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Zinc (II) Removal from Aqueous Solution by Biosorption with Aerobic Granular Sludge

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Abstract: In this study, aerobic granular sludge was utilized as an effective biosorbent to remove Zn^{2+} from the aqueous solution. The results showed that the initial pH, contact time and Zn^{2+} concentration affected the biosorption process significantly. Typically, the adsorption capacity of Zn^{2+} by aerobic granular sludge increased with increasing pH in the ranges of 5.0-7.0 and decreased with increasing of pH in the ranges of 7.0-9.0, which indicated that pH at 7.0 was optimal choice for the Zn^{2+} adsorption by the aerobic granular sludge. The maximum removal of Zn^{2+} at pH 7.0 was found to be 131.47 mg g^{-1} at initial Zn^{2+} ion concentration of 120 mg L^{-1} by the aerobic granular sludge. In addition, the adsorption ability of two kind of anaerobic granular sludge from different reaction stage at one reactor for Zn^{2+} remove was compared. It was founded that the anaerobic granular sludge from the end of aerobic stage had better adsorption ability than that from the end of anaerobic stage, which could be attributed to be the species and quantity of microbial in the aerobic granular sludge.

Key words: Aerobic granular sludge, biosorption, zinc

INTRODUCYION

Heavy metal wastewater has become a global environmental concern due to serious health threaten to humans (Mohammed-Azizi *et al*., 2013; Kim *et al*., 2012; Ibrahim *et al*., 2013; Hizal *et al*., 2013). The traditional approaches, such as precipitation, oxidation/reduction, ion exchange, filtration, electrochemical processes and membrane separations exhibit several disadvantages, such as high cost, incomplete removal, low selectivity, high energy consumption and generation of toxic slurries in metals removing (Valdman and Leite, 2000). Due to its potential applications in environmental protection and recovery of toxic or strategic heavy metals, microorganisms has been paid much attention to the removal of metal ions (Volesky, 2007; Wang and Chen, 2006; Bishnoi *et al*., 2007). However, the majority of biosorbents explored in previous studies were small particles with low density, poor mechanical strength and little rigidity, which would result in difficult separation of the treated effluent from the biosorbents. To overcome these weaknesses, cell immobilization technology is developed and various immobilized biomaterials have been applied in the metal biosorption successfully (Akhtar *et al*., 2008; Van Hullebusch *et al*., 2004; Akar *et al*., 2009; Ursoiu *et al*., 2012).

Aerobic granules, a novel type of microbial aggregates, have been extensively used to remove nitrogen, phosphorus and refractory compounds from wastewater on a laboratory scale (Liu and Tay, 2004; Dangcong *et al*., 1999; Cassidy and Belia, 2005). Due to the strong microbial structure, excellent settleability, high biomass retention and resistance to toxic compounds, aerobic granules is accounted as an excellent biomaterial for the removal of heavy metals (Adav *et al*., 2008; Liu *et al*., 2003). In this study, the feasibility of aerobic granules for the removal of Zn^{2+} from aqueous solution was investigated and the effect of initial pH, contact time and Zn^{2+} concentration were studied to optimize the biosorption operation. In addition, in order to realize the activity of different kinds of aerobic granules to Zn^{2+} biosorption, the Zn^{2+} remove by the two kind of anaerobic granular sludge from different reaction stage at one reactor were carried out.

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MATERIALS AND METHODS

Aerobic granules and growth condition: Aerobic granular sludge was taken from the SBR reactor and took sodium acetate as the only carbon source. The SBR operation cycle was anaerobic for 2 h first and aerobic for 4 h afterwards. For biosorption test, the aerobic granular sludge samples were harvested at the end of anaerobic and aerobic and were then gently washed three times using de-ionized water to remove the surface soluble ions. Mature granules were nearly spherical in shape and had a compacted, integrated structure (Fig. 1). The mean size of the granular sludge was around 0.5~1 mm.

Chemicals: The stock solution of Zn^{2+} was prepared by dissolving Zn $(NO_3)_2 \bullet 6H_2O$ (Analytical grade) in the de-ionized water and further diluted to the concentrations required for the experiments.

Batch biosorption experiments: To study the effect of pH, the Zn^{2+} solution was adjusted to the desired pH (5.0-9.0) with HCl or NaOH. In the biosorption isotherm study, a series of Zn^{2+} solution with various concentrations (40-120 mg L^{-1}) were mixed with 5mL aerobic granular sludge suspension at pH 7.0 for 5 h. In the biosorption kinetics study, contact time was changed from 0 to 300 min and samples were collected at different intervals.

In the experiment of Zn adsorption by different granular sludge, the aerobic granular sludge was obtained from the end of the aerobic reaction or anaerobic reaction stage in a one reaction cycle. The biosorption experiments were performed with 250 mL triangular flasks containing 50 mL Zn^{2+} solution as follows: Put 5 mL aerobic granular sludge suspension in 250 mL triangular flask and inject 50 mL Zn^{2+} solution on Guohua SHA-C reciprocating oscillator (300 times min^{-1}, 25°C) to the required time. After the adsorption, the sample was centrifuged by Pingfan DD5M centrifuge at 5000 r min^{-1} for 15 min and then was filtered by the filter paper to obtain the filtrate. Finally, The Zn^{2+} concentration of filtrate was determined by the Varian flame atomic absorption spectrometry AA240FS.

In the study, the biosorption capacity of Zn^{2+} was defined as follows:

$$Q = \frac{Co - C}{Co} \times 100\%$$

where, Co and C are the initial and final of Zn^{2+} concentrations (mg L^{-1}).

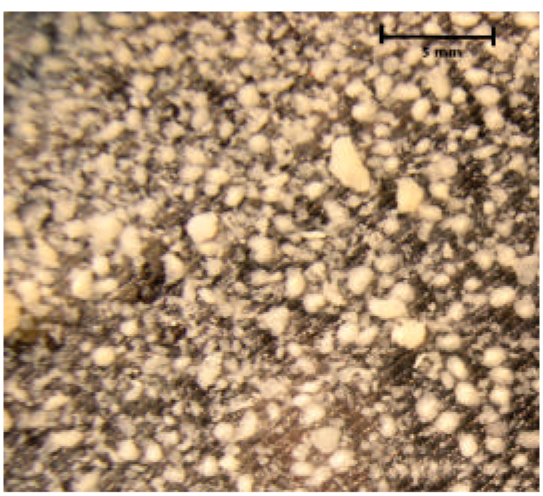

Fig. 1: Appearance of aerobic granules from the end of aerobic stage

RESULTS AND DISCUSSION

Effect of pH: Figure 2 shows the effect of pH on the Zn^{2+} biosorption at the 25°C with different concentration of Zn^{2+} and 5 mL aerobic granular sludge suspension from the end of aerobic stage. It can be seen that pH play an important role in the biosorption of Zn^{2+}. At pH was 5~7, as the pH increase from 5.0 to 7.0, the biosorption capacity obviously increased and reached the maximum value at pH 7.0. When the pH further increased (>7.0), the biosorption capacity began to decrease. This result indicated that pH 7.0 was the optimum choice for the Zn^{2+}

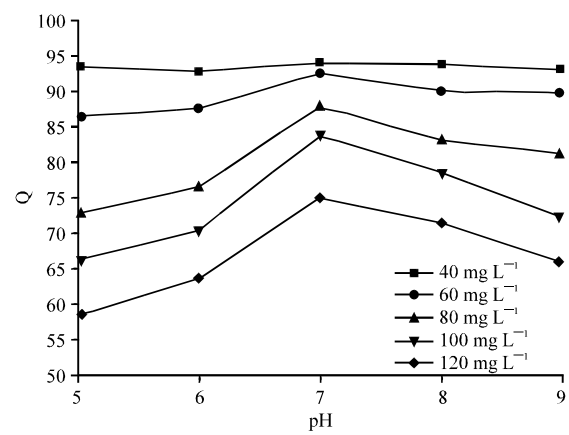

Fig. 2: Effect of pH on the Zn^{2+} biosorption at the 25°C with different concentration of Zn^{2+} and 5 mL aerobic granular sludge suspension from the end of aerobic stage

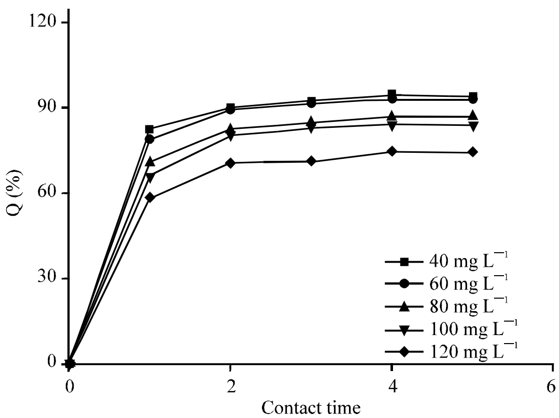

Fig. 3: Effect of contact time and initial Zn^{2+} concentration on the Zn^{2+} biosorption at pH 7.0 and 25°C with 5 mL aerobic granular sludge suspension from the end of aerobic stage

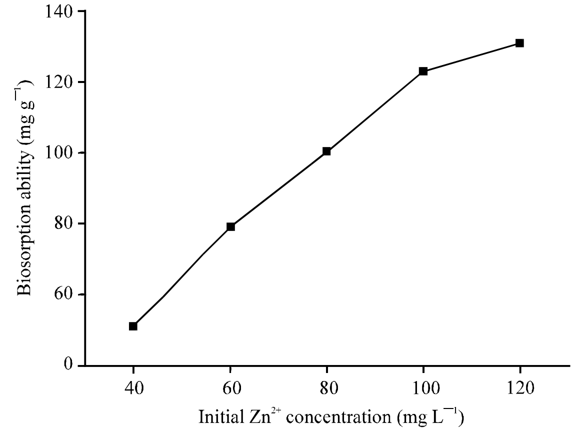

Fig. 4: Biosoption ability of Zn^{2+} by the aerobic granular sludge from the end of aerobic stage

remove by the aerobic granular sludge. It has been reported that the effect of pH on the uptake could be explained by the different activity of the functional groups and metal chemistry in solution (Yao *et al*., 2009). At acidic condition, protons (H^+) would compete for active sites on the cell of aerobic granules and thus restrict the interaction between metal ions and the biomass (Murphy *et al*., 2007). As the pH increased (5.0-7.0), more negative charge functional groups such as carboxyl, amine, hydroxyl and phosphate groups were exposed as active sites, which could react with metal ions (Klimmek *et al*., 2001). It has been reported that the biosorption of heavy metal by the aerobic granules increased with the pH increasing because of precipitation of heavy metal under the alkaline condition (Liu *et al*., 2003; Yao *et al*., 2009). However, in this experiment, the biosorption of Zn^{2+} by the aerobic granular sludge decreased as the pH increase from 7.0 to 9.0. This result was different with previous reports. The related mechanism need for further study.

Effect of contact time and initial Zn^{2+} concentration: Figure 3 illustrated the effect of contact time and initial Zn^{2+} concentration on the Zn^{2+} biosorption at pH 7.0 and 25°C with 5 mL aerobic granular sludge suspension from the end of aerobic stage. Two stage of biosorption behavior was observed. At first stage, biosorption capacity sharply increased within the first 2 h due to the rapid surface sorption, which was in agreement with previous studies (Tunali and Akar, 2006; Dixit *et al*., 2010; Zewail and El-Garf, 2010). During the second stage (2~5 h), a slower uptake of Zn^{2+} was observed as a result of free binding sites becoming saturated gradullly. Biosorption equilibrium can be achieved at 5 h.

Figure 3 also showed that the biosorption capacity decreased with the increase of initial Zn^{2+} concentration. When Zn^{2+} concentrations were 40, 60, 80, 100 and 120 mg L^{-1}, the biosorption capacity of Zn^{2+} by aerobic granular sludge biosorption capacity were correspondingly 94, 93.0, 87, 83.8 and 74.6% at pH 7.0 and 5 h, respectively. This indicated that biosorption capacity of Zn^{2+} by the aerobic granular sludge was decreased. However, biosorption ability increased along with the increase of Zn^{2+} concentration actually as shown in Fig. 4. The aerobic granular sludge adsorption ability values were 55.58, 82.06, 102.23, 123.82 and 131.47 mg g^{-1} dry sludge, respectively.

Biosorption kinetics: According to the reports about the heavy metal biosorption by the aerobic granular sludge (Yao *et al*., 2009; Wang *et al*., 2010), the models of pseudo-second-order was suitable to describe the kinetic data of Zn^{2+} biosorption by the aerobic granular sludge. The pseudo-second-order models is generally expressed in forms as follows:

$$\frac{t}{Qt} = \frac{1}{kQmax^2} + \frac{1}{Qmax}t$$

where, Qt is the Zn^{2+} sorption capacity at time t, k (min^{-1}) is rate constants of pseudo-second-order.

Table 1 lists the results of kinetic parameters of the pseudo-second-order models at different Co. It can

Table 1: Lists the results of kinetic parameters of the pseudo-second-order models at different Co

	Pseudo-second-order	
Co	k	R^2
40	0.0583	0.9998
60	0.0501	0.9997
80	0.0409	0.9996
100	0.0397	0.9985
120	0.0388	0.9989

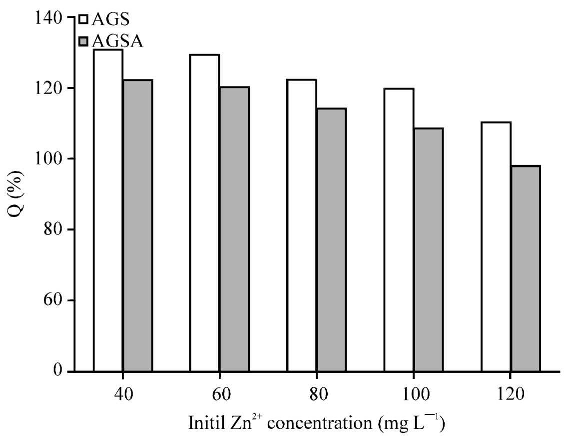

Fig. 5: Effect of different phases of aerobic granular sludges to Zn^{2+} biosorption (AGS: Aerobic granular sludges from the end of aerobic stage, AGSA: Aerobic granular sludges from the end of anaerobic stage)

be found that pseudo-second-order model described the biosorption process more effectively. This result implied that the biosorption of Zn^{2+} with aerobic granular sludge could be best described by the pseudo-second-order model. It also can be noticed that the kinetics rate of pseudo-second-order model was relatively higher (k>0.0583) at lower Co (= 40 mg L^{-1}), which provided significant practical importance as it would facilitate the process in the treatment of dilute Zn^{2+} contaminated wastewater.

Effect of aerobic granular sludges in different phases: In order to test the effect of different phases of aerobic granular sludges to Zn^{2+} biosorption, two kinds of aerobic granular sludges from the end of aerobic stage (AGS) and the end of anaerobic stage (AGSA). Figure 5 shows that the biosorption of Zn^{2+} by the two kinds of aerobic granular sludges from the end of aerobic stage and the end of anaerobic stage at pH 7.0 and 25°C. It was founded that the biosorption capacity of Zn^{2+} by the anaerobic granular sludge from the end of aerobic stage was higher than that from the end of anaerobic stage in the all different initial Zn^{2+} concentration. According to the Xu and liu's (2008) report, the mechanism of heavy metals biosorption by the aerobic granular sludges could be attributed to the surface adsorption and intracellular adsorption. Surface adsorption refers to the active sludge microbe's extracellular polymerizer (chitin, chitosan and so on) with quadridentate group like -OH, -COOH, -HN_2, PO_4^{3-} and –HS, which carried out precipitation, complexation, ion exchange and adsorption with metal ions. Intracellular adsorption was realized by metal ions' coalescent with intracellular permease and hydrolase. It was well known that more species and quantity of microbial in the aerobic granular sludge in the aerobic condition, which indicate that the activity of microbial metabolic in the aerobic condition was stronger than that of in the anaerobic condition, further to result in the higher activity of Zn^{2+} biosorption by the aerobic granular sludges from the end of aerobic stage.

CONCLUSION

This study investigated the feasibility of aerobic granules as a novel effective biosorbent for Zn^{2+} removal. The results about the effect of the initial pH, contact time and Zn^{2+} concentration shows that the adsorption capacity of Zn^{2+} by aerobic granular sludge increased with increasing pH in the ranges of 5.0-7.0 and decreased with increasing pH in the ranges of 7.0-9.0. And the maximum removal of Zn^{2+} at pH 7.0 was found to be 131.47 mg g^{-1} at initial Zn^{2+} ion concentration of 120 mg L^{-1} by the aerobic granular sludge. In addition, the different of two kind of anaerobic granular sludge from different reaction stage at one reactor to the Zn^{2+} biosorption was carried out. It was founded that the anaerobic granular sludge from the end of aerobic stage had better adsorption ability than that from the end of anaerobic stage, which could be attributed to be the species and quantity of microbial in the aerobic granular sludge.

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