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ORIGINAL PAPER

Study of hybrid immobilized biomass of *Pleurotus sajor-caju* and *Jasmine sambac* for sorption of heavy metals

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Abstract The current study deals with evaluation of biosorption feasibility for removal of Cu(II) and Zn(II) by hybrid immobilized biosorbent of *Pleurotus sajor-caju* and *Jasmine sambac*. Batch adsorption experiments were carried out to assess the effect of pH, initial metal concentration, biomass dose, temperature and time. The biosorption efficiency of Cu(II) and Zn(II) ions for hybrid immobilized biosorbent increases with rising pH values. The hybrid immobilized biosorbent illustrated the highest biosorption capability at pH 5 for Cu(II), 6 for Zn(II), at 0.05 g/100 mL dose and 100 mg/L initial metal concentration of both ions. Uptake kinetics followed the pseudo-second-order model and equilibrium was described by Langmuir and Freundlich isotherms. Adsorption ratios of Cu(II) and Zn(II) were best fitted to Langmuir isotherm. The best temperature for ion uptake was found to be 30 °C.

Keywords Batch mode · Biosorption · Copper · pH · Zinc

Introduction

Water is one of the few resources used by all earth's inhabitants. As a consequence, the pollution that spews from industries in Asia, a fire in Australia, volcanic eruption in Cango or weathering of rocks in Japan can have a detrimental impact on people and the environment locally or an ocean. The world's rivers carry as much as 24 million tons of sediments to the ocean each year. In addition to natural sources, battery production, petrochemicals, refineries, mining, fertilizers, fossil fuel burning, steel, wood pulping and all other industries are also potential source of pollution (Busari et al. 2012). Minamata tragedy in Japan was due to pollution of heavy metals in aquatic streams.

Presence of Cu(II) and Zn(II) may cause many harmful effects such as deposits in brain, skin and liver (Mukhopadhyay et al. 2007; Vullo et al. 2008) by Cu(II) and respiratory incapacitation such as increased breathing rate, coughing, decrease in oxygen uptake efficiency, muscular stiffness and loss of appetite by Zn(II).

Many techniques have been introduced for control of water pollution including chemical precipitation, adsorption, membrane separation, evaporation, reverse osmosis, ion exchange, ultrafiltration and electrodialysis (Shah et al. 2009).

But most of these methods have several inherent limitations including incomplete metal removal, sensitive operating conditions and production of secondary sludge (Dursun et al. 2003). Moreover, economical and technical factors also limit the feasibility of such processes (Jnr and Spiff 2005; Bai et al. 2010). Thus, an imperative requirement for the development of innovative and low-cast technique leads to the discovery of biosorption.

Biosorption is the aptitude of biological materials to remove heavy metals through metabolically mediated or physicochemical pathways of uptake (Norton et al. 2004; Cao et al. 2010). Many metal-binding mechanisms are involved in biosorption such as micro precipitation, complexation, physical adsorption and coordination (Babel and Kurniawan 2003).

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It has been revealed that microorganisms such as fungi can be used for removal of heavy metals because of their low cost, efficiency and eco-friendly choice (Ahlawalia and Goyal 2005; Cabuk et al. 2005).

Cell immobilization is a wonderful technique to fix biomass on suitable natural or synthetic material support for a range of physical and biochemical unit operations (Al-Rub et al. 2004). Natural or synthetic polymers such as silica gel, alginate or polyurethane (Chen et al. 2006; Anjana et al. 2007) can be used for immobilization.

Immobilization of biomass can solve many issues by proposing ideal size, mechanical potency and inflexibility (Pan et al. 2005). The benefits also include renewal of biomass, simple solid–liquid disjointing and minimal clogging in continuous-flow schemes (Anjana et al. 2007).

In the present study, hybrid immobilized biosorbent is developed by merging two biosorbents, namely the hyphal biomass of *P. sajor-caju* and fibrous network of *J. sambac*, by skill of immobilization in which these two perform as complementary partners.

Although copious natural resources of cellulosic characteristics and hyphal biomass have been suggested as biosorbents, but very less work has been done by their hybrid immobilization. Therefore, this study is focused on biosorption of Cu^{2+} and Zn^{2+} by using hybrid immobilized biosorbent of *P. sajor-caju* and *J. sambac*. The current study was conducted in March 2009 at Analytical Chemistry Laboratory, University of Agriculture, Faisalabad.

Materials and methods

The biosorption of Cu(II) and Zn(II) by hybrid immobilized biosorbent of *P. sajor-caju* and *J. sambac* was studied according to method described by Iqbal et al. (2007). The fungus *P. sajor-caju* was grown in Mushroom Laboratory and *J. sambac* waste biomass was collected from Institute of Horticultural Sciences, University of Agriculture, Faisalabad.

Preparation and sieving of biomass

Water-washed biomass was oven-dried at 65 °C for 72 h. Dried biomass was cut and ground with the help of food processor (Kenwood, FP-220) and then sieved through Octagon sieve (OCT-DIGITAL 4527-01). Sieving was performed to eliminate any large size particles and to get adsorbent of known and homogenous size.

Microorganism and culture medium

The fungal strain used in the present study as test fungus (*P. sajor-caju*) was obtained by subculturing on potato dextrose agar slants. for preparation of hyphal suspension, 7-day old cultures cultivated on potato dextrose agar plates at 35 ± 2 °C were used. Hybrid matrix was prepared according to the method described by Iqbal et al. 2007.

Immobilization of biomass

One gram of each live and dead hybrid biosorbent matrix of *P. sajor-caju* and *J. sambac* was dissolved in 100 mL of 2 % sodium alginate. The mixtures were homogenized by high-speed mixing. These mixtures were then reacted carefully with 0.1 M $CaCl_2$ to obtain the uniform-sized beads of biomass. The hybrid immobilized live and dead biosorbents were used in evaluation of all experimental parameters.

Cu(II) and Zn(II) solutions

Stock solutions (100 mg/L) of Cu(II) and Zn(II) were prepared by dissolving $ZnSO_4 \cdot 7H_2O$ and $CuSO_4 \cdot 5H_2O$ in 1,000 mL of deionized distilled water (DDW), respectively.

Batch biosorption studies

In this study, fixed volume of Cu(II) and Zn(II) solutions (100 mL of 100 mg/L) were taken in each 250-mL conical flasks. The influence of various factors such as pH, 1–5 for Cu(II) and 1–6 for Zn(II), biosorbent dose (0.05–0.4 g/100 mL), initial metal concentration (25–800 mg/L), contact time (15–1,440 min) and temperature(30–60 °C) on sorption process was studied in batch mode.

Determination of Cu(II) and Zn(II) contents in the solutions

Atomic absorption spectrophotometer was used for the determination of Cu(II) and Zn(II) concentration in the aqueous medium. Concentration difference process was used for calculating metal uptake. Mass balance equation was used for calculating metal uptake.

$$q = V(C_i - C_e)/M$$

Statistical analysis

All data records symbolize the mean of three independent values. Microsoft Excel 2007 was used to perform statistical analysis.

Results and discussion

Influence of pH

pH is a significant parameter that influences biosorption process (Dursun 2006). pH of solution effects metal-binding sites of cell surface. In order to assess the effect of this factor, batch equilibrium studies were performed by using dead and live hybrid immobilized biosorbents of *P. sajor-caju* and *J. sambac* with various initial pH value ranges (1–5 for Cu^{2+} and 1–6 for Zn^{2+}). The pH > 5 for Cu(II) and >6 for Zn(II) was avoided because precipitation of metal hydroxides have been reported to occur at these pH values (Lisa et al. 2004; Xiao and Huang 2009). The results showed that metal uptake by dead and live hybrid immobilized biosorbent of *P. sajor-caju* and *J. sambac* increase with rise in pH. Dead and live hybrid immobilized biosorbent exhibited maximum equilibrium uptake of Cu(II) (90.89 and 84.56 mg/g) and Zn(II) (89.98 and 83.88 mg/g) ions at the highest investigated pH values (5 for Cu(II) and 6 for Zn(II)). This may be attributed to the fact that with increase in pH, number of H^+ ions decline and positively charged metal ions get chance to easily bind the biomass surface (Souag et al. 2009). At low pH values, concentration of H^+ ions is very high, they compete with metal ions and get priority to bind the surface, leading to reduction of metal uptake (Ajmal et al. 2000; Annadurai et al. 2002).

At pH values lower than 5 for Cu(II) and lower than 6 for Zn(II), removals were low. Similar trend at lower pH values is previously studied (Bayramoglu and Arica 2008; Rani et al. 2010). As the hybrid immobilized biosorbent of *P. sajor-caju* and *J. sambac* showed maximum adsorption for Cu(II) and Zn(II) ions at pH 5 and 6, respectively, these pH ranges were used for further studies (Fig. 1a, b).

Effect of initial metal concentration

The significance of this parameter is linked to the optimum concentration combined with the maximum efficiency in the sorption technique. It supplies important driving force to overcome mass transfer resistance of Cu(II) or Zn(II) between aqueous solution and hybrid immobilized biosorbent (Xue-jiang et al. 2006; Zubair et al. 2008). Adsorption abilities of different metals are different at different initial metal concentrations. The influence of initial concentration

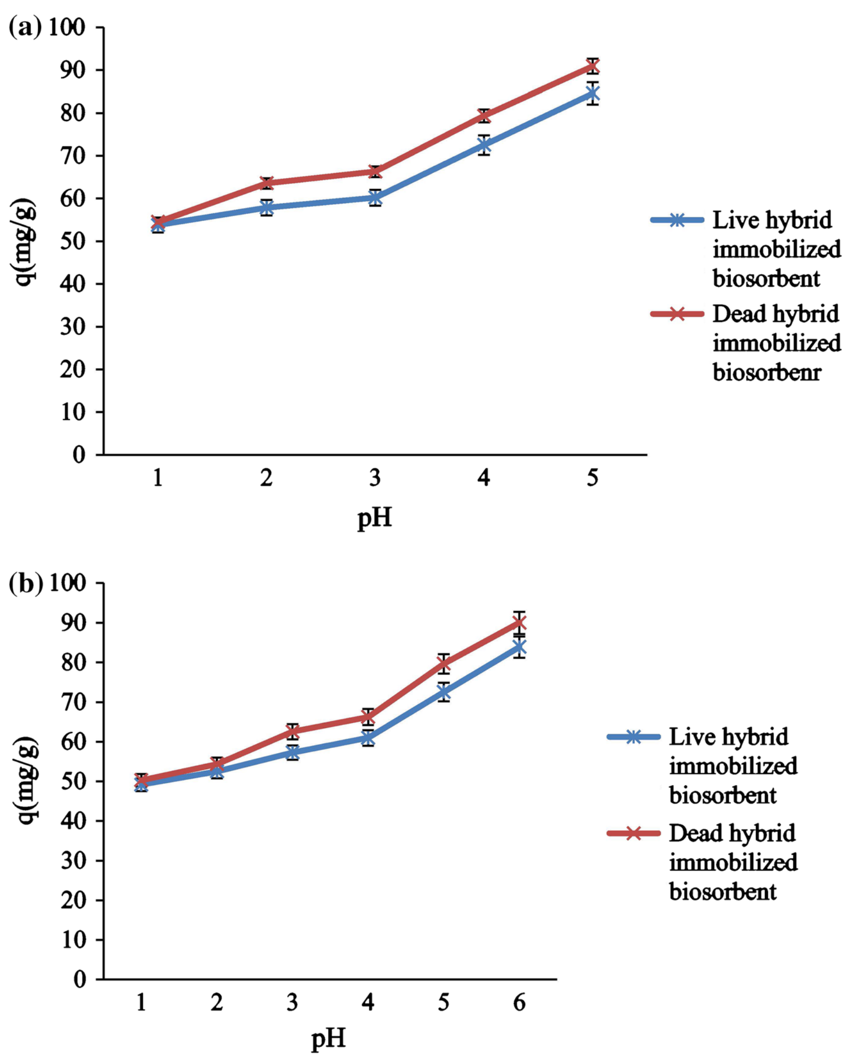

Fig. 1 **a** Effect of pH on uptake of Cu(II) by hybrid immobilized biosorbent. **b** Effect of pH on uptake of Zn(II) by hybrid immobilized biosorbent

on uptake capacities of Cu(II) and Zn(II) was studied by dead hybrid immobilized biosorbent of *P. sajor-caju* and *J. sambac* at pH 5 and 6 for Cu(II) and Zn(II), respectively, by changing the concentration of the system from 25 to 800 mg/L. Results obtained in this study showed that there is a linear relationship between sorption capacity and initial metal ion concentration, and this is also supported by the literature (Ashraf et al. 2010). This study also depicted an inversely proportional relationship between percentage removal and initial metal concentration, as reported previously (Bai et al. 2010). When metal ions at very low concentration are applied to biosorbent, then greater metal uptake occurs, because large number of vacant sorption sites is available for metal binding (Ashraf et al. 2010). At higher concentration, low-metal uptake may be attributed to saturation of sorption sites. Adsorption processes and different ion exchange mechanisms are involved in metal-uptake process (Busari et al. 2007) (Fig. 2).

Adsorption isotherms

Langmuir and Freundlich models were used to describe biosorption for single metal system. Langmuir model is based on monolayer adsorption over a homogenous adsorbent surface (Bai et al. 2010). Uptake capabilities and common equilibrium sorption procedure can also be explained by Langmuir model. For achievement of equilibrium data, initial concentrations of Cu(II) and Zn(II) were changed, but mass of adsorbent in all the samples was kept constant. The Langmuir model takes the form of equation:

$$C_e/q_e = 1/X_m K_L + C_e/X_m$$

In this study, Langmuir isotherm model proved to be in good correlation with results of initial metal ion concentration. The Freundlich relationship is based on heterogeneous adsorption. It does not indicate a finite uptake capacity of the sorbent and can thus only be reasonably applied in the low to intermediate concentration ranges. Freundlich equation is:

$$\log q_e = 1/n \log C_e + \log k$$

R^2 values achieved by Langmuir model showed that Freundlich isotherm model is not applicable because R^2 value is <0.98 (Table 1).

Effect of biosorbent dose

Biosorbent dose is a considerable factor that should be measured for successful metal sorption. Biosorption of

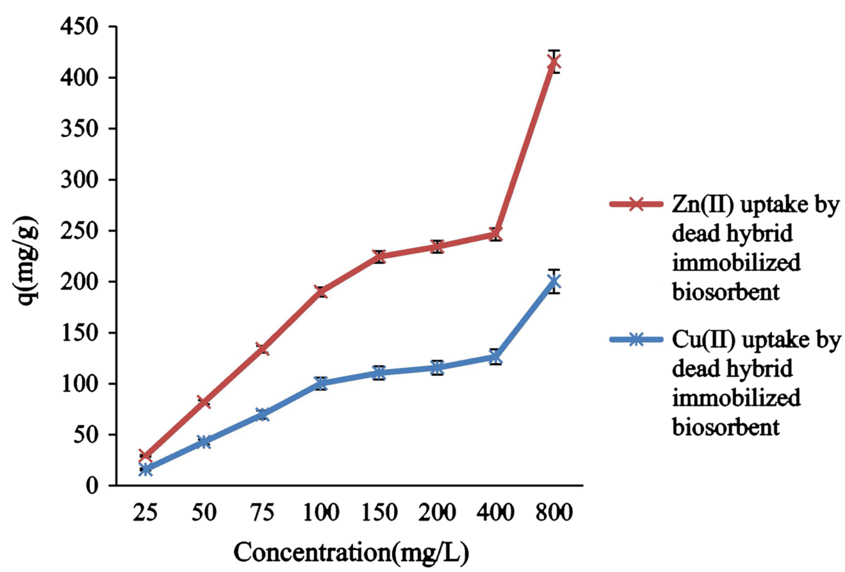

Fig. 2 Effect of concentration on uptake of Cu(II) and Zn(II) by hybrid immobilized biosorbent

Table 1 Langmuir and Freundlich isotherm constant for Cu^{2+} and Zn^{2+} dead hybrid immobilized biosorbent

Metals	Langmuir isotherm parameters			Experimental	Freundlich isotherm parameters			
	X (q_{max}) mg/g	K_L (L/mg)	R^2	q (mg/g)	q (mg/g)	K (mg/g)	R^2	1/n
Cu	217.39	0.0121	826	217.21	200.35	18.71	4,837	3,723
Zn	227.27	0.0122	835	225.82	213.04	18.04	5,148	3,886

X = maximal sorption capacity, K_L = Langmuir isotherm constant, q = sorption capacity, R^2 = correlation coefficient, K and n = Freundlich constant (K indicates sorption capacity and n indicates sorption intensity)

Cu(II) and Zn(II) was investigated with varying biosorbent doses (0.05–0.4 g/100 mL), but pH 5 for Cu(II) and 6 for Zn(II), initial metal concentration (100 mg/L), and temperature (30 °C) were kept constant. Maximum uptake capacity of Cu(II) (90.89 mg/g) and Zn(II) (89.98 mg/g) was examined with biomass dose of 0.05 g/100 mL.

It was observed from the results that sorptive capacity decreased with increase in biosorbent dose. Similar results were proposed previously (Hanif and Bhatti 2012).

This may be due to reduction in amount of metal ion concentration as compared to vacant binding sites (Mukhopadhyay et al. 2007). The study also explored that percent removal of Cu(II) and Zn(II) increased with increase in biosorbent dose. It may be attributed to the fact that higher the biosorbent dose, greater the available surface area and higher the chances of percent removal of metals (Witek-Krowiak et al. 2011) (Fig. 3).

Influence of temperature

The effect of temperature on uptake of Cu(II) and Zn(II) by dead and live hybrid immobilized biosorbents of *P. sajor-caju* and *J. sambac* was studied by altering temperature between 30 and 60 °C and keeping all other parameters constant [pH 5 for Cu(II) and 6 for Zn(II), initial metal concentration 100 mg/L, dose 0.05 g/100 mL and contact time 24 h].

The maximum sorption capacity of live (84.56 mg/g) and dead (90.89 mg/g) hybrid immobilized biosorbents for Cu(II) was observed at 30 °C. This temperature was also optimum for highest removal of Zn(II) (83.88 and 89.98 mg/g) by live and dead hybrid immobilized biosorbents, respectively. It was observed that metal uptake capacity decreased by enhancing temperature. These results are in agreement with the previous literature

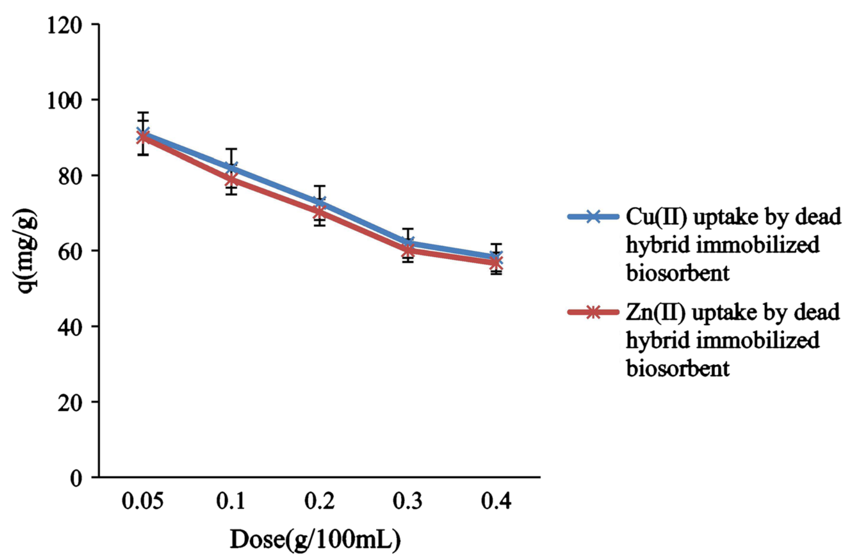

Fig. 3 Effect of dose on uptake of Cu(II) and Zn(II) by hybrid immobilized biosorbent

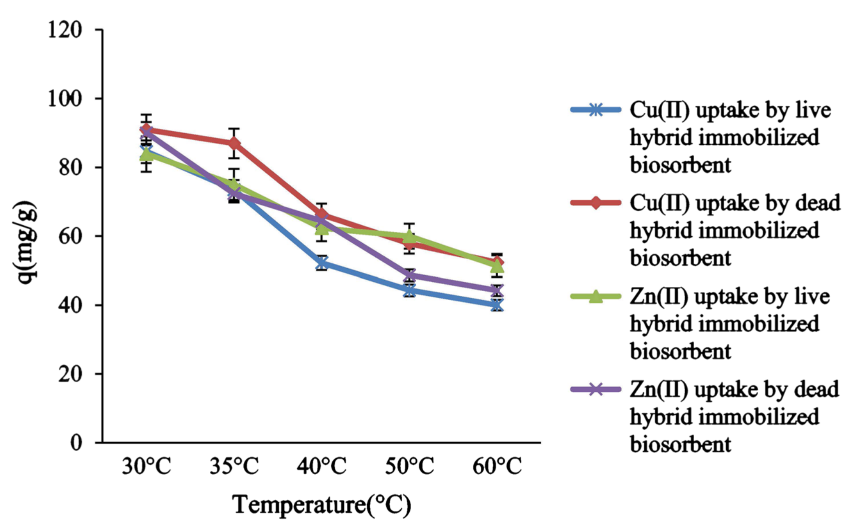

Fig. 4 Effect of temperature on uptake of Cu(II) and Zn(II) by hybrid immobilized biosorbent

(Shafqat et al. 2008). At increased temperature conditions, sorption of metal ions decreases because of decline in attractive forces between surface of biosorbent and metal ions (Jnr and Spiff 2005; Shafqat et al. 2008). It was also explored by the results that adsorption of Cu(II) and Zn(II) ions by live and dead hybrid immobilized biosorbents is exothermic in nature (Rostami and Joodaki 2002) (Fig. 4).

Effect of time

Contact time is one of the most significant parameter in the biosorption process. The influence of time (15–1,440 min) on the biosorption of Cu(II) and Zn(II) by dead and live hybrid immobilized biosorbents of *P. sajor-caju* and *J. sambac* is shown in Fig. 5. The experiment was performed at constant value of pH 5 for Cu(II) and 6 for Zn(II), biosorbent dose 0.05 g/100 mL, 30 °C temperature and initial metal concentration (100 mg/L).

It is depicted from the Fig. 5 that the percentage removal of Cu(II) and Zn(II) enhances by rising contact time. The sharp increase was observed in first 60 min. This may be attributed to extracellular binding and following sorption phase was slow due to intracellular binding (Bhatti et al. 2007). Maximum percentage removal for Cu(II) and Zn(II) by dead hybrid immobilized biosorbent of *P. sajor-caju* and *J. sambac* was 90.24 and 89.84 mg/g in first 120 min. Maximum percentage removal of Cu(II) and Zn(II) by live hybrid biomass was 83.95 and 82.18 mg/g, respectively, in first 120 min. The results are in agreement with previous study (Shafqat et al. 2008).

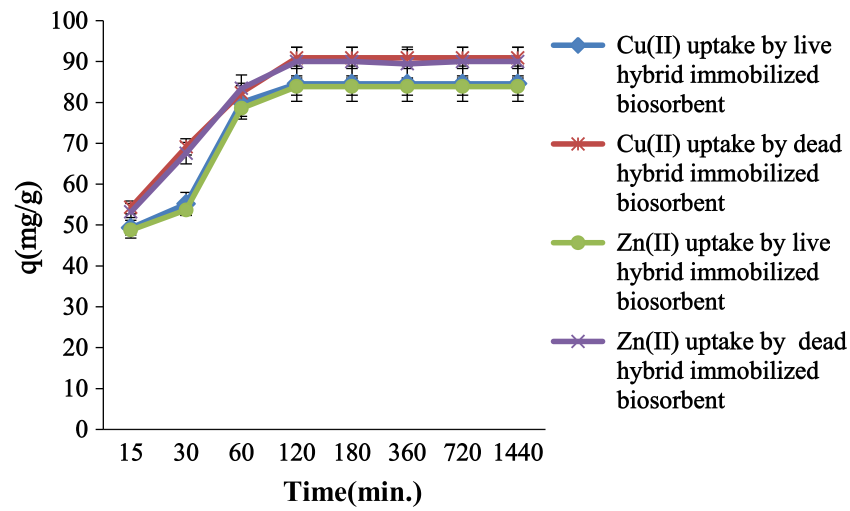

Fig. 5 Effect of time on uptake of Cu(II) and Zn(II) by hybrid immobilized biosorbent

Table 2 Comparison of pseudo-first-order and pseudo-second-order kinetic models for Cu^{2+} and Zn^{2+} biosorption on dead and live hybrid immobilized biomass

Metals	Biosorbent	Pseudo-first-order kinetic model			Experimental	Pseudo-second-order kinetic model		
		q_e (mg/g)	$K_{1,ads}$ (min^{-1})	R^2	q (mg/g)	q_e (mg/g)	$K_{2,ads}$ (g/mg min)	R^2
Cu^{2+}	Dead hybrid immobilized biosorbent	17.08	6.90×10^{-4}	0.273	90.25	90.90	2.13×10^{-3}	999
	Live hybrid immobilized biosorbent	24.40	4.61×10^{-4}	0.235	83.92	84.74	1.93×10^{-3}	999
Zn^{2+}	Dead hybrid immobilized biosorbent	17.54	6.90×10^{-4}	0.279	89.91	90.09	2.24×10^{-3}	999
	Live hybrid immobilized biosorbent	24.77	4.60×10^{-4}	0.2415	83.18	84.03	1.91×10^{-3}	999

q_e = amount of sorbed metal at equilibrium, $K_{1,ads}$ = rate constant for pseudo-first-order model, q = amount of sorbed metal, $K_{2,ads}$ = rate constant of pseudo-second-order model

After this equilibrium period, no considerable change in quantity of biosorbed metal ions was observed with increase in contact time, and thus, it was characterized as the optimum contact time. This fact is also supported by the literature (Arbanah et al. 2012). Active sorption sites in a system are in fixed amount and only one ion is adsorbed by one active site. The uptake of Cu(II) and Zn(II) by the hybrid immobilized biosorbent surface is fast initially, because a large number of vacant sites are available; but with passage of time, active sorption sites are occupied and uptake rate significantly decreases. The similar explanation was proposed by earlier workers (Arbanah et al. 2012; Abdulrasaq and Basiru 2010) (Fig. 5; Table 2).

Conclusion

In the present work, Cu(II) and Zn(II) removal by hybrid immobilized biomass of *P. sajor-caju* and *J. sambac* was evaluated. The study was carried out with dead and live hybrid immobilized biomass, and it was examined that dead hybrid immobilized biomass depicted the better results than live hybrid immobilized biomass. It was also noticed that pH, initial metal ion concentration, biosorbent dose, temperature and time influenced the uptake capacity of biosorbent. The maximum capacity of Cu(II) and Zn(II) was 90.89 and 89.98 mg/g, respectively, with dead hybrid immobilized biomass. Equilibrium data for studied metal ions followed the Langmuir model. The results indicated that hybrid immobilized biosorbent has improved metal uptake capacity.

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