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Biosorption of Strontium Ions by Magnetically Modified Yeast Cells

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The sorption of Sr^{2+} ions from aqueous solutions on magnetically modified fodder yeast (*Kluyveromyces fragilis*) cells and their subsequent desorption were studied. The Sr^{2+} sorption increased with increasing pH and reached a plateau between pH 4.0 and 7.0. The changes of temperature slightly influenced the sorption process. The sorption values were 19.5 mg g^{-1} and 53.5 mg g^{-1} for 10 mg L^{-1} and 40 mg L^{-1} Sr^{2+} solutions respectively after 20 min incubation at a pH higher than 4. The Langmuir isotherm was successfully used to fit experimental data; the maximum adsorption capacity was 140.8 mg g^{-1} under optimal conditions. The adsorbed Sr^{2+} ions can be desorbed with nitric acid (0.1 mol L^{-1}).

Keywords adsorption; desorption; magnetically modified yeast cells; strontium

INTRODUCTION

One of the most important problems during the reprocessing of spent fuels in the nuclear industry is the management and safe disposal of high level waste (HLW) and the uptake and separation of long-lived actinides radionuclides and fission products from nuclear waste solutions. Strontium-90, a β -emitter with 0.546 MeV energy, and a half-life of 28.6 years, is one of the most important fission products present in HLW. The content of strontium is roughly 10 times higher than that of cesium, and, as a heat-generator, it causes problems during depositing and vitrifying of HLW (1). Moreover, strontium behaves much like as calcium and deposits in plants by uptake from the soil; food recycle results in ^{90}Sr accumulation in the animal bones and teeth and damaging blood-producing cells (2). So, it is considered to be one of the most hazardous fission

products in the environmental protection field. Therefore, the separation of long-lived ^{90}Sr from HLW prior final disposal is important not only to reduce the volume of disposal waste and to simplify HLW vitrification, but also from the viewpoint of environment protection, to reduce the damage of all biological components.

Currently, simple and rapid methods for separation and determination of trace levels of strontium have become a focus in the field of radionuclide research. A number of methods have been reported, such as typical liquid/liquid extraction process (3,4), solid phase extraction (5,6), and other partitioning methods (7–9). Biosorption of heavy metal ions from aqueous solutions based on the use of biomass as an absorbent is considered to be a very promising process. This procedure has been successfully used for the removal of different types of organic and inorganic xenobiotics and metal ions, including strontium ones (2).

Recently magnetic separation techniques, which are usually based on the application of magnetically responsive composite materials have already found many important applications in various areas of biosciences, medicine, biotechnology, environmental technology, etc. (10,11). In order to simplify the separation processes, magnetically modified microbial biomass has been developed as an adsorbent and used for the separation of different kinds of compounds, such as heavy metal ions (12) or water soluble dyes (13).

Magnetically modified *Kluyveromyces fragilis* cells (fodder yeast) have been used recently as an efficient biosorbent for the removal of several organic dyes (14). This material is inexpensive and can be prepared in a simple way. Whole yeast cells and isolated cell walls are known to adsorb different metal ions efficiently (15,16). In this work magnetic fodder yeast cells were used as an adsorbent for the removal of Sr^{2+} ions from aqueous solutions.

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MATERIALS AND METHODS

Materials

Magnetically modified *Kluyveromyces fragilis* cells were prepared as described recently (14). Strontium nitrate standard (GBW(E) 080242) was obtained from National Research Center for Certified Reference Materials, Beijing, China. Common chemicals were provided by Alfa-Aesar, or Beijing Chemicals, China. Ultrapure water was obtained by Milli-Q System ($18.2 \text{ M}\Omega \cdot \text{cm}^{-1}$).

Adsorption of Sr^{2+} on Magnetically Modified *Kluyveromyces fragilis* Cells

Magnetically modified *Kluyveromyces fragilis* cells were suspended in ultrapure water (final concentration $13 \text{ mg (dry weight) mL}^{-1}$). Portions ($0.05 \sim 4.9 \text{ mL}$) of strontium nitrate solution (100 mg L^{-1}) and appropriate volumes of ultrapure water were mixed in 10 mL test tubes, then 0.1 mL of the yeast suspension was added under mixing and the total volume of the suspension was made up to 5.0 mL . The suspensions were mixed at appropriate temperature for 1 h . Subsequently, magnetic cells were separated from the suspension using a permanent magnet and concentration of Sr^{2+} in the clear supernatant was measured using the Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Element2, Finnigan Ltd., Germany). The concentration of Sr^{2+} was determined from the calibration curve. The amount of Sr^{2+} adsorbed on a unit mass of adsorbent was calculated using the following formula (1):

$$Q_{eq} = \frac{(C_i - C_{eq}) \cdot V_1}{V_2 \cdot \alpha} \quad (\text{mg g}^{-1}) \quad (1)$$

where Q_{eq} is the absorbed amount of Sr^{2+} after reaching equilibrium, C_i and C_{eq} are the initial and equilibrium Sr^{2+} concentrations in the aqueous phase, V_1 is the total volume of the suspension (5 mL), V_2 is the volume of the added magnetic yeast suspension (0.1 mL), and α is the dry mass of the adsorbent in 1 mL of suspension (g mL^{-1}); α was 0.013 during the experiments.

Desorption of Strontium Ions

Desorption of bound Sr^{2+} from magnetic *Kluyveromyces fragilis* cells was performed in $0.1 \text{ mol L}^{-1} \text{ HNO}_3$ solution under mixing at room temperature for 1 h . Then the suspension was separated using a permanent magnet and the concentration of Sr^{2+} was determined in the clear supernatant using ICP-MS. To study the reusability of the biosorbent, the material after the desorption step was extensively washed with water and used again.

Effect of pH Value and Temperature

The dependence of magnetic *Kluyveromyces fragilis* cells biosorption capacity on the pH value of strontium nitrate

solution (ranging from pH 1.0 to 7.0, determined with the pH meter PHS-3C, Shanghai REX Instrument Factory, China) was investigated at the Sr^{2+} concentration 10 mg L^{-1} and the incubation time 1 h at room temperature. Temperature dependence of biosorption was examined in the range $4 \sim 30^\circ \text{C}$ at pH 7.0, incubation time 1 h and Sr^{2+} concentration range 1 mg L^{-1} to 40 mg L^{-1} .

RESULTS AND DISCUSSION

Effect of pH Value

The data in Fig. 1 indicate the effect of pH on the biosorption of Sr^{2+} . As can be seen, the biosorption capacity of magnetic yeasts for Sr^{2+} increased with increasing pH. The biosorption value reached almost a plateau between pH 4.0 and 7.0, indicating that increased pH of the solution favors complexes formation between the ionized anionic groups on the yeast cells and Sr^{2+} . Lower affinity for Sr^{2+} observed in low pH values ($\text{pH} < 2.0$) can be explained by the competition of protons and Sr^{2+} for binding sites on the cell wall (17,18). Using $10 \text{ mg L}^{-1} \text{ Sr}^{2+}$ initial concentration and 20 min incubation time, the adsorption capacity of the magnetic biosorbent was 19.5 mg g^{-1} under optimal conditions ($\text{pH} > 4$).

Effect of Temperature

Temperature can influence adsorption of target compounds both in the positive and negative way. Figure 2 shows the influence of different temperature on the biosorption capacity of magnetic *Kluyveromyces fragilis* cells for Sr^{2+} . As can be seen, an increase of the temperature (examined in the range $4 \sim 30^\circ \text{C}$) caused gradual increase of adsorbent biosorption capacity; however, only a minor influence of temperature was observed and that is why

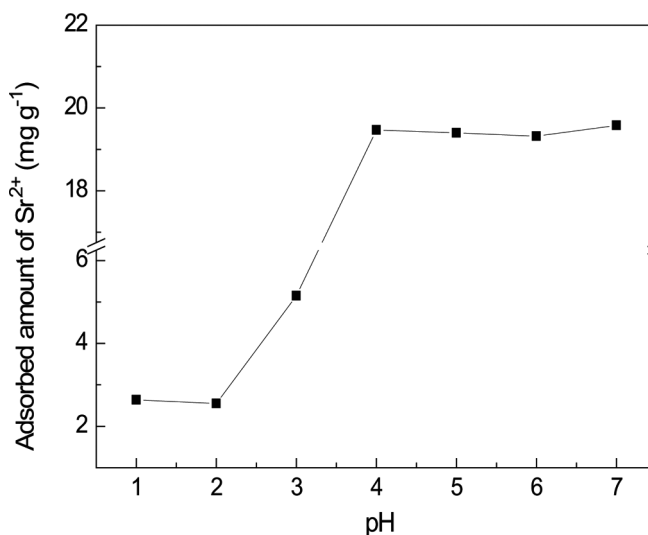


FIG. 1. Effect of pH on biosorption of Sr^{2+} on magnetically modified yeast cells (Sr^{2+} concentration: 10 mg L^{-1} ; temperature: 20°C ; incubation time: 1 h).

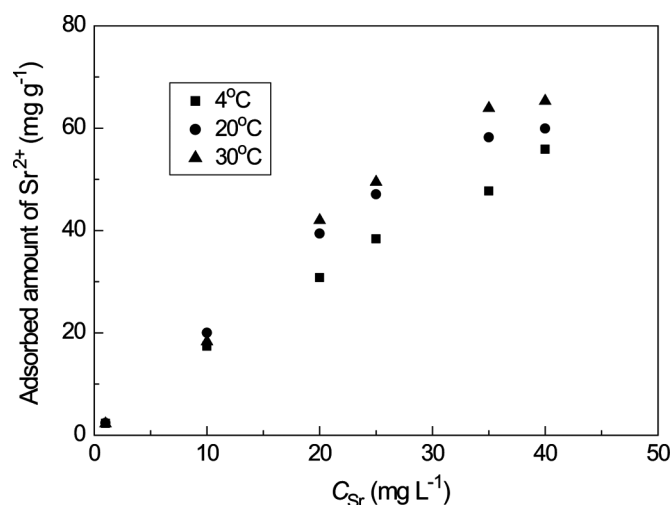


FIG. 2. Effect of temperature on Sr^{2+} biosorption on magnetically modified yeast cells (pH: 7.0; incubation time: 1 h).

the subsequent experiments were performed at room temperature (20°C).

Time Dependence of Sr^{2+} Biosorption

The kinetics of biosorption of Sr^{2+} by the magnetic *Kluyveromyces fragilis* cells from neutral solutions was investigated when the concentrations of Sr^{2+} were 10 mg L⁻¹ and 40 mg L⁻¹ respectively. The results are shown in Fig. 3. As can be seen, the amount of Sr^{2+} adsorbed on magnetic yeast cells quickly increased during the prolonged incubation time. Though the adsorption process reached equilibrium within 10~20 min, an incubation time of 1 h was used for all subsequent experiments.

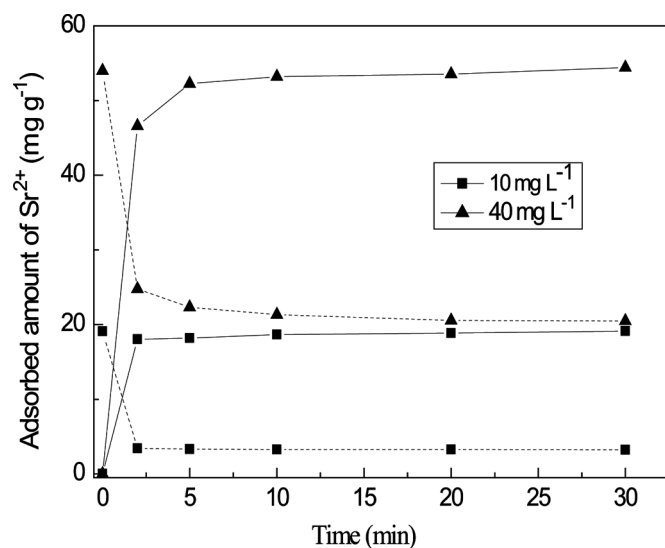


FIG. 3. Sr^{2+} adsorption and desorption as a function of time, using magnetic yeast cells as adsorbent. — adsorption curve; desorption curve.

For 20 min incubation time, the biosorption capacity of magnetic yeast cells was 19.5 mg g⁻¹ for 10 mg L⁻¹ Sr^{2+} solution and 53.5 mg g⁻¹ for 40 mg L⁻¹ Sr^{2+} solution.

Sr^{2+} Desorption from Magnetic Yeast Cells

At the same time, desorption of bound Sr^{2+} with 0.1 mol L⁻¹ HNO₃ was studied. The desorption ratio was calculated from the amount of Sr^{2+} absorbed on the magnetic yeast cells and the final concentration of Sr^{2+} in the desorption solution. The courses of desorption, after charging the biosorbent with Sr^{2+} from 10 mg L⁻¹ and 40 mg L⁻¹ solutions are shown in Fig. 3. As can be seen, the desorption process was fast and reached the equilibrium within 10~20 min. After desorption, the amount of Sr^{2+} still adsorbed accounted for 16.7% and 36.6% of the biosorption capacity of the adsorbent for Sr^{2+} ions after loading with 10 mg L⁻¹ and 40 mg L⁻¹ solutions, respectively. These results confirm the influence of pH value on the adsorption process, i.e., that the Sr^{2+} binding groups on the yeast cells exhibit lower affinity in lower pH values conditions.

Repeated adsorption/desorption cycles were performed to examine the reusability and metal recovery efficiency of the biosorbent. The Sr^{2+} adsorption capacity decreased from 19.5 to about 13 mg g⁻¹ at the third adsorption step after repeated loading with 10 mg L⁻¹ solution and 20 min incubation time.

Effect of Sr^{2+} Concentrations

The effect of Sr^{2+} initial concentrations (ranging from 1 mg L⁻¹ to 100 mg L⁻¹) on biosorption capacity of the magnetic *Kluyveromyces fragilis* cells was studied at pH 7 and contact time of 1 h, respectively (Fig. 4). The amount of Sr^{2+} absorbed on the magnetically modified yeast cells

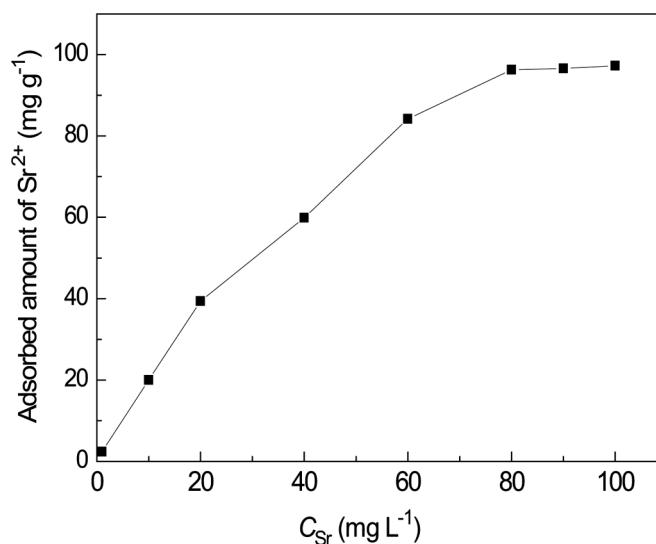


FIG. 4. Dependence of Sr^{2+} adsorption on magnetic yeast cells on Sr^{2+} initial concentration (pH: 7.0; temperature: 20°C; incubation time: 1 h).

quickly increased with the increase of the Sr^{2+} concentration. When the Sr^{2+} concentration increased to 80 mg L^{-1} and more, the biosorption capacity remained constant, indicating saturation of the binding sites on the surface of the yeast cells. The cell walls of *Kluyveromyces fragilis* cells are composed from several types of biopolymers, such as glucan (28%), mannan (31%), proteins (13%), lipids (8%), chitin and chitosan (2%) (18,19). These compounds provide different types of groups (including metal-binding groups) such as carboxy-, hydroxyl-, amino-, sulfhydryl-, sulfate-, phosphate-, etc. Sr^{2+} binding can be attributed to ion exchange, adsorption, complexation, microprecipitation, and crystallization processes occurring on the cell wall. According to the previous studies, Sr^{2+} ions form stable bonds especially with nitrogen- or oxygen-ligands (20). The maximum biosorption capacity was 97.2 mg g^{-1} for the magnetically modified yeast cells at the highest initial Sr^{2+} concentration used (100 mg L^{-1}). From the viewpoint of the adsorbed amount, the magnetically modified *Kluyveromyces fragilis* cells may be considered as a superior biosorbent for Sr^{2+} and be appropriate to use in reprocessing processes to eliminate Sr^{2+} .

Sorption Isotherms

To develop a possible large-scale “magnetic biomass” separation process, it is necessary to determine the biosorption capacity of the magnetic biocomposite. Previous literature has reported many isotherm models to calculate the maximum adsorption capacity of adsorbents. However, the most commonly used models are the Langmuir and Freundlich ones.

In the present study, Langmuir and Freundlich models have been used. A mathematical description of the Langmuir isotherm is expressed as (2):

$$Q_{\text{eq}} = bQ_{\text{max}}C_{\text{eq}}/(1 + bC_{\text{eq}}) \quad (2)$$

where Q_{eq} (expressed in mg g^{-1}) is the amount of the adsorbed Sr^{2+} ions per unit mass of magnetically modified cells at equilibrium and C_{eq} (expressed in mg L^{-1}) is the equilibrium concentration of the unbound (free) metal ions. Q_{max} is the maximum adsorption capacity (mg g^{-1}) and b is a constant related to the enthalpy of adsorption (expressed in L mg^{-1}). The value of Q_{max} represents a practical limiting adsorption capacity when the adsorbent surface is fully covered with metal ions.

The Freundlich isotherm describes a more complicated adsorption model where interactions occur between adsorbed molecules. The Freundlich isotherm is expressed as (3):

$$Q_{\text{eq}} = kC_{\text{eq}}^{1/n} \quad (3)$$

where the constants k and $1/n$ are characteristic constants, k being related to the adsorption capacity. Constant $1/n$

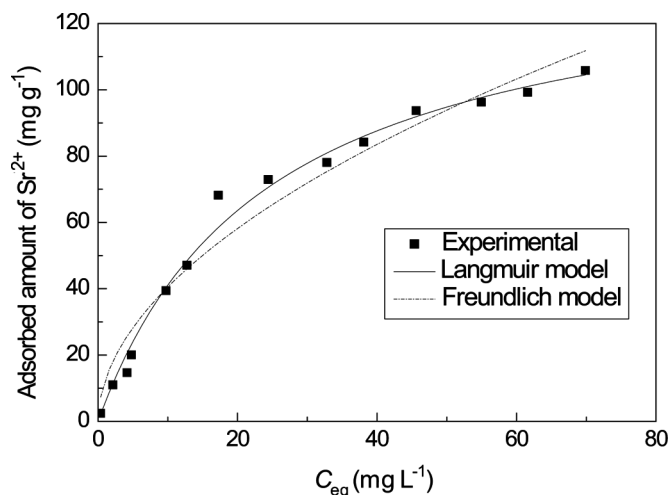


FIG. 5. Equilibrium adsorption isotherms of Sr^{2+} using magnetic yeast cells as adsorbent (pH: 7.0; temperature: 20°C ; incubation time: 1 h).

represents the intensity of the reaction, with a value less than one indicating favorable adsorption.

The sorption isotherms for Sr^{2+} binding on magnetic *Kluyveromyces fragilis* cells in neutral medium are given in Fig. 5. The biosorption capacity for Sr^{2+} obtained from the experimental data was 97.2 mg g^{-1} yeast cells using 100 mg L^{-1} Sr^{2+} initial concentration. The isotherm model parameters were obtained by non-linear least square fitting of the experimental data using Microcal Origin 6.0 program (Microcal Software Inc., 2000). The values of constants or binding parameters obtained by fitting Langmuir and Freundlich models to the experimental adsorption data obtained during adsorption of Sr^{2+} on the magnetic *Kluyveromyces fragilis* cells, as well as the values of correlation coefficients are shown in Table 1. The Langmuir model fits well the experimental isotherm data and suggests that monolayer adsorption is the prevalent mechanism. The Langmuir model predicts a maximum biosorption capacity of 140.8 mg g^{-1} (dry weight) for Sr^{2+} binding on magnetic *Kluyveromyces fragilis* cells. This value is high, and is fully comparable or better when

TABLE 1
Calculated constants and correlation coefficients obtained by fitting Langmuir and Freundlich models to the experimental data

Langmuir	Freundlich
$b = 0.041$	$k = 12.16$
$Q_{\text{max}} = 140.8 \text{ mg/g}$	$1/n = 0.52$
$R^2 = 0.9901$	$R^2 = 0.9302$

TABLE 2

Comparison of maximum adsorption capacities Q_{\max} of studied magnetic yeast cells with other materials

Adsorbent	Q_{\max} [mg g ⁻¹]	Reference
Magnetic <i>Kluyveromyces fragilis</i> cells	140.8	This paper
<i>Amaranthus spinosus</i> root tissue powder	12.89	(21)
	23.85 (NaOH treatment)	
Activated carbon	44.4	(22)
Mucilaginous seeds of <i>Ocimum basilicum</i>	247	(2)
Carbon material from pecan shells	180	(23)
<i>Shewanella alga</i>	6.9	(24)
<i>Shewanella putrefaciens</i>	6.6	(24)
Pakistani coal powder	140	(25)
Zeolite synthesized from fly ash	85.5	(26)

compared with maximum adsorption capacities for other adsorbents described recently (see Table 2).

CONCLUSIONS

Biosorption has become an important approach to remove heavy metals and radionuclides ions from aqueous solutions. In this paper magnetically modified yeast cells were used to study the biosorption and desorption of Sr²⁺ from solutions. The results demonstrated that the adsorbed amount of Sr²⁺ increased with the enhanced Sr²⁺ concentration, reaching the plateau at around 80 mg L⁻¹ Sr²⁺ initial concentration at 20°C and pH = 7. The biosorption capacity was affected by the pH value and constant adsorption was found at the pH range from 4 to 7. The biosorption process was fast and the equilibrium was reached in approximately 20 min. Desorption was carried out by using 0.1 mol L⁻¹ HNO₃ and approximately 80% of adsorbed Sr²⁺ was released. In addition, magnetic modification of the yeast cells enabled their simple removal from the suspensions using a permanent magnet. This biosorbent exhibits superparamagnetic behavior at room temperature so the particles show magnetic properties when placed within a magnetic field, but retained no residual magnetism when removed from the field (14). From these results, we can draw a conclusion that magnetically modified yeast cells represent a promising adsorbent which could be used to concentrate and separate Sr²⁺ ions from liquid high level waste originating during spent fuels reprocessing.

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