



Adsorption of uranium from aqueous solution on heat and acid treated sepiolites

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ABSTRACT

In this work adsorption of uranium on natural, heat and acid treated sepiolite was studied. For acid treatment HCl and H₂SO₄ were used separately. Heat and acid treatment caused some changes in sepiolite such as surface area, micropore volume (cm³/g) and average pore diameter (Å). Different amounts of Mg ions were extracted from the lattice depending on the type of acid. After acid treatment with HCl, the amount of Mg left in the sepiolite changed a little. During H₂SO₄ treatment the sepiolite structure was progressively transformed into amorphous silica. These heat and acid treatments changed adsorption capacity and mechanism of uranium on sepiolite. Data obtained from the adsorption experiments were applied to Langmuir and Dubinin–Radushkevich (D–R) adsorption isotherms. Using these isotherms different adsorption capacities were found for natural and treated sepiolite samples. The capacity values were 3.58×10^{-3} , 3.14×10^{-3} , 2.78×10^{-3} and 1.55×10^{-3} mol/g for HCl treated, heat treated, natural and H₂SO₄ treated sepiolite samples, respectively. In order to evaluate the adsorption mechanism adsorption energies were calculated by the D–R isotherm. According to the adsorption energy values uranium fixed to the natural and heat treated sepiolite surface with ion exchange (12.75 and 12.12 kJ/mol, respectively). Simple physical attractions were the driving force for adsorption on HCl and H₂SO₄ treated ones (6.62 and 6.87 kJ/mol, respectively).

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1. Introduction

Because of their high adsorption capacity and abundance in nature, clay minerals are frequently preferred by researchers for adsorption experiments. To improve their adsorption capacity various surface modification methods like heat and acid treatment are applied by many researchers (Gonzalez-Pradas et al., 2005; Tan et al., 2004; Kara et al., 2003; Dekany et al., 1999; Torro-Palau et al., 1996; Balci, 1996).

Sepiolite is a fibrous clay mineral, which is formed of sheets of tetrahedral and octahedral oxides. It has fine channels running parallel to the fibre axis (Balci, 1996). The half unit cell of sepiolite is (Si₁₂)(Mg₈)O₃₀(OH)₄(OH₂)₄8H₂O (Martin Vivaldi and Fenoll Hach-Ali, 1969). Its structure, consisting of ribbons alternating with available open channels along the fibre axes, has provided sepiolite with good adsorption properties (Calliere et al., 1982; Gonzalez-Pradas et al., 2005). Several studies of heat and acid treatment of sepiolite have been studied to change its surface specifications (Sabah et al., 2002; Cetisli and Gedikbey, 1999;

Balci, 1996; Dekany et al., 1999; Torro-Palau et al., 1996; Gonzalez-Pradas et al., 2005). Adsorption of metal ions on sepiolite was studied by many investigators (Kara et al., 2003; Sanchez et al., 1999; Brigatti et al., 2000; Helios-Rybicka, 1985).

In this study we investigated the effect of natural, heat and acid treated sepiolite samples on the adsorption of uranium on sepiolite. The effects of the treatment on the surface properties and uranium adsorption were investigated.

2. Materials and methods

2.1. Heat and acid treatment to the sepiolite

Sepiolite sample used in this study was obtained from Eskisehir in Turkey. The particle size of the sample used was < 5 μm. To clean the clay surface from soluble impurities, 250 ml distilled water was added onto 25 g sepiolite and shaken for 3 h at 25 °C. After this the samples were dried at 40 °C in an oven for 72 h (step1). These samples were labelled as S. For heat treatment, 120 g sample S was put in a muffle furnace at 200 °C for 3 h and then labelled as S200. For acid treatment process, 120 g S200 sample was put into contact with 200 ml %10 H₂SO₄ and stirred for 3 h at 100 °C. After the acid treatment clay samples were

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washed with distilled water several times, then dried at 40 °C oven for 72 h and labelled as SS200. Same experiments were repeated using HCl instead of H₂SO₄ and samples were labelled as SC200.

2.2. Adsorption experiments

The initial uranium solution concentrations were 1 × 10⁻³, 1 × 10⁻², 3 × 10⁻², 5 × 10⁻², and 7 × 10⁻² M. The adsorption experiments were carried out using the batch method. About 40 mg of sepiolite sample was kept in contact with 5 ml of solution. All of the batch adsorption tests were performed in glass bottles in duplicate. After shaking for 1 h at constant agitation speed (200 rpm) at 25 °C the two phases were separated by centrifugation at 300 rpm for 5 min. Equilibrium concentrations of uranium were determined by measuring β-radiation of the aqueous phase with ERD Mullard G-M tube type MX 123. Five replicate measurements were made in each case, and mean activities of remaining solutions were recorded. The measurement time was set to 5 min. The adsorption percentages were calculated from the depletion of uranium from solution and comparison with blank, where no clay had been added. Therefore counting efficiency of instrument was not taken into account.

The XRD patterns were obtained on a SIEMENS Diffractometer D5000 and materials data MDI Jade 7.5 2005.

3. Results and discussion

As seen in Fig. 1 adsorption of uranium on natural sepiolite reached equilibrium in 1 h.

The relationship between the distribution coefficient (K_D) and the ratio of solution's volume to the adsorbant mass (V/m) at 1 × 10⁻² M solutions can be seen in Fig. 2.

The data obtained from adsorption experiments were applied to Langmuir and D-R isotherms (Figs. 3 and 4). The Langmuir isotherm is given by the following equation (McKay and Porter, 1997):

$$\frac{1}{q_e} = \frac{1}{q_m^L} + \frac{1}{K_L q_m^L} C_e \tag{1}$$

where q_m^L and K_L, the Langmuir constants, are the saturated monolayer sorption capacity and adsorption equilibrium constant,

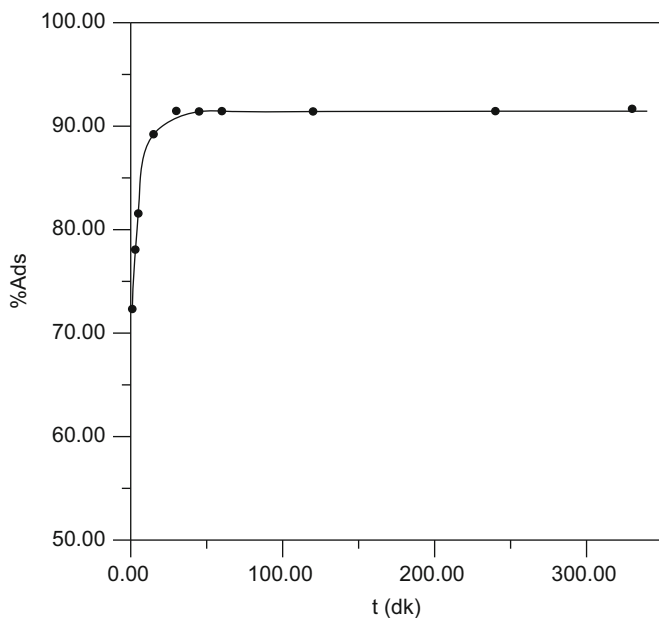


Fig. 1. Adsorption % versus time (min) for adsorption of uranium on natural sepiolite.

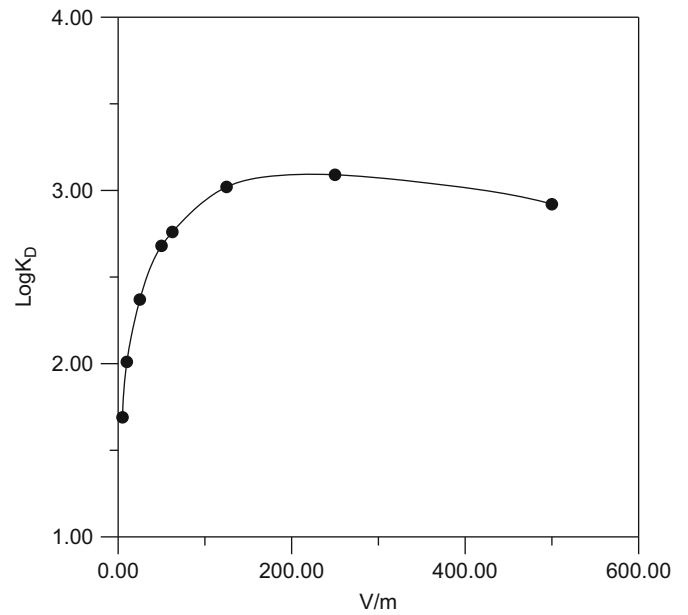


Fig. 2. Log K_D versus V/m for uranium adsorption on natural sepiolite.

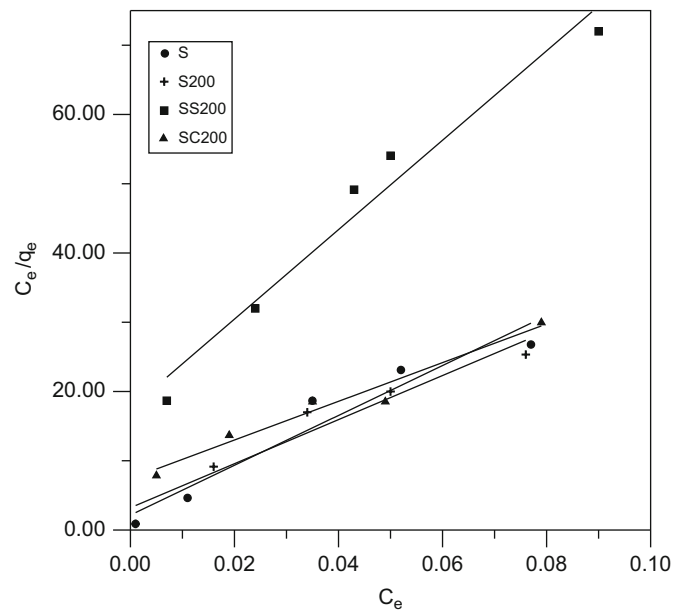


Fig. 3. Langmuir adsorption isotherm of uranium adsorption on natural, heat and acid treated sepiolite.

respectively. C_e is the equilibrium solution concentration, q_e is the amount of uranium adsorbed at equilibrium. A plot of 1/q_e versus 1/C_e would result in a straight line with a slope of (1/K_Lq_m^L) and intercept of 1/q_m^L (Fig. 3). q_m^L values showed maximum adsorption capacities for uranium adsorption on sepiolite (Table 1).

The D-R isotherm is more general than the Langmuir isotherm, because it does not assume a homogeneous surface or constant sorption potential (Aksoyoglu, 1989; Kilislioglu and Bilgin, 2003). The D-R equation is

$$q_e = q_m \exp(-K\varepsilon^2) \tag{2}$$

where K is a constant related to the adsorption energy, q_m the theoretical saturation capacity, ε the Polanyi potential, equal to RT exp(1+1/C_e). The linear form of Eq. 2 is

$$\ln q_e = \ln q_m - K\varepsilon^2 \tag{3}$$

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