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Biosorption of zinc(II) from aqueous solution by dried activated sludge

Chunping Yang[1,2,3,*], Jiaqiang Wang[1,3], Min Lei[1,3], Gengxin Xie[1,3], Guangming Zeng[1,3], Shenglian Luo[1,2]

1. College of Environmental Science and Engineering, Hunan University, Changsha 410082, China. E-mail: chunpingyang@gmail.com
2. School of Environmental and Chemical Engineering, Nanchang Hangkong University, Nanchang 330063, China
3. Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, China

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Abstract

The biosorption potential of dried activated sludge as a biosorbent for zinc(II) removal from aqueous solution was investigated. The effects of initial pH, contact time, initial zinc ion concentration, and adsorbent dosage on the biosorption processes were determined, and the equilibrium data were modeled by the Langmuir and Freundlich isotherms. The Langmuir isotherm model (R^2 = 0.999) was proved to fit the equilibrium data much better than the Freundlich isotherm model (R^2 = 0.918). The monolayer adsorption capacity of dried activated sludge for zinc(II) was found to be 17.86 mg/g at pH of 5 and 25°C. The kinetic data were tested using pseudo first- and second-order models. The results suggested that the pseudo second-order model (R^2 > 0.999) was better for the description of the adsorption behavior of zinc(II) onto the dried activated sludge. Fourier transform infrared spectral analysis showed that the dominant mechanism of zinc(II) biosorption onto the dried activated sludge was the binding between amide groups and zinc ions.

Key words: activated sludge; biosorption; isotherms; kinetics; zinc(II)
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Introduction

Industrialization and urbanization often lead to an increase in the discharge of toxic metals, such as cadmium, lead, zinc, chromium and copper, into the environment, which results in a consequent damage to ecosystems and human health. Hence, the presence of heavy metals in natural water or industrial wastewater is a subject of great interest in environmental science and is one of the most serious worldwide environmental problems (Axtell et al., 2003). Conventional methods for the removal of heavy metals include chemical precipitation (Matlock et al., 2002), ion exchange (Feng et al., 2000), electrodialysis (Mohammadi et al., 2005), and membrane separations (Qdais and Hassan, 2004). All these methods have various disadvantages, specifically, high capital investment and operating cost, incomplete removal, low selectivity, high energy consumption, or the generation of residual sludge (Celaya et al., 2000).

In recent years, biosorption has emerged as an alternative to overcome these shortcomings of conventional methods for heavy metals removal (Wang and Chen, 2006, 2009). The advantages of biosorption for removal of heavy metals include low cost, free availability, possible reuse, high adsorption capacity, and flexible operation. The biosorbents investigated or used include fungi (Fan et al., 2008), algae (Kratochvil and Volesky, 1998), yeast (Goksungur et al., 2005) and bacteria (Sag and Kutsal, 2000). Biomaterials, such as waste sludge, digested sludge, and waste biomass, are also suitable for the recovery of heavy metals (Aksu et al., 2002; Bux et al., 1999; Pamukoglu and Kargi, 2006, Wang et al., 2006), and the regeneration of these biosorbents is usually unnecessary.

In this article, waste activated sludge from a municipal wastewater treatment plant was used as an adsorbent to remove zinc ions from aqueous solutions. The effects of several parameters such as contact time, initial pH of the solution, initial zinc(II) concentration, and adsorbent dosage on zinc removal were evaluated respectively. In addition, the adsorption isotherms were described by using the Langmuir and Freundlich models, and the kinetic experimental data were correlated by the pseudo first- and second-order kinetic models. The mechanism of zinc(II) biosorption onto the dried activated sludge was also studied using Fourier transform infrared (FT-IR) spectral analysis.

1 Materials and methods

1.1 Preparation of biosorbent

Activated sludge was taken from the Guozhen Municipal Wastewater Treatment Plant, Changsha, China.

* Corresponding author. E-mail: chunpingyang@gmail.com

Activated sludge biomass was centrifuged at 4000 r/min for 5 min, dried at 95°C to constant weight, and then grounded to powder. The powder was sieved to obtain particle sizes under 250 μm, and was further used as the biosorbent.

1.2 Chemicals and equipments

In this work, all chemicals were in analytical grade and were used without further purification. Double deionized water was used throughout the experiments. The stock solution of zinc(II) (1000 mg/L) was prepared by dissolving $ZnSO_4 \cdot 7H_2O$ into double deionized water, and was further diluted to the required concentrations using double deionized water. All glassware used was soaked in 10% (*V/V*) nitric acid solution for 24 hr and cleaned repeatedly with double deionized water.

A pH meter (Leici PHS-25, China) was employed for pH measuring. A PerkinElmer AAnalyst 700 atomic absorption spectrometer (AAS) (Massachusetts, USA) was used to analyze zinc ion concentrations in solutions. The FT-IR spectra of unloaded biosorbent and zinc(II) loaded biosorbent were taken using KBr discs at 400–4000 cm^{-1} using a FT-IR (WQF-410, China) spectrometer.

1.3 Batch biosorption procedure

Batch biosorption experiments were carried out in 250 mL Erlenmeyer flasks containing 100 mL of solution with various metal concentrations (20–200 mg/L). The dried activated sludge dosages ranging from 0.5 to 3 g/L were added to the solutions. The pH of the solutions was adjusted using 0.1 mol/L HCl and 0.1 mol/L NaOH prior to experiment. The flasks were stirred on a rotary shaker at 150 r/min. All the experiments were carried out at room temperature (25°C). The optimum pH value and contact time for the biosorption were determined, and adsorption kinetics was investigated using batch biosorption experiments at an initial zinc concentration of 100 mg/L, a fixed dosage of the dried activated sludge of 2 g/L, and pH of 5.0. Biosorption isotherms were examined by mixing a dosage of 2 g/L dried activated sludge with zinc(II) solution at pH 5.0 at various initial concentrations ranging from 20 to 200 mg/L. The samples taken from flasks after adsorption were centrifuged at 8000 r/min for 10 min, and the metal concentration in the supernatant was determined using AAS. All experiments were conducted in triplicates and average values were used in the data analysis. Blank experiments were run through all experiments. The adsorbed metal concentrations were obtained from the difference between initial and final metal concentrations in aqueous solution.

The amount of zinc ion adsorption onto the dried activated sludge can be calculated by Eq. (1).

$$Q = (C_i - C_f)\frac{V}{M} \quad (1)$$

where, Q (mg/g) is the zinc ion adsorption capacity; C_i (mg/L) and C_f (mg/L) are the initial and final metal ion concentrations in the solution, respectively; V (L) is the solution volume; M (g) is the amount of biosorbent used.

2 Results and discussion

2.1 Effect of pH on zinc(II) biosorption

Biosorption of zinc(II) onto the dried activated sludge as a function of pH was studied and the results are shown in Fig. 1. The experiments were conducted using 100 mg/L of initial zinc(II) solutions at initial pH values ranging from 2.0 to 5.0. The metal solutions were contacted with the biosorbent (2 g/L) for 5.0 hr at 25°C. The zinc adsorption capacity increased sharply when solution pH increased from 2 to 5 (Fig. 1). Similar results have been reported for other types of biosorbents (King et al., 2008; Yan and Viraraghavan, 2003). Biosorption at pH above 5.0 was not carried out to avoid zinc ions precipitation. Therefore, the optimum initial pH value for zinc(II) biosorption was obtained at 5.0 and used for all other biosorption experiments in this study.

Earlier studies on heavy metal biosorption have shown that pH is one of the most important parameters affecting the biosorption process (Fan et al., 2008; Gong et al., 2005). The change in pH values can usually result in the change of the surface charge on the biosorbent. At high pH values, electrostatic repulsion decreases owing to the reduction of positive charge density on the biosorbent, which leads to an increase in metal adsorption capacity (King et al., 2008). Therefore, zinc adsorption capacity increased with increase in pH value.

2.2 Effect of contact time on zinc(II) biosorption and its kinetics

The time course studies on the zinc(II) biosorption for solutions of 100 mg/L with biosorbent dosage of 2 g/L were performed by conducting batch biosorption experiments. The relationship between zinc adsorption capacity and adsorption time in Fig. 2 indicates that zinc adsorption capacity increased obviously during the first 60 min of the metal-biosorbent contact. This phenomenon could be attributed to the instantaneous utilization of the most readily available adsorbing sites on the adsorbent surface. The equilibrium was reached within 180 min. Therefore, the zinc(II) adsorption capacity and the concen-

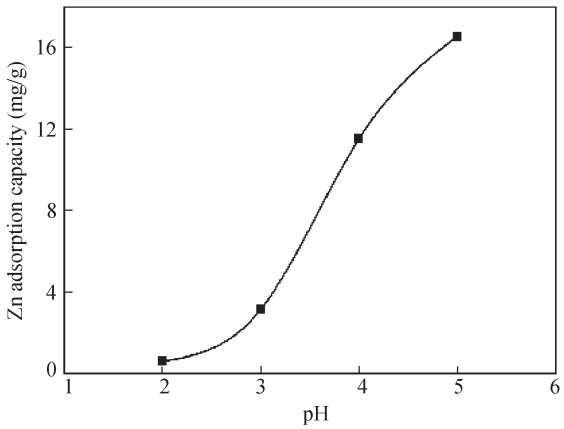

Fig. 1 Effect of pH on zinc adsorption by dried activated sludge. Conditions: initial zinc(II) concentration 100 mg/L; adsorbent dosage 2 g/L; contact time 5 hr; temperature 25°C.

tration of the unadsorbed zinc(II) at the end of 180 min were given as the equilibrium values of Q_e (mg/g) and C_e (mg/L), respectively. All other biosorption experiments were conducted at contact time of 180 min.

In order to better understand the kinetics of zinc(II) biosorption on the dried activated sludge, the Lagergren's pseudo first- and second-order models were used to describe the experimental data.

The linear form of the pseudo first-order equation by Lagergren (Kumar et al., 2006) is given as Eq. (2):

$$\log(Q_e - Q_t) = -\frac{k_1}{2.303}t + \log Q_e \quad (2)$$

where, Q_e and Q_t (mg/g) are the adsorption capacities at equilibrium and at time t, respectively, and k_1 (min^{-1}) is the rate constant of pseudo first-order sorption. The values of k_1, and the calculated $Q_{e,cal}$ were obtained from slope and intercept of straight line in Fig. 3a. These values along with the corresponding correlation coefficient (R^2) were found as: $k_1 = 0.062\ min^{-1}$, $Q_{e,cal} = 9.33$ mg/g, and $R^2 = 0.951$.

The value $Q_{e,cal}$ determined from the model is not in a good agreement with the experimental value of $Q_{e,exp}$ (16.5 mg/g). Therefore the pseudo first-order model is not suitable to model the adsorption of zinc(II) onto the dried activated sludge.

The pseudo second-order model is more suitable for the description of the kinetic behavior of adsorption in which chemical sorption is the rate-controlling step. This equation is in the following form (Ho and McKay, 1999):

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{1}{Q_e}t \quad (3)$$

where, Q_e and Q_t (mg/g) are the adsorption capacity at equilibrium and at time t, respectively, and k_2 (g/(mg·min)) is the rate constant of pseudo second-order sorption. The plot of t/Q_t versus t (time) (Fig. 3b) could provide a linear relationship, from which the coefficients Q_e and k_2 can be calculated. These values and the corresponding correlation coefficient (R^2) were as follows: $k_2 = 0.0156$ g/(mg·min), $Q_{e,cal} = 16.95$ mg/g, and $R^2 > 0.999$.

The correlation coefficient (R^2) for the pseudo second-order equation is high and the calculated $Q_{e,cal}$ value agrees with the experimental $Q_{e,exp}$ (16.5 mg/L). This strongly suggests that the biosorption of zinc(II) onto the dried activated sludge can be represented more appropriately by the pseudo second-order model.

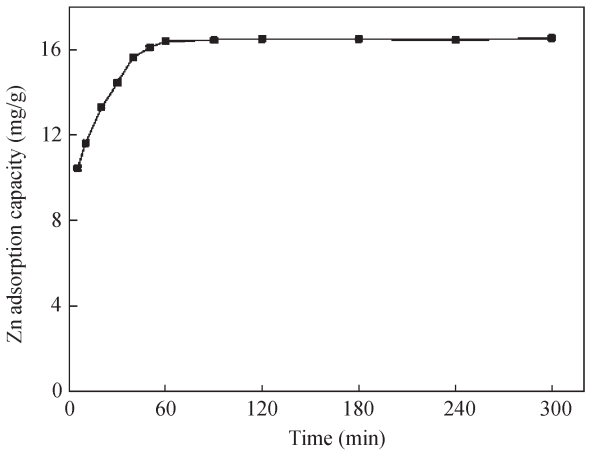

Fig. 2 Effect of contact time on zinc adsorption by dried activated sludge. Conditions: initial zinc(II) concentration 100 mg/L; biosorbent dosage 2 g/L; pH 5; temperature 25°C.

2.3 Effect of adsorbent dosage on zinc(II) biosorption

The effect of dosage of the dried activated sludge on zinc(II) removal was examined at the dosage range from 0.5 to 3 g/L for a contact duration of 180 min. The initial zinc ion concentration was 100 mg/L. The result showed that an increase in adsorbent dosage resulted in an increase in zinc removal efficiency (Fig. 4). This is owing to the increase of adsorbent mass (more surface area available for adsorption) that would result in a greater availability of reactive groups (increase in the number of binding sites) (Kumar et al., 2006). Similar trend was also observed for zinc removal using *Pinus sylvestris* ovulate cones as the adsorbent (King et al., 2008).

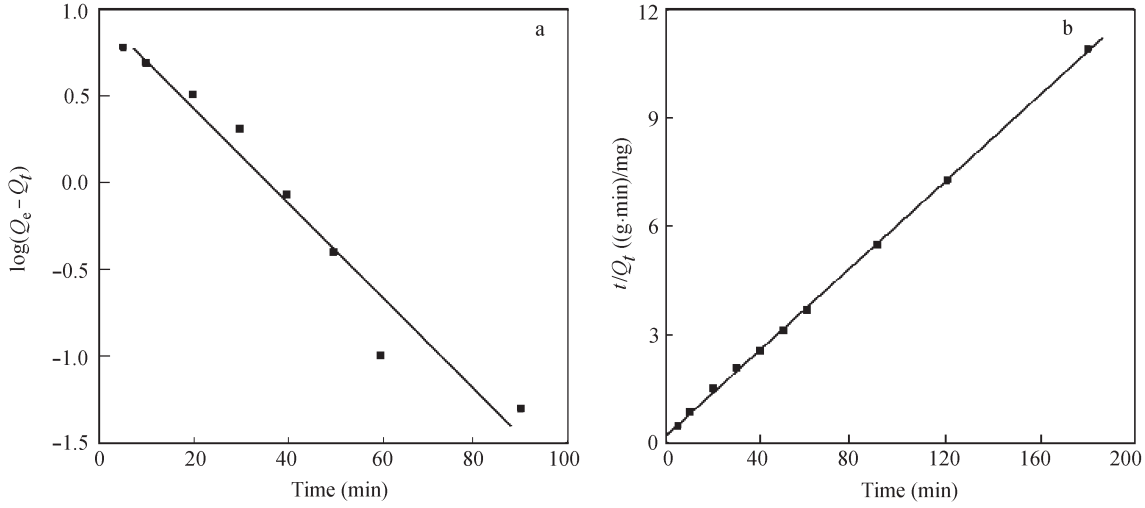

Fig. 3 Biosorption kinetics of zinc ions onto dried activated sludge. (a) the pseudo first-order model; (b) the pseudo second-order model. Conditions: initial zinc(II) concentration 100 mg/L; biosorbent dosage 2 g/L; pH 5; temperature 25°C.

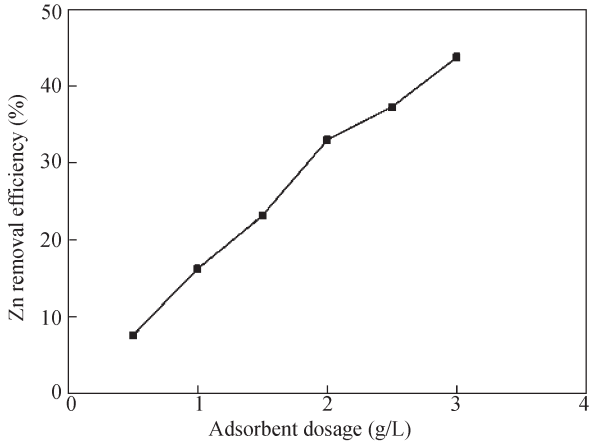

Fig. 4 Effect of biosorbent dosage on zinc removal from solution. Conditions: initial zinc(II) concentration 100 mg/L; pH 5; contact time 3 hr; temperature 25°C.

2.4 Effect of initial concentration on zinc(II) biosorption

The effect of initial zinc ion concentration on the biosorption of zinc(II) by the dried activated sludge was investigated when the initial zinc ion concentration ranged from 20 to 200 mg/L at an adsorbent dosage of 2 g/L. The experimental results showed that the metal adsorption capacity increased while the removal efficiency of zinc(II) decreased with an increase in initial zinc ion concentration (Fig. 5). The increase in zinc(II) adsorption capacity was resulted from the increase in the driving force, i.e., concentration gradient of adsorption (King et al., 2008). The decrease in the removal efficiency of zinc ions might be owned to insufficient binding sites for adsorption. Kumar et al. (2006) also thought that a lower removal efficiency at higher concentrations was due to the saturation of binding sites.

2.5 Biosorption isotherms

Two different biosorption isotherms, the Langmuir and the Freundlich isotherms, were used to correlate the equilibrium data.

The Langmuir isotherm has been successfully applied to many pollutant adsorption processes and has been the most widely used adsorption isotherm for the adsorption of a solute from a liquid solution. A basic assumption of the Langmuir theory is that adsorption takes place on a homogeneous surface by monolayer sorption.

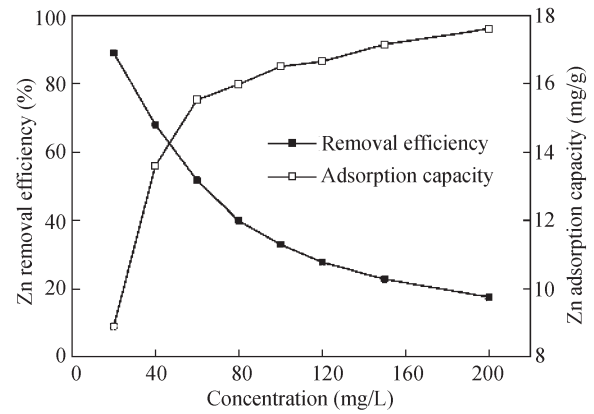

Fig. 5 Effect of zinc ion concentration on the biosorption of zinc by dried activated sludge. Conditions: biosorbent dosage 2 g/L; pH 5; contact time 3 hr; temperature 25°C.

The Langmuir isotherm has the following form (Eq. (4)) (Fan et al., 2008):

$$Q_e = \frac{Q_{max} C_e}{K + C_e} \tag{4}$$

which may be written in linearized form as follows:

$$\frac{C_e}{Q_e} = \frac{C_e}{Q_{max}} + \frac{1}{bQ_{max}} \tag{5}$$

where, Q_e (mg/g) is the equilibrium metal adsorption capacity of the adsorbent; C_e (mg/L) is the equilibrium metal ion concentration in the solution; Q_{max} (mg/g) is the maximum adsorption capacity of the adsorbent; and b (L/mg) is a constant related to adsorption energy. A plot of C_e/Q_e versus C_e in Fig. 6a indicated a straightline of slope $1/Q_{max}$ and an intercept of $1/(Q_{max}b)$. The Langmuir model constants and correlation coefficient were: Q_{max} = 17.86 mg/g, b = 0.227 L/mg, and R^2 = 0.999.

The Freundlich isotherm proposes a monolayer sorption with a heterogeneous energetic distribution of active sites, accompanied by interactions between adsorbed molecules. The Freundlich isotherm has the following form (Eq. (6)) (Fan et al., 2008):

$$Q_e = K_f C_e^{1/n} \tag{6}$$

which may be written in linearized form as follows:

$$\ln Q_e = \ln K_f + \frac{1}{n} \ln C_e \tag{7}$$

where, Q_e (mg/g) is the equilibrium metal adsorption capacity of the adsorbent; K_f (mg/g) and n (g/L) are Freundlich constants related to adsorption capacity and intensity of adsorption, respectively; and C_e (mg/L) is the equilibrium metal ion concentration in the solution. From the slope and intercept of the line in Fig. 6b, the Freundlich model constants and correlation coefficient could be obtained as: K_f = 8.55 mg/g, n = 6.51 g/L, and R^2 = 0.918.

On the basis of the correlation coefficients (R^2), the Langmuir isotherm was found to represent the equilibrium adsorption data better than the Freundlich isotherm. The maximum adsorption capacity (Q_{max}) of the dried activated sludge was found to be 17.86 mg/g for zinc(II). The Q_{max} of the dried activated sludge in this study was compared with those in some other reported studies (Table 1). Differences in the adsorption capacity data are most likely due to different properties of each adsorbent, such as structure, functional groups, and surface area.

2.6 FT-IR analysis

In order to find out which functional groups play major roles in the adsorption of zinc ions, FT-IR spectral analysis of unloaded dried activated sludge and zinc(II) loaded dried activated sludge were carried out. Figure 7 shows

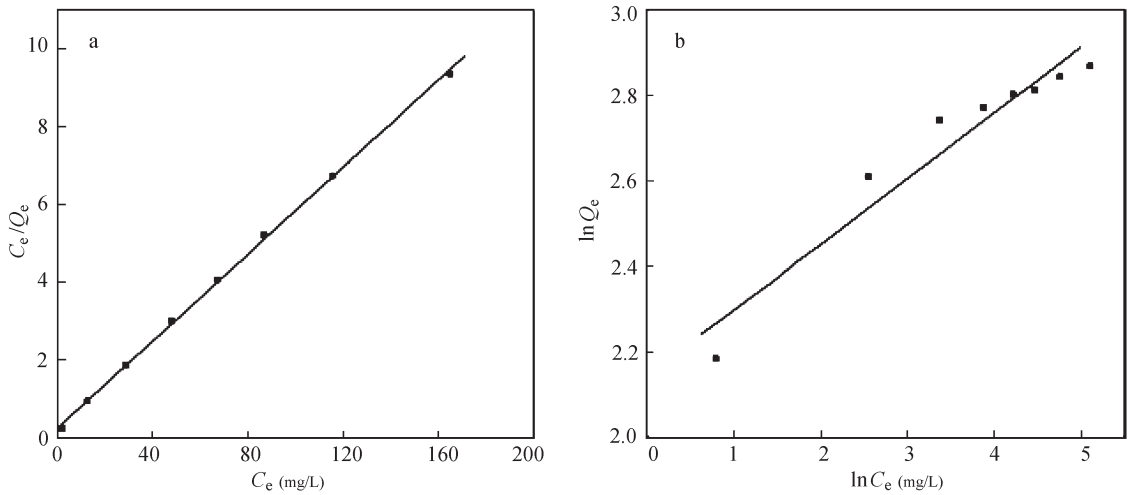

Fig. 6 Biosorption isotherms of zinc ions onto dried activated sludge. (a) Langmuir isotherm; (b) Freundlich isotherm. Conditions: biosorbent dosage 2 g/L; pH 5; contact time 3 hr; temperature 25°C.

Table 1 Maximum adsorption capacities for zinc(II) adsorption using different adsorbents

Adsorbent	Q_{max} (mg/g)	pH	Reference
Activated carbon	31.11	4.5	Mohan and Singh, 2002
Coir	8.6	5.5	Kathrine and Hansen, 2007
Barley straw	5.3	5.5	Kathrine and Hansen, 2007
Coniferous bark	7.4	5.5	Kathrine and Hansen, 2007
S. cumini L.	35.84	6	King et al., 2008
Fontinalis antipyretica	14.7	5.0	Martins et al., 2004
Botrytis cinerea biomass	12.98	5.0–6.0	Tunali and Akar, 2006
Activated sludge	17.86	5.0	Present study

the FT-IR spectra and the various functional groups corresponding to the absorption bands. The band at 3296 cm^{-1} is attributed to the stretching of –OH and –NH in activated sludge polymeric compounds. The adsorption bands at 2924 and 2854 cm^{-1} are caused by asymmetric vibration of –CH and symmetric vibration of –CH, respectively. The adsorption band at 1637 cm^{-1} could be attributed to C═O stretching conjugated to a –NH deformation of –CN (amide I) group of protein peptide bond and chitosan. The 1543 cm^{-1} band is caused from the stretching vibration of –CN and the deformation vibration of –NH (amide II) of peptide bond of proteins. A band 1410 cm^{-1} is formed from the deformation vibration of phenolic –OH and C═O stretching of carboxylate. A 1036 cm^{-1} band could be drawn out by the vibration of –C–O–C and –OH of polysaccharides. FT-TR spectra analysis after zinc biosorption showed that there was a substantial decrease in the adsorption intensity of amide group at 1637 and 1543 cm^{-1}, which indicated that the amide group played the main role in binding zinc. Similar results were also reported in literature for cadmium and lead removal using dried activated sludge (Wang et al., 2006). The group of –OH is also involved in zinc binding to some extent.

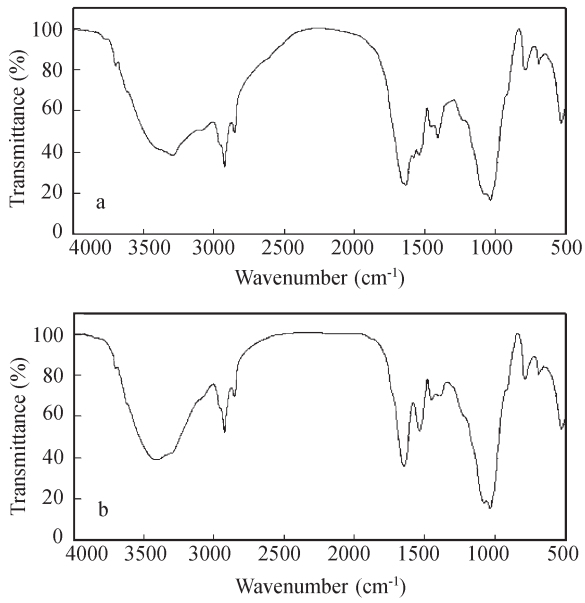

Fig. 7 FT-IR spectra of dried activated sludge. (a) before zinc(II) adsorption; (b) after zinc(II) adsorption.

3 Conclusions

This study focuses on the biosorption of zinc(II) from aqueous solution using the dried activated sludge as a low-cost biosorbent. The biosorption characteristic has been examined at various pH values, contact time, biomass dosages, and initial concentrations. The zinc adsorption capacity increased with increase in pH values ranging from 2 to 5. The extent of zinc removal was directly related with the dosage of the dried activated sludge in the solution. The zinc removal efficiency increased from 7.5% to 43.7% with increase in adsorbent dosage from 0.5 to 3 g/L at an initial zinc ion concentration of 100 mg/L. The uptake of zinc ions by the dried activated sludge increased from 8.9 to 17.6 mg/g, and the zinc removal efficiency decreased from 89.0% to 17.6% when the initial zinc ion concentration increased from 20 to 200 mg/L.

The Langmuir model can describe the equilibrium data for biosorption of zinc ions onto the dried activated sludge better than the Freundlich model, and the maximum adsorption capacity was 17.86 mg/g. The kinetic data for

the biosorption followed well the pseudo second-order kinetic model. The FT-IR spectral analysis indicated that amide group was mainly responsible for zinc(II) removal and that hydroxide group was also involved in zinc(II) binding to some degree. Based on these results, it can be concluded that as a low-cost biosorbent, the dried activated sludge, can be used for zinc(II) ions removal from aqueous solutions.

Acknowledgments

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