



Effect of pH, ionic strength, temperature and humic substances on the sorption of Ni(II) to Na-attapulgite

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ABSTRACT

Sorption of Ni(II) on Na-attapulgite as a function of pH and ionic strength is investigated under ambient conditions. The results indicate that sorption of Ni(II) is strongly dependent on ionic strength at pH < 8, and independent of ionic strength at pH > 8. Results of the desorption tests of Ni(II) from bare and HA bound attapulgite hybrids indicate that the sorption is reversible at pH 6.0. Outer-sphere complexation or ion exchange may be the main sorption mechanism of Ni(II) to attapulgite at pH < 8, whereas the uptake of Ni(II) at pH > 8 is mainly dominated by inner-sphere complexation. The presence of FA and HA decreases Ni(II) sorption to FA/HA-attapulgite hybrids at pH > 7 obviously, and the influence at pH < 7 is weakly. Sorption of Ni(II) to Na-attapulgite increases with increasing temperature. The thermodynamic data (ΔH^0 , ΔS^0 , ΔG^0) are calculated from the temperature-dependent sorption isotherms, and the results suggest that sorption process of Ni(II) on attapulgite is spontaneous and endothermic.

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1. Introduction

Attapulgite $[(\text{Mg}, \text{Al})_4(\text{Si})_8(\text{O}, \text{OH}, \text{H}_2\text{O})_{26} \cdot n\text{H}_2\text{O}]$ is a hydrated magnesium aluminum silicate present in nature as fibrillar mineral [1,2]. It presents in natural environment and its structure was illustrated in 1940, and later refined by many authors [3–6]. Attapulgite is a kind of di-octahedral clay mineral, formed by discontinuous octahedral layers elongated in the C-direction alternated with continuous tetrahedral ones. The apex of SiO_4 tetrahedral point upward and downward conforms two chains which causes the special micro-aperture and micro-channel structures in the C-direction. Under ambient conditions and room temperature, three kinds of water generally present on attapulgite particles: (i) sorption water on the mineral surface by physical effect; (ii) zeolitic water in the micro-channel by weakly bound effect; and (iii) crystalline water that tightly bound water-molecules completing with the coordination of the (Mg and Al) cations at the borders of each octahedral layer (i.e., structural water). Besides the three kinds of water, attapulgite also contains structural hydroxyl groups (Al–OH and Mg–OH) [4,5,7,8] and other sorption sites such as cations of tetrahedron, and the fringe complex water of structure with Mg^{2+} and silicic-hydroxyl groups (Si–OH) [9]. Because of its special structure, attapulgite can be modified by cationic surfactant and has

permanent negative charges on its surface, to enhance the contaminant retention, to adsorb and to retard contaminant migration on its surface or in its micro-channel. In addition, some isomorphous substitutions in the tetrahedral layer, such as Al^{3+} for Si^{4+} , develop negatively charged adsorption sites able to electro-statically adsorb cation ions [10,11]. In last decades, attapulgite has been intensively investigated as adsorbent in the removal of organic contaminants and heavy metal ions from the solutions and heavy metal polluted soils in the natural environment [12–15].

Nickel is a toxic metal ion present in wastewater and most of nickel is used in steel factories, electroplating and ceramic industry, battery and accumulator manufacturing [16,17]. Sorption of Ni(II) on clay minerals, which are ubiquitous in the environment, has been widely studied in the last decades. Many kinds of sorbents such as solid humic acid, chelating resins, raw rice bran, hematite, and montmorillonite have been used to remove Ni(II) [18–20]. Many mechanisms have been synchronously postulated for Ni(II) sorption, including surface complexation, ion exchange, surface precipitation/coprecipitation and diffusion into particle micropores [21–23]. The property of negatively surface charge of attapulgite makes it a suitable material for preconcentration and solidification of metal ions from large volume of solutions. To the best of our knowledge, the study of Ni(II) on attapulgite, especially the thermodynamic data of Ni(II) sorption on attapulgite and the effect of humic substances on Ni(II) uptake to humic-attapulgite hybrids, is still scarce.

The main purposes of this paper are: (1) to investigate the sorption behavior of Ni(II) on attapulgite; (2) to study the influence of

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ionic strength on Ni(II) sorption; (3) to evaluate the effect of pH on Ni(II) sorption; (4) to study the effect of temperature on Ni(II) sorption and to calculate the thermodynamic parameters of Ni(II) sorption on attapulgite; (5) to compare the influence of humic acid and fulvic acid on Ni(II) sorption in the ternary systems; (6) to study the sorption and desorption behavior of Ni(II) at different experimental conditions; and (7) to discuss the sorption mechanism of Ni(II) on attapulgite.

2. Experimental

2.1. Materials

All chemicals used in the experiments were purchased in analytical purity and used without any purification. The sample of attapulgite was achieved from KaiXi Co. (Gansu, China), and was treated with 5% hydrochloric acid for 24 h, then was immersed into 20% NaCl solution for 60 h; followed by washing with distilled water until no chloride was detected in supernatant with 0.01 M AgNO₃. Thus achieved sample was dried at 105 °C for 2 h in order to eliminate the free water. At last the sample was milled and passed through a 320-mesh screen and then used in the experiments [8,24,25].

Soil humic and fulvic acids were extracted from the soil of Gansu province (China), and have been characterized in detail. The main elements of HA are: C 60.44%, H 3.53%, N 4.22%, O 31.31% and S 0.50%; and those of FA are: C 50.15%, H 4.42%, N 5.38%, O 39.56% and S 0.49%.

2.2. Experimental procedure

All the experiments were carried out under ambient conditions. The stock suspension of attapulgite, NaNO₃, and nickel stock solution were added in the polyethylene tubes to achieve the desired concentrations of different components. It was necessary to note that before the addition of nickel solution, attapulgite and NaNO₃ were first shaken for 2 days to achieve the equilibration of Na⁺ with attapulgite. For the experiments in the presence of HA/FA, HA/FA was equilibrated with attapulgite for 2 days before the addition of nickel solution. The pH values of the solution were adjusted by adding negligible volumes of 0.1 or 0.01 M HNO₃ or NaOH. After the suspensions were stirred for 2 days, the solid and liquid phases were separated by centrifugation at 7500 rpm for 30 min.

The concentration of Ni(II) was analyzed by spectrophotometry at wavelength of 530 nm by using Ni butanedione dioxime complex. The concentration of FA and HA was analyzed by UV-vis spectrophotometry at wavelength of 294 nm. All the experimental data were the averages of duplicate or triplicate experiments. The relative errors of the data were about 5%.

For desorption experiments, the suspension of attapulgite was centrifuged (7500 rpm, 30 min) 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.

3. Result and discussion

3.1. XRD analysis

XRD analysis (Fig. 1) showed that the crystal structures of untreated attapulgite and attapulgite treated with NaCl did not undergo any changes. One can see that there is no difference in the peak positions ($2\theta=8.34$, 27.49, 35.01 and 42.6) of the two samples. It must be noticed that the peak at $2\theta=8.34$ has inter-

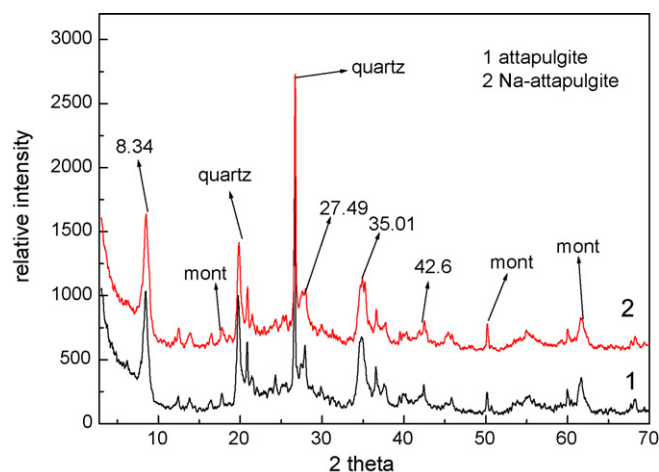


Fig. 1. XRD of attapulgite and Na-attapulgite.

planar distance (d) = 1.064 nm and is attributed to the basal plane of attapulgite structure [26]. The intergrowth-minerals of montmorillonite and quartz are also found in the sample, and have been marked in Fig. 1.

3.2. Effect of contact time

The removal of Ni(II) from solution to Na-attapulgite as a function of contact time was shown in Fig. 2. The removal of Ni(II) by Na-attapulgite was calculated by the following equation:

$$\text{Sorption\%} = \frac{C_0 - C_{eq}}{C_0} \times 100\% \quad (1)$$

where C_0 is the initial concentration of Ni(II), and C_{eq} is the equilibrium one in supernatant after centrifugation. As can be seen from Fig. 2, the sorption of Ni(II) on attapulgite was very quickly and the removal of Ni(II) to attapulgite maintained level after 2 h contact time. The quickly removal of Ni(II) from aqueous solution to attapulgite is essential for the application of attapulgite in the removal of Ni(II) from large volume of solutions. After the addition of attapulgite to solution, the sorption of Ni(II) to attapulgite occurs immediately and can be followed by the separation of attapulgite from solution quickly. In the following experiments, 2 days

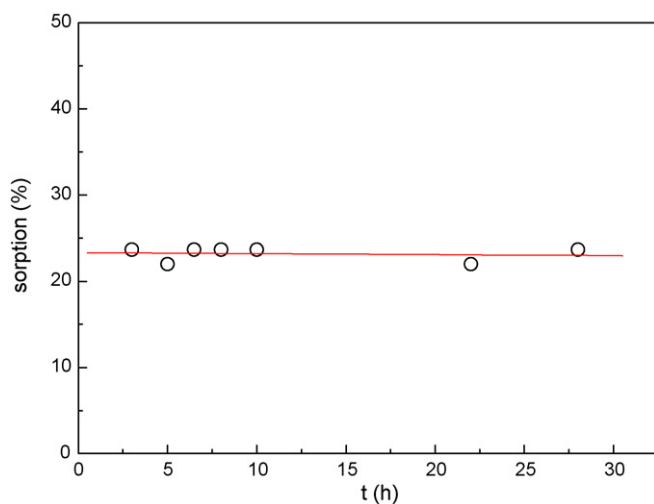


Fig. 2. Effect of time on the sorption of Ni(II) to Na-attapulgite. $C_{\text{Ni(II)initial}} = 8.52 \times 10^{-5}$ mol/L, pH 6.0 ± 0.1 , $T = 18 \pm 2$ °C, $I = 0.01$ M NaNO₃, $m/V = 0.8$ g/L.

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