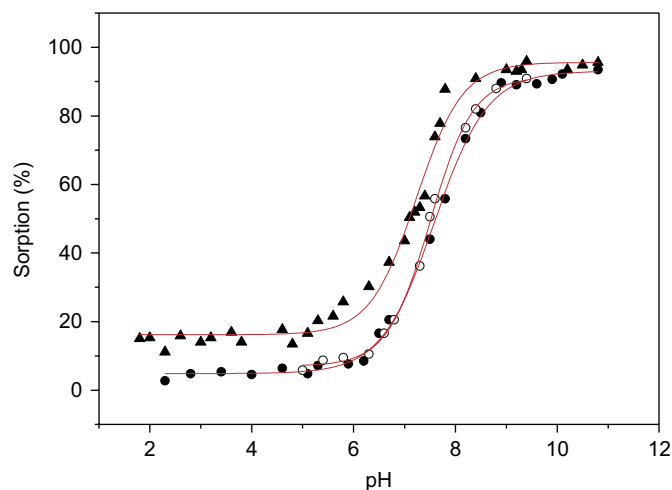
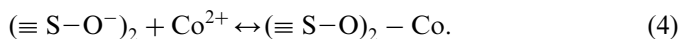


Fig. 4. Effect of pH on the sorption of $\text{Co}(\text{II})$ on montmorillonite. $m/V = 1.6 \text{ g/L}$, $3.3 \times 10^{-4} \text{ mol/L}$, $C(\text{KNO}_3) = 0.01 \text{ mol/L}$. (▲) Presence of FA; (●) absence of FA, $C(\text{KNO}_3) = 0.01 \text{ mol/L}$; (○) absence of FA, $C(\text{KNO}_3) = 0.1 \text{ mol/L}$.

The montmorillonite surface may also be negatively charged, and thus provide sorption sites for cobalt ions:



Tewari et al. (1972) and Tewari and Lee (1975) studied the sorption of cobalt on oxides and found that the sorption increases markedly from pH 5 to 8. Such pH-dependent sorption indicates that surface complexation is the main mechanism of $\text{Co}(\text{II})$ sorption on montmorillonite (Wang et al., 2005b,c; Baeyens and Bradbury, 1997; Takahashi et al., 2002; Chen et al., 2007a).

The pH dependence of cobalt sorption on fulvic acid-bound montmorillonite is also shown in Fig. 4. The initial concentration of FA in the suspension was 8 mg/L, and the UV-vis spectrophotometric analysis indicated that ~95% FA was adsorbed on montmorillonite in our experiments.

It can be seen from Fig. 4 that the presence of FA enhances the sorption of Co(II) on montmorillonite at pH 2–9. At the pH values, the negative charged FA is easily adsorbed on the positive charged montmorillonite surface, and thereby the surface-adsorbed FA can form strong complexes with Co(II) on montmorillonite. The interaction of cobalt ion with surface-adsorbed FA is stronger than that of cobalt with bare montmorillonite. Therefore, the presence of FA enhances the sorption of Co(II) in the ternary Co–FA–montmorillonite system.

Many possible modes of humic substances sorption on oxides or clays have been described (Tan et al., 2007b; Chang et al., 2007; Gu et al., 1994). The presence of FA in solution affects the sorption of cobalt on montmorillonite, and the species of cobalt on FA-bound montmorillonite is mainly dominated by both FA and montmorillonite; the formation of ternary complexes such as $\equiv\text{S}-\text{O}-\text{Co}-\text{FA}$ or $\equiv\text{S}-\text{O}-\text{FA}-\text{Co}$ occurs on the surface of fulvic acid-bound montmorillonite (Takahashi et al., 2002; Wang et al., 2004c). Microscopic investigation to study the possible species of Co(II) on bare or FA–montmorillonite and the macromolecular structure of FA is necessary.

3.4. Effect of ionic strength

Fig. 5 shows the dependence of $\log K_d$ of cobalt sorption on bare montmorillonite as a function of KNO_3 concentration. As can be seen from Fig. 5, the $\log K_d$ values were almost independent of ionic strength under our experimental conditions, although the $\log K_d$ values were slightly higher at very low KNO_3 concentrations. One can draw the conclusion that the sorption of cobalt on bare montmorillonite is insensitive to the ionic strength. The results of this work are very similar to those of cobalt sorption on kaolinite and bentonite (Yu et al., 2006; Dong et al., 2000).

The sorption of Co(II) on montmorillonite in 0.01 and 0.1 M KNO_3 solutions, respectively, at the pH range 5–10

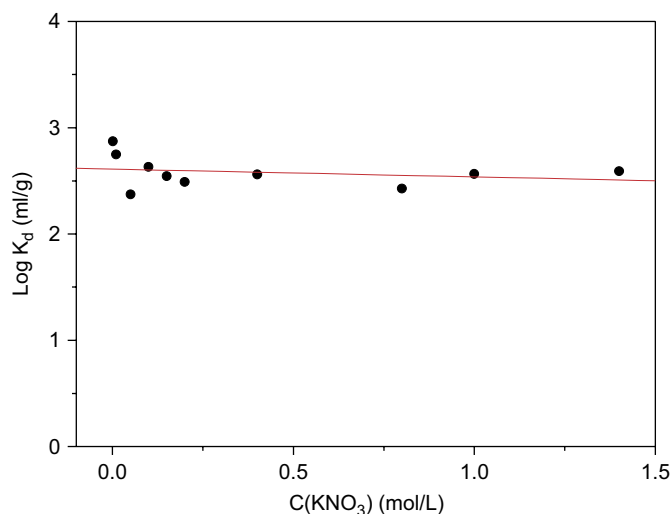


Fig. 5. Effect of ionic strength on the sorption of Co(II) on montmorillonite. pH 5.5, $m/V = 1.6 \text{ g/L}$, $3.3 \times 10^{-4} \text{ mol/L}$.

is also shown in Fig. 4. It is clear that there is no difference within the experimental uncertainties in the sorption of Co(II) on montmorillonite in the two different ionic strength conditions. The sorption of Co(II) is independent of ionic strength and pH values at $\text{pH} < 6$, and strongly dependent on pH at $\text{pH} > 6$. The pH-dependent and ionic strength-independent sorption of Co(II) on montmorillonite at $\text{pH} > 6$ indicates that the sorption of Co(II) is mainly dominated by inner-sphere surface complexation rather than by cation exchange mechanisms (Yu et al., 2006; Baeyens and Bradbury, 1997; Chen and Wang, 2006; Wang et al., 2005d). It is well known that inner-sphere surface complexation is strongly pH-dependent and weakly ionic strength-dependent; the surface sites change with the change of pH values, and thus the sorption exhibits pH dependence.

The distribution coefficient, K_d , of Co(II) sorption on montmorillonite was calculated from

$$K_d = \frac{q}{C_{\text{eq}}} = \frac{C_0 - C_{\text{eq}}}{C_{\text{eq}}} \frac{V}{m}, \quad (5)$$

where q is the concentration of cobalt in the solid phase after sorption (mol/g), C_{eq} is the concentration of cobalt in the solution phase after sorption (mol/L), C_0 is the initial concentration (mol/L), V is the volume of the solution (L), and m is the mass of montmorillonite in solution (g).

3.5. Sorption–desorption isotherms

Fig. 6 shows the sorption isotherms of cobalt on montmorillonite at $\text{pH} 7.7 \pm 0.1$ and 7.3 ± 0.1 , respectively. From Fig. 6, one can see that the sorption isotherms of cobalt on montmorillonite are linear at both pH values; this suggests that the sorption of cobalt is far from saturation. The experimental data can be fitted by the Freundlich model ($q = K_{\text{FI}}(C_{\text{eq}})^n$) very well.

The sorption of positively charged Co^{2+} onto the positively charged montmorillonite at $\text{pH} < 9$ is the direct evidence of formation of chemical bonds between the montmorillonite surface and Co^{2+} . The strong sorption of Co^{2+} on montmorillonite suggests the formation of precipitation, the chemical sorption or the strong complexation of Co(II). However, the concentration of Co(II) used here was so low that pure $\text{Co}(\text{OH})_2$ precipitation cannot be formed. Thus, a transition from the adsorption to the surface-induced precipitation took place and the montmorillonite surface acted as a template for the formation of the surface precipitation (Yu et al., 2006).

The desorption isotherms of cobalt for decreasing pH values (Method 2) are also shown in Fig. 6. The sorption experiments were performed at $\text{pH} 7.7 \pm 0.1$. After 2 days of sorption equilibration, a negligible volume of HNO_3 was added to each test tube and then the test tubes were shaken for 2 days. The pH values of the suspensions were measured to be $\text{pH} 7.3 \pm 0.1$ and then the contents were centrifugated for the separation of solid phase from liquid phase. It can be seen that the desorption isotherm is lower

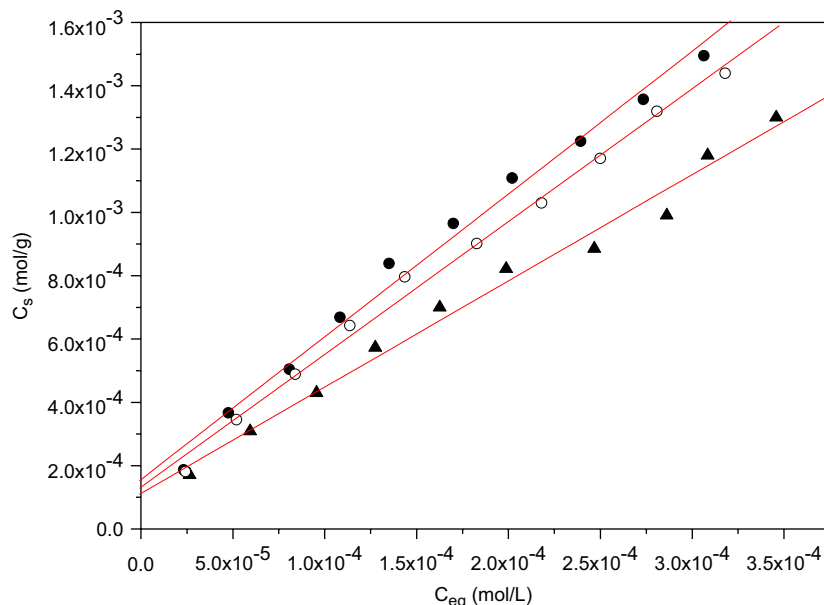


Fig. 6. Sorption and desorption isotherms of Co(II) on montmorillonite as a function of pH values. $m/V = 1.6$ g/L, $C(\text{KNO}_3) = 0.1$ mol/L. (●) Sorption, pH 7.7; (○) desorption, pH 7.3; (▲) sorption, pH 7.3.

than the sorption isotherm at pH 7.7 but higher than the sorption isotherm at pH 7.3, which indicates that the sorption is irreversible. The irreversible sorption also indicates that inner-sphere complexes are formed on montmorillonite. Only part of the surface-adsorbed Co(II) is desorbed from montmorillonite surfaces to solution by decreasing the pH values.

Fig. 7 shows the sorption and desorption isotherms of cobalt on montmorillonite at $\text{pH } 7.3 \pm 0.2$ (Method 1). As can be seen from Fig. 7, both isotherms of sorption and desorption are linear and the isotherm of desorption is higher than that of sorption. This illustrates that sorption–desorption irreversibility occurred in the sorption of cobalt on montmorillonite. The sorption of cobalt on montmorillonite is not reversible. The results derived from Methods 1 and 2 show that only part of the surface-adsorbed Co(II) can be desorbed from montmorillonite surface to solution, where most of adsorbed Co(II) is not easily desorbed from montmorillonite by changing the experimental conditions. The results of this work indicate that strong inner-sphere surface complexes are formed at the surface of montmorillonite by chemical sorption rather than by physical sorption. Generally, physical sorption is weak and the adsorbate could be desorbed from solid phase to liquid phase easily by changing the experimental conditions, while the chemical sorption is strong and the adsorbate is not easily desorbed from solid to liquid phases.

The concentration of cobalt on montmorillonite in desorption tests (Method 1) is calculated from

$$C'_s = C_s - \left(C'_{\text{eq}} - \frac{C_{\text{eq}}}{2} \right) \frac{V}{m}. \quad (6)$$

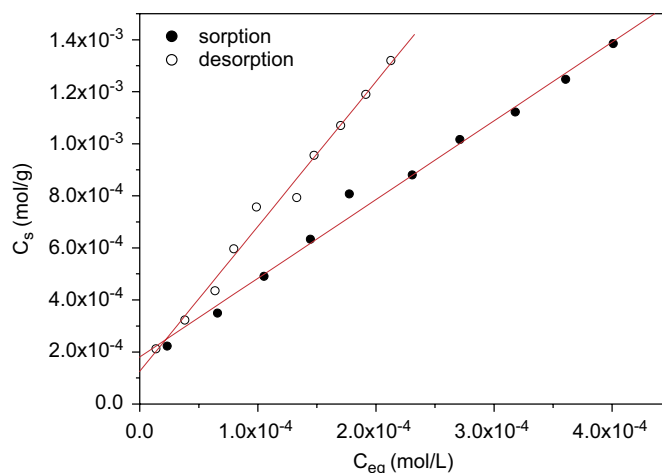


Fig. 7. Sorption and desorption isotherms of Co(II) on montmorillonite as a function of pH values. $m/V = 1.6$ g/L, $C(\text{KNO}_3) = 0.1$ mol/L. (●) Sorption, pH 7.3; (○) desorption, pH 7.3.

Here C'_s is the concentration of cobalt on the solid phase after desorption (mol/g), C_s is the concentration of cobalt on the solid phase after sorption (at the beginning of the desorption test) (mol/g), C'_{eq} is the concentration of cobalt in solution after desorption (mol/L), C_{eq} is the concentration of cobalt in solution after sorption (at the beginning of the desorption test) (mol/g), V is the volume of the solution (L) and m is the mass of montmorillonite in solution (g).

The average K_d values of Co(II) sorption and desorption at pH 7.3 are listed in Table 2. The sorption–desorption irreversibility factors of Co(II) on montmorillonite studied in the two methods are also listed in Table 2. From the K_d values of Co(II) sorption at pH 7.3 derived from the two methods, one can see that there is no difference with

Table 2
The distribution coefficients of Co(II) sorption–desorption on montmorillonite

| | pH | $\bar{K}_{d(\text{sorp})}$ (mL/g) | $\bar{K}_{d(\text{desorp})}$ (mL/g) | Irreversibility factor (%) ^a |
|----------|-----|--------------------------------------|--|--|
| Method 1 | 7.3 | 4100 ± 650 | 7350 ± 940 | 44 |
| | 7.7 | 5830 ± 880 | | |
| Method 2 | 7.3 | 4140 ± 550 | 5250 ± 700 | |
| | 7.3 | | | |

a

$$\text{Irreversibility factor} = \frac{\bar{K}_{d(\text{desorp})} - \bar{K}_{d(\text{sorp})}}{\bar{K}_{d(\text{desorp})}} \times 100\%.$$

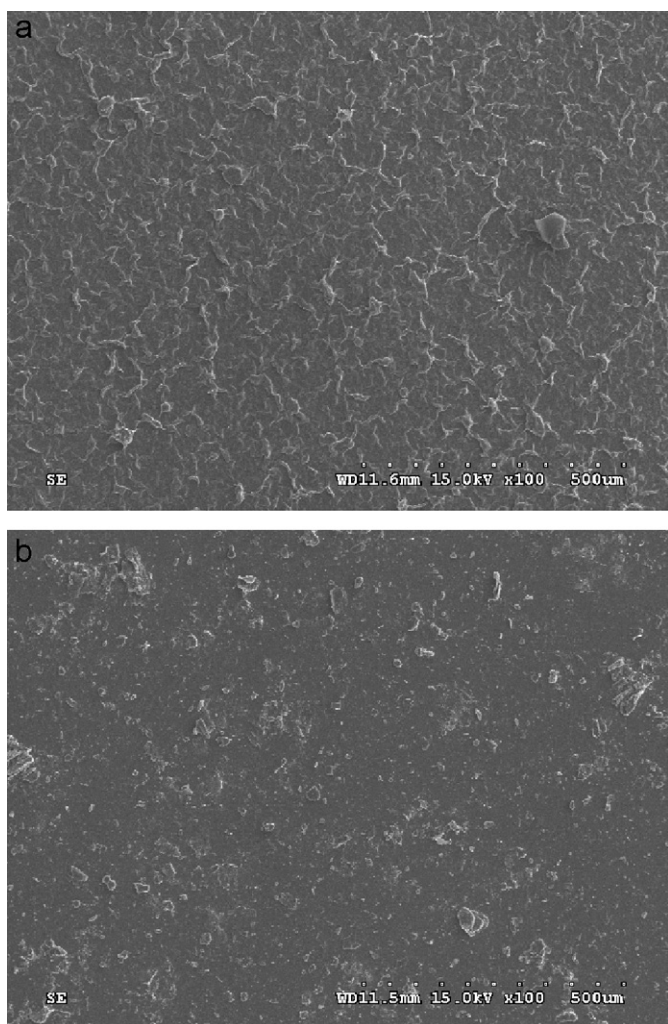


Fig. 8. SEM images of bare montmorillonite and FA–montmorillonite colloids.

experimental uncertainties. However, the K_d values of desorption are quite different: $\bar{K}_{d(\text{desorp})}$ (derived by changing pH value) is much lower than that (derived by changing the equilibration concentration of Co(II)), which suggests that sorption–desorption of Co(II) on montmorillonite is influenced more strongly by pH values than by

solution concentration. The sorption of Co(II) on montmorillonite is influenced by many factors such as pH, ionic strength, solution concentration, solid content, etc. The methods of changing one or more factors to investigate the sorption and desorption properties of Co(II) should be clarified more precisely. Different results will be achieved from different methods. The high K_d values of Co(II) sorption on montmorillonite suggest that the montmorillonite is a very suitable candidate for preconcentration and sorption of Co(II) from large volumes of solutions.

3.6. SEM images of bare and FA-bound montmorillonite

Fig. 8 shows the SEM images of bare montmorillonite (Fig. 8A) and FA-bound montmorillonite (Fig. 8B), respectively. It is clear that the particle size of FA–montmorillonite is much larger than that of montmorillonite. FA has high oxygen and carboxy groups, which can form complexes with montmorillonite. The presence of FA in montmorillonite makes the aggregations of montmorillonite; thereby, the sizes of FA–montmorillonite colloids become bigger than the size of bare montmorillonite particles. SEM analysis is a good method to observe the surface structure of colloids, which is important to evaluate the physicochemical behavior of radionuclides in the natural environment (Chen et al., 2007b).

4. Conclusions

From the experimental results mentioned above, the following conclusions can be achieved:

1. Sorption of Co(II) on montmorillonite is strongly dependent on pH values, and weakly dependent on ionic strength.
2. Sorption of Co(II) on montmorillonite is mainly dominated by surface complexation rather than by cation exchange.
3. The presence of fulvic acid enhances the sorption of Co(II) on montmorillonite under our experimental conditions. The complexes between FA and Co(II) is rather stronger than the complexes of Co(II) and surface functional groups of montmorillonite.
4. The sorption of Co(II) on montmorillonite is not reversible. Sorption of Co(II) on montmorillonite occurs on both reversible and irreversible sites.
5. Montmorillonite is a suitable candidate for preconcentration and solidification of Co(II) from large volumes of solutions.
6. Sorption–desorption of Co(II) on montmorillonite is affected more strongly by pH values than by solution concentration.

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