

Physicochemical parameters of Hg(II) ions adsorption from aqueous solution by sepiolite/poly(vinylimidazole)



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ABSTRACT

The sepiolite/Poly(vinylimidazole) (sepiolite/PVI) composite was prepared by in situ polymerization of vinyl imidazole (VIM) and sepiolite, which was not modified. Fourier Transform Infrared (FTIR) analysis, X-ray diffraction (XRD) spectra, Scanning Electron Microscopy (SEM) images and Differential Scanning Calorimetry (DSC) exhibited that sepiolite/PVI composite was successfully produced. The theoretical maximum adsorption capacity was calculated to be 345.3 mg g^{-1} (concentration range of $100\text{--}1000 \text{ mg L}^{-1}$) in Hg(II) solution at pH 6.5 at 298 K. The adsorption kinetics and isotherms were also investigated for the adsorption process. The adsorption isotherms data were well fitted to the Langmuir model for the adsorption of Hg(II) onto sepiolite/PVI. The thermodynamic parameters (ΔG° , ΔH° , and ΔS°) were calculated which the composite could be considered as a spontaneous, endothermic and mainly physical adsorption process. The equilibrium parameter (R_L) indicated that sepiolite/PVI composite was useful for Hg(II) removal from aqueous solutions.

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1. Introduction

Hg(II) is potential carcinogenic and very toxic metal found in various industrial wastewaters, which originate from electroplating, paint and battery manufacture and mining activities [1,2]. Exposure to Hg(II) leads to various disorders and diseases in the body due to the toxic effects of these ions even at very low concentrations [3,4]. Therefore, in order to minimize the harmful effects of Hg(II), removal of mercury from waste waters is very important before it poses a treat.

The conventional methods used to remove Hg(II) from wastewaters are chemical precipitation, reverse osmosis, coagulation, ion-exchange and solvent extraction [2,5,6]. These methods are costly and have disadvantages such as generation of toxic sludge, incomplete metal removal and high energy requirements. Therefore, adsorption which is an alternative to these treatment methods is considered to be an effective and economical method for removal of Hg(II) from waste water [7–10]. Still, there is a need for efficient and environmentally friendly systems for reducing Hg(II) contamination. Therefore, developing an adsorbent with large surface area and small diffusion resistance is important. Recently

the adsorption method using a polymer/clay composite adsorbent has been applied to the removal of wastewater containing Hg(II) owing to its convenience and simplicity [6,4,11–13].

Nitrogen-rich polymers have been used for mercury adsorption due to a high affinity of nitrogen to mercury [14,15]. Among these polymers poly(vinylimidazole) (PVI) is a well-known polymer used in studies because of relatively simple to synthesize and imidazole groups of PVI have complexation properties with catalytic divalent metallic ions [16], low cost, antimicrobial activity and biodegradability [17,18] and low cost. In a previous study [11], n-vinyl imidazole copolymer beads were used for Hg(II) adsorption from aqueous solutions.

In recent years, polymer/clay composites have attracted great interest in adsorption process because they often indicate remarkably improved physical and mechanical properties when compared either to the polymer matrix (for example, polymers have relatively low surface area) [19]. Clays are chosen for polymeric composites due to large specific surface areas and excellent chemical stability [20]. So far, most of the investigation on nanocomposite including clay is mainly focused on montmorillonite with layered structure [17,18], whereas only a few works involves sepiolite [19,21–26]. Sepiolite has attained increasing interest in recent years because it exhibits extraordinary performance on the flame resistance, barrier properties, mechanical properties and thermal stability produced from its special

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Notation

C_e	Concentration of Hg(II) ions at equilibrium (mg L^{-1})
C_0	Initial concentration of Hg(II) ions in solution (mg L^{-1})
E_a	Activation energy of adsorption (kJ mol^{-1})
E_{fe}	Free energy of adsorption (kJ mol^{-1})
ΔG°	Gibbs free energy of adsorption (J mol^{-1})
ΔH°	Isosteric enthalpy of adsorption (J mol^{-1})
ΔS°	Entropy change of the adsorption process ($\text{J mol}^{-1} \text{K}^{-1}$)
q_e	The amount of Hg(II) ions adsorbed on the adsorbent at equilibrium (mg g^{-1})
q_t	The amount of Hg(II) ions adsorbed on the adsorbent at any time (mg g^{-1})
q_m	The maximum amount of Hg(II) ions adsorbed per unit mass adsorbent (mg g^{-1})
Q_L	The maximum amount of Hg(II) ions adsorbed per unit mass adsorbent (mg g^{-1})
K_L	The Langmuir constant related to the affinity of binding sites (mL mg^{-1})
n	The heterogeneity factor
K_F	The Freundlich constant
Q_{D-R}	The maximum amount of Hg(II) ions adsorbed per unit mass adsorbent (mg g^{-1})
K_{D-R}	The Dubinin-Radushkevich constant ($\text{mol}^2 \text{J}^{-2}$)
ϵ	The polanyi potential (J mol^{-1})
R_L	The dimensionless separation factor
k_1	The rate constant of pseudo first-order adsorption (min^{-1})
k_2	The rate constant of pseudo second-order adsorption ($(\text{g mg}^{-1}) \text{min}^{-1}$)
k_R	The rate constant for the modified Ritchie's-second-order model (min^{-1})
k_i	The intraparticle diffusion rate constant ($\text{mg g}^{-1} \text{min}^{-0.5}$)
R^2	Linear regression coefficient
t	Time (min)
T	Temperature (K)

structure [27,28]. But, there is no report on sepiolite/PVI composite for Hg adsorption yet.

Sepiolite is a phyllosilicate that contains a continuous two-dimensional tetrahedral sheet and differs from other layer silicates in that it lacks continuous octahedral sheets. The sepiolite has the TOT layer-fibrous structure. This structure contains in fact fragments of TOT structures that extend along the axis [29,30]. The presence of silanol groups (Si–OH) at the edges of the tunnels arise due to the discontinuity of the silica sheets. Because of the tunnels in the structure, the specific surface area of sepiolite is up to $300 \text{ m}^2 \text{ g}^{-1}$. The presence of silanol groups (Si–OH) can cause to

enhance the interfacial interaction between polymer and sepiolite. Therefore, good dispersion of sepiolite within the polymer matrix leads to improve the thermal and mechanical properties of polymers [17,18].

The physical mixing between sepiolite and polymers could hardly achieve desired dispersion due to aggregation of sepiolite in the polymer matrix. For this reason, surface organic modification of sepiolite can increase its compatibility with polymers before applying physical mixing [19,26]. Another approach is in situ polymerization usually yields polymer/clay composites in which the clay is well dispersed [17,31–33]. Therefore, the method may help to prevent aggregation of clay in the host matrix.

In this study, sepiolite/PVI composite was prepared by situ polymerization. Sepiolite without any chemical treatment was directly used for the preparation of the composite. The sepiolite/PVI composite samples were characterized by X-ray diffraction (XRD), fourier transform infrared spectroscopy (FTIR), scanning electron microscope (SEM) and differential scanning calorimetry (DSC). According to the results, our present study provided a good dispersion of sepiolite in the polymer matrix and an improvement on thermal properties of polymer. In the present study, a sepiolite/PVI composite was also used to remove heavy metal Hg(II) from aqueous solutions. The adsorption conditions of Hg(II) onto the composite have been investigated and also the kinetics and thermodynamics of the adsorption process were evaluated.

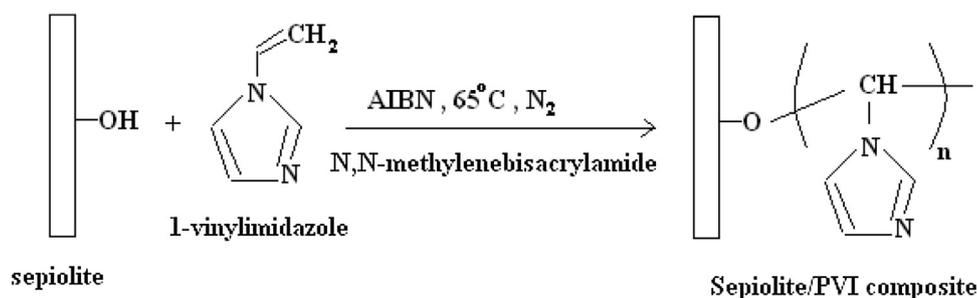
2. Experimental

2.1. Materials

The sepiolite samples were obtained from Aktaş Lületaş Co. (Eskişehir, Turkey). The chemical composition of the sepiolite determined by XRF is 53.47% SiO_2 , 23.55% MgO , 0.71% CaO , 0.43% NiO , 0.19% Al_2O_3 and 0.16% Fe_2O_3 and exhibited a 21.49 loss was exhibited upon ignition. The specific surface area of the sepiolite was $342 \text{ m}^2 \text{ g}^{-1}$ [34] 1-vinylimidazole (VIM) and azobisisobutyronitrile (AIBN) were purchased from Aldrich and Fluka, respectively. All other chemicals were of reagent grade and purchased from Merck (Darmstadt, Germany).

2.2. Preparation of sepiolite/PVI composite

The sepiolite sample was ground and sieved $75 \mu\text{m}$ size fraction. Then, it was dried at 105°C for 24 h and used for further experiments. sepiolite/PVI composite was prepared by in situ polymerization as follows. AIBN was crystallized in methanol before use. 10 mL 1-vinylimidazole, 10 mL deionized water and 0.16 g AIBN and calculated sepiolite were mixed in the flask. The prepared suspension was stirred strongly for 3 h in the ultrasonic bath for good dispersion of sepiolite and then *N,N'*-methylenebisacrylamide (0.2 g) was added in the mixture. The mixture was placed in a water bath at 65°C under nitrogen atmosphere for



Scheme 1. Proposed the formation process of sepiolite/PVI composite.

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