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Effect of competitive interference on biosorption of cadmium by immobilized *Lentinus edodes* residue

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ABSTRACT

Use polyvinyl alcohol-Na-alginate (PVA-SA) to immobilize *Lentinus edodes* residue for Cd^{2+} removal. In a single-metal solution, the biosorption of Cd^{2+} reached equilibrium within 7 h which can be well described by a pseudo-second-order model, with equilibrium biosorption 0.2008 mg/g; 4–7 was the suitable pH value for Cd^{2+} biosorption by immobilized fungus, a much wider range than that required by mobilized fungus. Unlike the single-metal solution, the Cd^{2+} biosorption ratio in a two-metal solution increased linearly as the pH values increased. When the concentration of the interferential ion (Cu^{2+}/Pb^{2+}) increased, Cd^{2+} biosorption decreased significantly ($p = 0.01$). Isotherm analysis showed that Cd^{2+} biosorption increased as the initial concentration in a single-metal solution increased. When the initial Cd^{2+} concentration in the two-metal solution increased, the biosorption increased at first, but when the Cd^{2+} concentration was over 90 mg/g, a remarkable decrease occurred. Langmuir, Freundlich, Dubinin–Radushkevich and Langmuir–Freundlich isotherm models were fit to the experimental data. The Langmuir model fit the Cd^{2+} isotherm biosorption best, with correlation coefficients of 0.9981 in Cd^{2+} single-metal solution and 0.9291 in a Cd^{2+}–Pb^{2+} solution. The D–R isotherm fit the Cd^{2+} isotherm biosorption in the Cd^{2+}–Cu^{2+} solution with a correlation coefficient of 0.9623.

Keywords: Immobilization; *L. edodes*; Two-metal solution; Interference; Cd^{2+}; Biosorption isotherm

1. Introduction

Sewage usually contains a variety of metal ions, so interference between ions cannot be ignored. Biosorption mainly relies on cell-surface functional groups, so if both competitor ions and target ions can combine with the same adsorption site, the existence of competitor ions will reduce the biosorption efficiency of the target ions. Due to the differences in affinity for the adsorption site, different competitive cations will have different effects cationic on the adsorption of target ions. Sar [1] found that effect of different ions on uranium and thorium adsorption by *pseudomonas* bacteria was as follows: $Fe^{3+} > Th^{4+} > Fe^{2+} > Cu^{2+} > Al^{3+}$ and the adsorption of uranium was reduced by 80% when Fe^{3+} was present. In addition, cations compete with anions in biological cell walls, which leads to a decline in adsorption. The extent of this decline depends on the bonding force between the metal cations

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and the anions. The more intense the bonding force is, the lower the effect.

Microbial immobilization technology uses physical or chemical means to bind cells or enzymes located in a limited region, keeping them active and reusable. Initially, this technology was used in fermentation and then it was introduced to water treatment in the late 1970s. Now this technology is a research focus because it overcomes difficulties such as the small size of a free cell, the dissociation of biosorbents from water and secondary pollution. This technology has a high efficiency and stability and the biosorbent can be easily purified, and microbial immobilization has great prospects in the field of wastewater treatment.

Many studies have shown that various kinds of edible fungus can accumulate heavy metal ions effectively [3–5]. In this study, *Lentinus edodes* residue was used as an adsorbent. Polyvinyl alcohol-Na-alginate (PVA-SA) embedding technology was used to immobilize *L. edodes* residue for Cd^{2+} removal from both a single-metal and a two-metal solution and to investigate the effect of cross-interference metal ions on Cd^{2+} adsorption. The kinetics of the single-metal solution, the effect of solution pH on the two-metal solution system and the effect of cross-interference metal ions on Cd^{2+} biosorption by PVA-SA *L. edodes* beads were investigated. Finally, Langmuir, Freundlich, Langmuir-Freundlich and Dubinin–Radushkevich (D–R) isothermal adsorption models were used to fit the Cd^{2+} isothermal adsorption process in single-metal and two-metal solutions.

2 Materials and methods

2.1. Biomass

L. edodes residue was obtained by cutting off the edible part of *L. edodes* and then grinding and sieving to select particles between 35 and 100 mesh sieves. *L. edodes* residue powder was dried in an oven at 50±1°C for 2 h and then stored in desiccators.

2.2. PVA-SA L. edodes beads

Five grams of polyvinyl(PVA) and 1 g of alcohol-Na-alginate(SA) were added to 100 mL of distilled water, mixed and dissolved with heating. The solution was cooled to 45–50°C and 3 g of *L. edodes* powder was added. A saturated boric acid solution containing 2% $CaCl_2$ was added to the mixture with 7–9 syringe needles and continuous stirring. After immobilization for 24 h without disturbance, the PVA-SA *L. edodes* beads were obtained. The PVA-SA *L. edodes* beads were washed in distilled water and dried 2 h in an oven at a temperature of 50±2°C.

2.3. Metal ion solution

The lead stock solution was made by dissolving 1.5990 g of lead nitrate in 100 mL of nitric acid and adding distilled water to a final volume of 1000 mL. The cadmium stock solution was prepared by dissolving 0.100 g of cadmium in 1:1(V/V) HCl followed by 1 mL of concentrated HNO_3 and then diluting with distilled water to 100 mL. Copper was dissolved in 5 mL of 1:1 HNO_3 and diluted with distilled water to 100 mL to obtain the copper stock solution.

The metal solution was made by diluting a stock solution and using HCl and NaOH to regulate the pH value.

2.4. Biosorption experiments

Batch biosorption experiments were carried out in 250 mL shake flasks containing 25 mL of stock solution ($CdCl_2$ and/or $Cu(NO_3)_2$ and/or $Pb(NO_3)_2$). Analytical grade reagents were used in all cases. Flasks were agitated in an HY-5 orbital shaker at 25°C.

To investigate Cd^{2+} biosorption kinetics by PVA-SA *L. edodes* beads, samples of 1.5 mL were taken at different times from the mixed adsorption flasks. The effect of the initial solution pH was tested by adjusting the pH of either the $CdCl_2$ and $Cu(NO_3)_2$ solution or the $CdCl_2$ and $Pb(NO_3)_2$ solution to between 1.0 and 7.0. The initial metal concentrations were measured after pH adjustment and before the addition of the PVA-SA *L. edodes* beads. To investigate the effect of interferential metal ions on Cd^{2+} biosorption, the Cd^{2+} concentration was fixed at 10 mg/L, the pH was fixed at 5–6 and the varied interferential metal ion (Cu^{2+}/Pb^{2+}) concentration varied from 0 to 30 mg/L.

Adsorption thermodynamics tests were conducted by varying the Cd^{2+} concentration from 0 to 120 mg/L and fixing the solution pH value at 5–6. The interferential metal ion concentration was 0 mg/L or 10 mg/L. The contents of the flask were shook and allowed to adsorb for 7 h at 25°C and the filtrate was analyzed for Cd^{2+}, Pb^{2+} and Cu^{2+} by flame atomic adsorption spectroscopy (AAS).

3. Results and discussion

The adsorption may be described by [6,7]:

$$q_e = (C_o - C_e)V / M \tag{1}$$

and the biosorption efficiency of metal ions may be calculated by

$$X = (C_0 - C_e) / C_0 \times 100 \tag{2}$$

where q_e is the amount of the heavy metal adsorbed onto the unit amount of the biomass (mg/g); X is the biosorption ratio (%); C_0 and C_e are the concentrations of heavy metal in the initial and equilibrium solution (mg/g); M is the mass of PVA-SA beads and V is the volume of the aqueous phase.

3.1. Adsorption curve and adsorption kinetics of Cd^{2+}in a single-ion solution

3.1.1. Cd^{2+} adsorption curve

Contact time is an important parameter for successful biosorption. Fig. 1 shows that the Cd^{2+}adsorption process can be divided into a fast phase and a slow phase. The adsorption rate increases from 0 to 53.51% within 1 h and increases gently after 1 h. Adsorption equilibrium was reached after 7 h with 0.2008 mg/g. The time required to reach equilibrium when adsorbing Cd^{2+} is much longer for when the mushroom is immobilized (7 h) than when it is mobile (1 h) [8].

3.1.2. Adsorption kinetics

Two widely used kinetic models for investigating the mechanism of adsorption are the pseudo-first-order and pseudo-second-order kinetic models [9–11]. Many studies show that the biosorption process of various bivalent heavy metal ions can be described by the pseudo-second-order dynamic equation [9,10,12]:

$$q_e = q_m bC_e / (1 + bC_e) \tag{3}$$

where q_e and q_t refer to the amount of metal ions adsorbed at equilibrium at any time(mg/g) and k_2 is the equilibrium rate constant of the pseudo second-order adsorption (g/mg·min). Plots of t/q_t vs. t are shown in Fig. 2. The correlation coefficient (R^2) is 0.9946, which indicates that pseudo second-order adsorption model is suitable for describing the adsorption of Cd^{2+} onto PVA-SA bead.

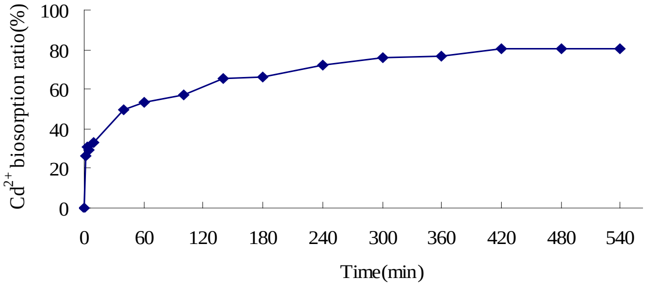

Fig. 1. Effect of time on Cd^{2+} biosorption onto PVA-SA beads at a biomass concentration of 20 mg/L, pH of 5–6 and 25°C.

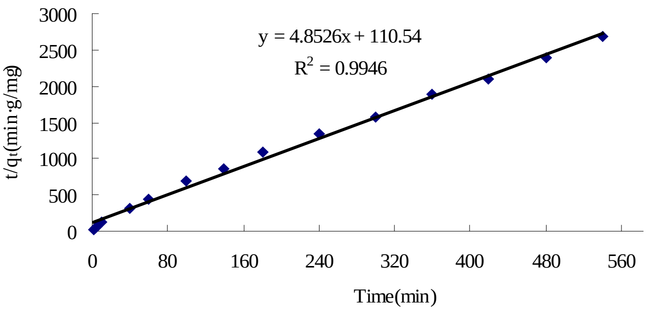

Fig. 2. Second-order modelling of Cd^{2+} biosorption onto PVA-SA beads at a biomass concentration of 20 mg/L, pH of 5–6 and 25°C.

3.2. Impact of pH

The pH is an important factor for metal ions adsorption. Fig. 3 shows the relation between pH values and the Cd^{2+} adsorption ratio in a solution of Cd^{2+}, Cd^{2+}–Cu^{2+} and Cd^{2+}–Pb^{2+}. When the pH value is small, the Cd^{2+} adsorption rate is lower in all three solutions. When the pH is low, H^+ (or H_3O^+) ions will compete with the metal ions at binding sites on the surface of the sorbent, thereby reducing the adsorption of the metal ions. When the pH value changes from 0 to 4, the Cd^{2+} adsorption in the single-metal solution increases rapidly. When the pH is above 4, the Cd^{2+} adsorption curve becomes approximately flat. At a pH of 4–7, a range that encompasses the typical pH range of mobilized fungi (pH 6–7) [8], the adsorption rate of Cd^{2+} in a two-metal-ion solution is much smaller than that in single-metal solution (Fig. 3). The Cd^{2+} adsorption rate increases linearly with increasing pH values. This increase may be the result of competition between the ions. Liu [13] pointed out that a strong dependence on pH suggests that the functional groups on the surface of the organisms and that the oxidation state and/or coordination of the metal ions may play an important role in the process of heavy metal adsorption by organisms.

3.3. Influence of the initial concentration of interferential ions on Cd^{2+} adsorption

Fig. 4 shows that different concentrations of Cu^{2+} or Pb^{2+} have a similar impact on Cd^{2+} adsorption. That is, a low concentration of interferential ions has a strong negative effect on Cd^{2+} biosorption, while for high concentrations, this negative disturbance is mild. Different interferential ions have different points of inflection on the Cd^{2+} adsorption curve. The points of inflection is at 1 mg/L in the Cu^{2+}–Cd^{2+} solution and 5 mg/L in the Pb^{2+}–

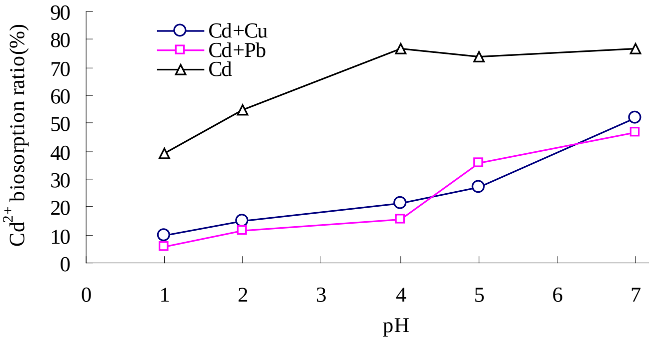

Fig. 3. Effect of pH on Cd^{2+} biosorption onto PVA-SA beads at a biomass concentration of 8 mg/L and 25°C.

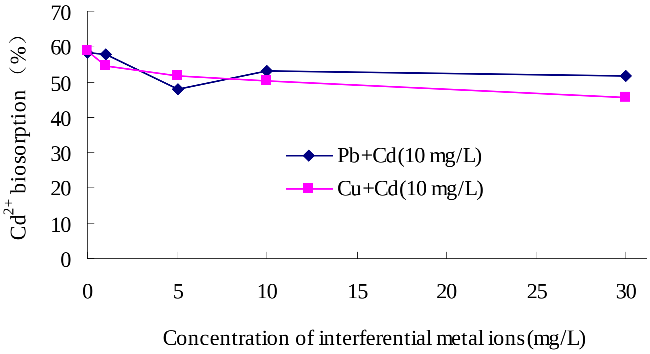

Fig. 4. Effect of interferential metal ions on Cd^{2+} biosorption onto PVA-SA beads at a biomass concentration of 8 mg/L and 25°C.

Cd^{2+} solution. After the turning points, the Cd^{2+} adsorption curve decreases more rapidly in the Cu^{2+}–Cd^{2+} solution than in the Pb^{2+}–Cd^{2+} solution. On one hand, these differences may be related to the physical and chemical characteristics of Cu^{2+}, Pb^{2+} and Cd^{2+} such as atomic number, atomic radius and the bonding of the ions with hydroxide groups on the surface of the absorbent. On the other hand, the differences may be ascribed to the functional groups on the surface of the absorbent. Tables 1–4 show the adsorption data for Cd^{2+} and the variance analysis for the three solutions described above. Both Pb^{2+} and Cu^{2+} significantly inhibit Cd^{2+} adsorption onto PVA-SA beads. This result is similar to the impact of competing ions on adsorption onto *auricularia* [18]. Table 2 shows that Pb^{2+} has a significant influence on Cd^{2+} adsorption onto

Table 1
Effect of Pb^{2+} concentration on Cd^{2+} biosorption onto PVA-SA beads

Cd^{2+} concentration (mg/L)	Pb^{2+} concentration (mg/L)	C_e (mg/L)	q_e (mg/g)	Cd^{2+} biosorption ratio (%)
10	0	4.1545	0.7307	58.50
10	1	4.2200	0.7225	57.80
10	5	5.1970	0.6004	48.03
10	10	4.680	0.6650	53.20
10	30	4.8210	0.6474	51.79

Table 2
Variance analysis of the effect of Pb^{2+} on Cd^{2+} biosorption

Variance source	Sum of squares	Degree of freedom	Mean square	F	Sig.
Between group	226.041	4	56.510	72.818	0.000
Within group	7.760	10	0.776		
Total variance	233.801	14			

Table 3
Effect of Cu^{2+} concentration on Cd^{2+} biosorption onto PVA-SA beads

Cd^{2+} concentration (mg/L)	Cu^{2+} concentration (mg/L)	C_e (mg/L)	q_e (mg/g)	Cd^{2+} biosorption ratio (%)
10	0	4.1505	0.7312	58.50
10	1	4.5550	0.6806	54.45
10	5	4.8125	0.6484	51.88
10	10	4.9600	0.6300	50.40
10	30	5.4470	0.5691	45.53

Table 4
Variance analysis of the effect of Cu^{2+} on Cd^{2+} biosorption

Variance source	Sum of squares	Degree of freedom	Mean square	F	Sig.
Between group	277.535	4	69.384	155.922	0.000
Within group	4.450	10	0.445		
Total variance	281.985	14			

PVA-SA beads ($p<0.01$). When the Pb^{2+} concentration is 5 mg/L, the negative effect on Cd^{2+} biosorption is strongest (Table 1) because Pb^{2+} competes with Cd^{2+} for binding sites. The presence of 1–30 mg/L Cu^{2+} decreases the Cd^{2+} adsorption rate by 12.07% (Table 3, Table 4). When the concentration of the interferential ions is greater than 30 mg/L, Cd^{2+} biosorption is more strongly restrained by Cu^{2+} than by Pb^{2+} (Fig. 4).

3.4. Adsorption thermodynamics

An accurate adsorption model is very important for optimizing the adsorption process. Langmuir, Freundlich, D–R and Langmuir–Freundlich isothermal adsorption models have been widely used for wastewater analysis. The Langmuir model assumes that the adsorption surface is uniform, that there is no interaction between the adsorbed molecules and that the adsorption occurs on a single-layer. The Langmuir model expression is as follows:

$$q_e = q_m b C_e / \left(1 + b C_e\right) \tag{4}$$

where q_e is the adsorption capacity (mg/g adsorbent), q_m is the maximum adsorption capacity (mg/g adsorbent), C_e is the metal ion concentration left in the solution when adsorption equilibrium is reached (mg/L) and b is the Langmuir adsorption constant, which is related to the affinity of the heavy metal ions for the binding site. q_m is a coefficient characteristic of the adsorbent and is determined by the nature of the adsorbent only, not the concentration of the adsorbent. The Freundlich model is an experimental model that assumes ions adsorption occurs as a monolayer and the absorbing surface is not uniform. The expression is as follows:

$$q_e = k C_e^{1/n} \tag{5}$$

where k and n are adsorption constants. k is related to the adsorption capacity of the adsorbent and n indicates the difficulty of adsorbing the metal ions onto the absorbent. When $1/n$ is between 0 and 1, the metal ions adsorb easily. The other parameters are the same as in Eq. (4). The expression for the Langmuir–Freundlich isotherm is as follows [14]:

$$q_e = q_m b_{LF} C_e^{\theta} / \left(1 + b_{LF} C_e^{\theta}\right) \tag{6}$$

where b and n are adsorption parameters and the other parameters are the same as in Eq. (4).

The expression of the D–R isotherm is as follows [15,16]:

$$\ln q_e = \ln q_m - \beta \varepsilon^2 \tag{7}$$

$$\varepsilon = RT \ln\left(1 + 1/C_e\right) \tag{8}$$

where β is a constant related to the energy ($kJ^2 \cdot mol^{-2}$), ε is the Polanyi potential and the other parameters are the same as in Eq. (4). The adsorption energy, E_s, can be calculated with the following equation:

$$E_s = (2\beta)^{-1/2} \tag{9}$$

3.4.1. Isothermal adsorption curves

Fig. 5 and Fig. 6 show the relation between the adsorption of the metal ions and the initial concentration of Cd^{2+} in the single-metal and two-metal solutions. In Cd^{2+} single-metal solution, the amount of Cd^{2+} adsorbed increases with the initial concentration of Cd^{2+}. The increase rate decreases with Cd^{2+} initial concentration increasing. When the initial concentration of Cd^{2+} is 120 mg/g, the adsorption capacity reaches 6.0469 mg/g, which is much higher than the maximum adsorptive capacity in the two-metal solution. The slope of the curve represents the affinity between the adsorbent and the metal ions. The greater the slope is, the greater the affinity. In the single-metal solution, the slope of the curve is larger and the presence of Pb^{2+} reduces the slope of the Cd^{2+} adsorption curve greatly (Fig. 5). When the Cd^{2+} concentration is less than 90 mg/g, the slope of the Cd^{2+} adsorption curve is reduced slightly (Fig. 6). When the Cd^{2+} concentration is greater than 90 mg/g, the slope of

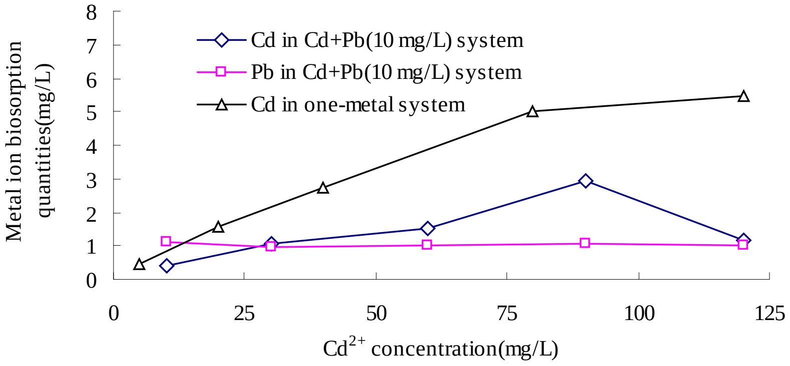

Fig. 5. The relationship between metal ion biosorption quantities and initial Cd^{2+} concentration in Cd^{2+} single-metal solution and Cd^{2+}–Pb^{2+} solution, at a biomass concentration of 8 mg/L, pH of 5–6 and 25°C.

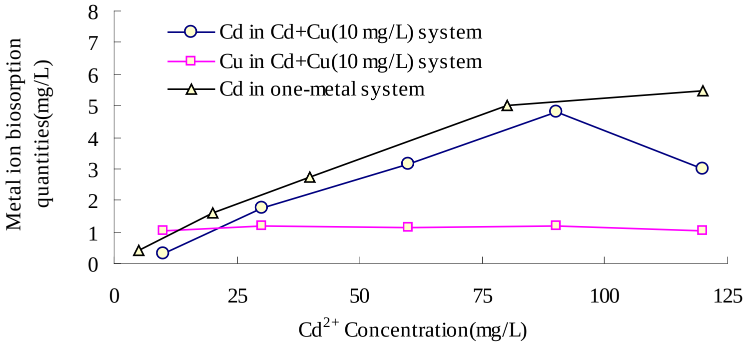

Fig. 6. The relationship between metal ion biosorption quantities and initial Cd^{2+} concentration in Cd^{2+} single-metal solution and Cd^{2+}–Cu^{2+} solution, at a biomass concentration of 8 mg/L, pH of 5–6 and 25°C.

the curve falls remarkably in both two-metal solutions. When the concentration of Cd^{2+} is 90 mg/g, the quantity of Cd^{2+} adsorbed in both the Cd^{2+}–Pb^{2+} solution and Cd^{2+}–Cu^{2+} solution reaches the maximum (Cd^{2+}–Pb^{2+}: 2.9544 mg Cd/g; Cd^{2+}–Cu^{2+}: 4.8075 mg Cd/g). Ferraz et al. [17] studied the effect of interferential ions on metal ions adsorbing to Saccharomyces cerevisiae and confirmed that there was competition between the metal ions for adsorption sites on the surface of cell walls. Fig. 5 and Fig. 6 indicate that when the Cd^{2+} concentration is low (0–90 mg/g), the competition for adsorption sites mostly comes from interferential ions. When the Cd^{2+}concentration is high (>90 mg/g), the amount of adsorbing Pb^{2+} and Cu^{2+} is basically unchanged, but the number of PVA-SA beads that are adsorbing Cd^{2+} decreases greatly because of an overabundance of Cd^{2+}, which means competition and repulsion between Cd^{2+} on the cell wall play an important role in the decrease in Cd^{2+} adsorbing.

3.4.2. Isothermal adsorption model

To determine the similarities and differences of the adsorption mechanisms and the adsorption laws for Cd^{2+} in single-metal and two-metal solutions, Langmuir, Freundlich, Langmuir–Freundlich and D–R isothermal adsorption models were fit to the experimental data (Tables 5–7). In the single-metal solution, all of the models except the D–R model can describe the isothermal adsorption of Cd^{2+}. Langmuir model is the best, with a correlation coefficient of 0.9981 and theoretical q_m of 6.4475 mg/g, which is slightly higher than the adsorbed amount when the initial concentration of Cd^{2+} is 120 mg/g (6.0469 mg/g). Langmuir–Freundlich model is the second-best model and the D–R model is the worst. Langmuir model fits the isothermal adsorption of Cd^{2+} in the Cd^{2+}–Pb^{2+} solution, with a correlation coefficient of 0.9291 and theoretical q_m of 2.6788 mg/g, which is slightly lower than the experimental q_m (2.9544 mg/g). The D–R model fits the isothermal adsorption of Cd^{2+} in the Cd^{2+}–Cu^{2+} solution with a correlation coefficient of 0.9623.

Table 5
Parameters of isotherm models for Cd^{2+} removal from one-metal system

Thermodynamic models	Parameters		R^2
	Symbol	Values	
Langmuir	q_m (mg/g)	6.4475	0.9981
	b	0.05916	
Freundlich	k	0.43102	0.9823
	n	1.5099	
Langmuir–Freundlich	q_m (mg/g)	11.3636	0.9913
	Θ	0.8485	
	b_{LF}	0.03437	
D–R	q_m (mg/g)	53.4529	0.4072
	B	—	
	E_s (kJ/mol)	10000	

Table 6
Parameters of isotherm models for Cd^{2+} removal from Cd+Pb system

Thermodynamic models	Parameters		R^2
	Symbol	Values	
Langmuir	q_m (mg/g)	2.6788	0.9291
	B	0.02524	
Freundlich	K	0.1778	0.6146
	N	1.8889	
Langmuir–Freundlich	q_m (mg/g)	20000	0.6146
	Θ	0.5294	
	b_{LF}	8.889E-07	
D–R	q_m (mg/g)	1.6318	0.7389
	B	1×10-5	
	E_s (kJ/mol)	223.607	

Table 7
Parameters of isotherm models for Cd^{2+} removal from Cd+Cu system

Thermodynamic models	Parameters		$R2$
	Symbol	Values	
Langmuir	q_m (mg/g)	–3.2051	0.9013
	B	–0.01274	
Freundlich	K	0.08490	0.7316
	N	1.0851	
Langmuir–Freundlich	q_m (mg/g)	33.3333	0.7323
	Θ	0.9655	
	b_{LF}	0.0024	
D–R	q_m (mg/g)	3.6616	0.9632
	B	3×10^{-5}	
	E_s (kJ/mol)	129.099	

4. Conclusions

With an adsorptive capacity of 0.2008 mg/g, PVA-SA beads absorb Cd^{2+} and reach equilibrium in a single-metal solution after 7 h, a much longer time than is required with mobilized fungus. Pseudo-second-order dynamic equations describe the process of PVA-SA beads adsorbing Cd^{2+} in a single-metal solution with a correlation coefficient of 0.9946. In a single-metal solution, when the pH changes from 0 to 4, the Cd^{2+} adsorption ratio increases rapidly, but when the pH is above 4, the Cd^{2+} adsorption curve become approximately flat. A pH between 4 and 7 is the optimal rang for Cd^{2+} adsorption. This range is much wider than the range for mobilized fungi. In a two-metal solution, the Cd^{2+} adsorption ratio is much smaller than in a single-metal solution and the Cd^{2+} adsorption ratio increases linearly with increasing pH. The presence of Cu^{2+} or Pb^{2+} reduces Cd^{2+} adsorption significantly and when the Cd^{2+} concentration is low, the interference of the competing metal ions is large. The interference lessens when the Cd^{2+} concentration is increased. When the initial concentration of Cd^{2+} is 120 mg/g, the Cd^{2+} adsorbed from a single-metal solution reaches 6.0469 mg/g. The number of PVA-SA beads adsorbing Pb^{2+} and Cu^{2+} remains basically unchanged in a two-metal solution. When the Cd^{2+} concentration is less than 90 mg/g, the presence of Pb^{2+} has a negative effect on Cd^{2+} adsorption, but when the Cd^{2+} concentration is greater than 90 mg/g, both Pb^{2+} and Cu^{2+} reduce Cd^{2+} adsorption greatly. When the Cd^{2+} concentration reaches 90 mg/g, Cd^{2+} adsorption from the two-metal solution reaches a maximum (Cd^{2+}–Pb^{2+}: 2.9544 mg Cd/g; Cd^{2+}–Cu^{2+}: 4.8075 mg Cd/g). The Langmuir isotherm model can describe the Cd^{2+} isothermal adsorption process in a single-metal solution with a correlation coefficient of 0.9981. The second-best model is the Langmuir–Freundlich model and the D–R model is the worst. The Langmuir model fits the Cd^{2+} isothermal adsorption process in the Cd^{2+}–Pb^{2+} solution ($R^2 = 0.9291$) with a theoretical q_m of 2.6788 mg/g, which is slightly smaller than the experimental value (2.9544 mg/g). The D–R model can fit the Cd^{2+} isothermal adsorption process in the Cd^{2+}–Cu^{2+} solution and the correlation coefficient is 0.9623.

Acknowledgments

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