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Biosorption behavior of Sr^{2+} using straw-derived biochar: equilibrium and isotherm study

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ABSTRACT

Rice straw-derived biochars carbonize at different temperature (550, 650, 750°C) used for Sr^{2+} removal. Experimental parameters affecting the sorption process like pH, contact time, sorbent concentration, and temperature were investigated. Sorption profile showed an initial rapid uptake which decreased and became almost constant after one hour. Optimum Sr^{2+} sorption was achieved at pH 6. Sorption kinetics good correlated with both pseudo-second-order and Elovich kinetic equations. Isotherm model of Dubinin–Radushkevich provides the best correlation of Sr^{2+} sorption. The present biochar shows better sorption performance (198 mg/g) of Sr^{2+} than many types of sorbents. Adsorption of Sr^{2+} on biochar was endothermic and increase upon solution temperature rises from 25 to 55°C. Present investigation emphasized that the rice straw-based biochar exhibited a high potential for Sr^{2+} removal from aqueous solution.

Keywords: Biochars; Sorption equilibrium; Sr^{2+}; Kinetics; Rice straw

1. Introduction

Biomass is a renewable energy resource and has a growing interest as a chemical feedstock source. The pyrolysis conversion of biomass into value-added products, namely solid char (biochar), liquid (bio-oil), and gas (biogas) has attracted tremendous research interest, mainly due to the rising energy demands and concerns over greenhouse gas emissions [1]. A great number of applications have been considered for liquid product (bio-oil), but the char has not received much attention.

Biochar is a newly constructed scientific term. According to Lehmann and Joseph, it is defined as "a carbon rich product when biomass such as wood, manure or leaves is heated in a closed container with little or unavailable air"[2]. Shackley et al. defined biochar more descriptively as "the porous carbonaceous solid produced by the thermochemical conversion of organic materials in an oxygen depleted atmosphere that has physicochemical properties suitable for safe and long-term storage of carbon in the environment" [3]. Indeed, various types of biomass including woody materials, animal wastes, crop residues, organic wastes, and sewage sludge have been used to produce valuable biochars via slow to intermediate pyrolysis processes [4]. Agricultural wastes or residues are wide available low-cost raw material to produce biochar, as well as bio-oil and gases [5].

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Rice straw is one of the main kinds of agricultural by-products. Large quantities of straws accumulate due to agricultural practices in Egypt. Although some residues are used as feed, fuel, or straw returned to the fields, millions of tons of rice straws are burnt annually. This represents a main source of air pollution. Carbonization of straw wastes to produce biochar, charcoal like product, has been suggested to avoid negative impacts of direct burning. As the biochar is resistant to biological decay, it is preserved in the terrestrial systems for much longer time than for example plant residues or compost; therefore, the beneficial effects are extended [6]. Therefore, carbonization through pyrolysis to produce biochar is a successful mean to avoid the negative impacts on human health and environment.

Now, there are many physicochemical processes for Sr^{2+} removal from waste streams such as chemical precipitation, flocculation, ion exchange, membrane process, immobilization method, and adsorption [7]. Among these, adsorption represents one of the most operative and cost-effective methods. Adsorption processes have been widely used in water and wastewater related industries. Researchers have investigated several sorbents to remove Sr^{2+} from aqueous solutions, such as multiwall carbon nanotube, minerals, silica materials, gel, oxides, and biomass [8]. Biosorption using carbon-based materials including biomass or waste bioproducts is considered as a promising biotechnology for metal ion removal from water, due to its simplicity, efficiency, low cost, and availability. Furthermore, these materials widely used as adsorbents for radioactive material due to its chemical, thermal, radiation stability and rigid porous structure and mechanical strength [9]. In a previous study, several carbon-based materials were produced and used for the selective removal of uranium [10–12], thorium [13], cesium, erbium [14], and Sr^{2+} [15,16].

This study aims to investigate Sr^{2+} sorption behavior on rice straw-based biochar as a function of experimental parameters such as solution pH, initial sorbent dosage, and contact time. Different mathematical models were used to study sorption isotherms and kinetics of the process. The calculated model parameters are useful in case of further scale up of the process.

2. Experimental

Reagent-grade chemicals were used in this study. Stock solution (1,000 mg/L) of Sr^{2+} chloride (purchased from Merck Company) was prepared by dissolving accurate amount in deionized water. Working solutions of the needed concentrations were prepared by continuous dilutions (5–100 mg/L). All sample bottles and glassware were cleaned, rinsed with deionized water, and oven-dried at 60°C.

Preparation of biochar was done using fluidized bed reactor. 500 gm of dried rice straw heated at 50°C/10 min under flow 300 mL/min of nitrogen. Heating continued up to 550, 650, and 750°C with one hour hold. After cooling, biochars were washed with distilled water, and dried at 120°C. The dried sample was crushed and sieved to obtain a particle size range of 0.5–1.0 mm, and stored in plastic bottles for later use. The obtained rice straw biochars at the desired temperatures were labeled as RS-550, RS-650, and RS-550, respectively.

Biochars surface area (BET surface area) was measured by nitrogen adsorption method after degassing for 1 h at 105°C using Quantachrome NOVA 1100e. pH of biochars was measured by mixing 2 g of biochar in 50 mL distilled water at 90°C for 20 min [17]. After cooling, the pH was measured with a Corning pH 10 portable pH meter (Acton, MA, USA). pH_{pzc} (point of zero charge) was determined by mass titration method described by Leon et al. [18]. Surface acidity and basicity of biochars were determined by Boehm method [19].

Batch adsorption experiments were conducted on orbital shaker (thermo scientific) at a shaking speed of 150 rpm, using 25 mL Erlenmeyer flasks with addition of specific amount of adsorbent to 10 mL of Sr^{2+} solution of given initial concentration. Kinetic experiments were performed in the condition of 25°C, 20 mg of the biochar, Sr^{2+} solution of 30 mg/L concentration, and pH (6.5) without adjusting. The flasks were shacked at room temperature. At different time intervals (10–120 min), sorbent was removed using filtration and Sr^{2+} concentration was measured using an atomic absorption (Varian, spectra-110-220/880 Australia Pty. Ltd.) equipped with nitrous oxide-acetylene burner at the wavelength of 460.73 nm. A calibration curve is constructed by running several samples of known Sr^{2+} concentration under the same conditions as the unknown. The amount the standard absorbs is compared with the calibration curve and this enables the calculation of the Sr^{2+} concentration in the unknown sample.

Influence of solution pH on the sorption was performed by mixing 20 mg of biochar into 10 mL Sr^{2+} solution of 30 mg/L in the condition of 25°C and different initial pH of Sr^{2+} solutions. The initial pH of Sr^{2+} solution was adjusted to values in the range of 2–10 by drop wise adding 0.1 N NaOH or HCl solutions. After 60 min of contact time, the concentrations of Sr^{2+} left in supernatant solutions were determined to study the effect of pH.

To study the adsorption isotherms, 20 mg of biochar was added into 10 mL of Sr^{2+} solutions of different initial concentrations (5–100 mg/L) in 25 mL flasks at 25˚C and pH 6.0 until the system reached adsorption equilibrium. The concentrations of Sr^{2+} left in the supernatant solutions were determined to calculate q at equilibrium.

The effect of temperature was investigated by carrying out batch adsorption study at different temperatures (25 and 55˚C) at pH (6.0) while other conditions were the same as the kinetic experiments. The temperature of Sr^{2+} solution was adjusted using a thermostatic water bath (Memmert WB29 Model).

Effects of sorbent dosages were conducted at different biochar concentrations (0.5–10 g/L) at fixed Sr^{2+} concentrations (30 ppm) and pH 6.0. Other conditions were the same as in pH effect experiments.

The Sr^{2+} uptake q_e (mg/g) was calculated from the mass balance as follows:

$$q_e = \frac{V(C_i - C_e)}{m} \tag{1}$$

Metal ion removal efficiency ($E\%$) was calculated as follows:

$$E(\%) = \frac{C_i - C_e}{C_i} \times 100 \tag{2}$$

where C_i and C_e are the initial and equilibrium concentrations of Sr^{2+} (mg/L) in aqueous solution, respectively, m is the weight of the sorbent (mg), and V represents the volume of the solution (mL).

3. Results and discussion

3.1. Biochar characterization

According to physicochemical properties of prepared biochars given in Table 1, surface area, S_{BET}, and total pore volume, V_p increases with increase pyrolysis temperature from 550 to 750˚C. Values of pH and pH_{pzc} are comparable which indicates no inorganic oxides constituents leach from these biochars. This is a good property in water treatment. Acidity and basicity results indicate amphoteric behavior of all biochars due to coexist of acidic and basic surface sites in all samples.

Table 1
Biochars characteristics

Biochar	RS-550	RS-650	RS-750
S_{BET} (m^2/g)	71.35	76.2	63.0
V_p (cc/g)	0.055	0.063	0.052
Basicity (meq/g)	6.22	8.13	3.06
Acidity (meq/g)	5.6	2.0	1.79
Sr^{2+} removal q_e (mg/g)	56.0	15.6	5.56
pH_{pzc}	9.5	9.3	9.4
pH	9.2	9.3	9.07

RS-550 biochar was found to be superior for Sr^{2+} removal, q (56.0 mg/g) in comparison to other biochars (15.6 and 5.56 mg/g). No particular feature of RS-550 biochar appears to be dominant in the removal of Sr^{2+}. Therefore, RS-550 biochar was used for further experiments to find out the influence of different experimental parameters on Sr^{2+} uptake.

3.2. Sorption profile

Fig. 1 shows the sorption profile of Sr^{2+} uptake as function with time at different temperature. Removal of Sr^{2+} rises with contact time and reaches equilibrium at 60 min. This short equilibrium time is one of the significant concerns for economical applications in wastewater treatment. The curves in Fig. 1 are smooth, single, and continuous until saturation, which represent monolayer coverage of Sr^{2+} on sorbent surface. There are two sections of the uptake profile in Fig. 1 which are (i) initial fast Sr^{2+} uptake between 0 and 30 min and (ii) more gradual removal up to 60 min of contact after which no further uptake of Sr^{2+} was observed. The 1st stage is owing to the accessibility of great number of available sorption sites for Sr^{2+} ion on adsorbent surface. The subsequent slower stage is due to competition among Sr^{2+} for limited number of remaining vacant sorption sites. Thus, the driving

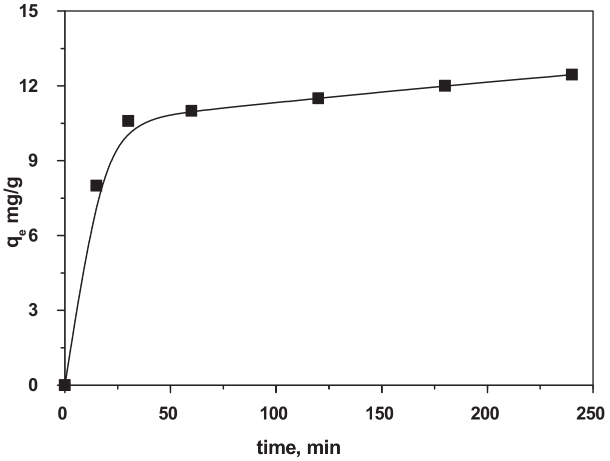

Fig. 1. Effect of contact time on Sr^{2+} sorption by RS-550 biochar.

concentration gradient between the bulk solution and the solid surface is the main factor controlling the kinetics of the system. A similar trend of rapid initial and subsequent slower sorption was reported by many researchers [20–22].

3.3. Effect of pH

pH is important factor for metal ions adsorption. It affects the metal ions solution chemistry and sorbent functional groups. Table 2 shows a gradual increase in Sr^{2+} adsorption capacity with increase in pH from 2 to 6 and remained constant after that. To explain the pH effect, you should take in consideration of biochar surface charge and interaction nature of the metal ion species in solution. Biochar derived from rice straw contains silica (SiO_2) and its hydrolysis produces silanol group –SiOH. Sr^{2+} removal may be takes place through ion exchange substitution reaction of Sr^{2+} ion and protons of the surface carboxyl and silanol groups. This can be represent by following reactions: [23]

$$\mathrm{Sr^{2+} + m(-COOH) \leftrightarrow Sr(OOC-)_m^{m-2} + mH^+} \tag{3}$$

$$\mathrm{Sr^{2+} + m(-SiOH) \leftrightarrow Sr(OSi-)_m^{m-2} + mH^+} \tag{4}$$

where −COOH = carboxyl group, −SiOH = silanol group, and mH^+ = number of protons released. At low pH, Eqs. (3) and (4) lie to the left due to high H^+ concentration that competes with Sr^{2+} for binding onto the biochar surface. Together, the main charges on biochar are positive which results in the lower uptake of positively charged Sr^{2+} on RS-550 biochar. By increased pH, Eqs. (3) and (4) precede to the right, ion exchange sites deprotonate, and negative charge on the biochar increases. As a result, active sorption sites available for Sr^{2+} increase and electrostatic attraction between the negatively charged biochar surface and the positively charged Sr^{2+} will occur. This means that Sr^{2+} ion removal is increased. Within this pH range, the ion exchange process is the major mechanism for removal of metal ion from solution. Further, a decrease in the solution pH compared to the initial pH was observed after equilibration. This suggests that H^+ ions liberated from the solid surface into the aqueous phase as a result of the exchange with Sr^{2+} cations. Similar trends were also observed for Sr^{2+} removal by gels-based polysaccharide derivatives and magnetically modified fodder yeast (*Kluyveromyces fragilis*) cells [24,25] and Coir pith [26]. In the following studies, experiments were done at pH 6.0 to avoid any possible hydroxide precipitation.

Table 2
Effect of solution pH on Sr^{2+} uptake by RS-550 biochar

pH	q_e (mg/g)
2	2.0
4	9.8
6	14.0
8	15.2
10	15.5

3.4. Effect of sorbent dosage

Table 3 shows that increasing the biochar concentration increase the percent removal (*R*%) but decrease the adsorption capacity (q_e) with a saturation level at high doses (6 g/L). This is may be due to resistance to mass transfer of Sr^{2+} from bulk liquid to sorbent solid surface becomes important at high sorbent loading. Moreover, in the same time, sorption capacity was decreased with increase of dosage. Higher sorbent dose causes particles aggregates and interference or repulsive forces between binding sites, therefore, decrease the interaction of Sr^{2+} with the sorbent and reduces the total surface area of the sorbent.

3.5. Sorption kinetics

To understand the sorption mechanism of RS-550 biochar for Sr^{2+}, the sorption kinetics were studied using pseudo-second-order [27] and Elovich kinetic models [28] by Eqs. (5) and (6), respectively.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{5}$$

$$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t \tag{6}$$

Table 3
Sorption capacity, q_e, and efficiency, *E*% of Sr^{2+} at different sorbent masses of RS-550 biochar

Mass (g/L)	*E*(%)	q_e (mg/g)
0.5	52	50
1	79	38.8
2	88	21.6
3	96	15.7
6	99	8.1
10	99.8	4.9

where q_e and q_t are amounts of ion adsorbed at equilibrium (mg/g) and time t (min), respectively. K_2 is the adsorption rate constants of pseudo-second-order (g/(mg min). $h = k_2 q_e^2$, where h is the initial adsorption rate (mg/g min). The parameters α and β in Elovich equation are the initial rate (mg/g min) and extent of surface coverage and activation energy for chemisorptions (g/mg), respectively. α (<1) in Bangham equation is constant.

The calculated value of pseudo-second-order and Elovich equation constants is given in Table 4. The pseudo-second-order model was found to explain the adsorption kinetics of Sr^{2+} onto RS-550 biochar most effectively with correlation coefficients 0.999. Furthermore, estimated q_e values of pseudo-second-order model accurately predict the adsorption kinetics over the entire working times. This result indicates that the pseudo-second-order model is more suitable than Elovich model for Sr^{2+} adsorption onto RS-550 biochar and that the adsorption complies with the pseudo-second-order reaction. Thus, supporting the basic assumption in the model that chemisorption plays a major role in this adsorption system [29] and rate of reaction is directly proportional to the number of active sites on the surface of adsorbent. According to pseudo-second-order model, the adsorption rate $\mathrm{d}q_t/\mathrm{d}t$ is proportional to the second order of $(q_e - q_t)$. Since RS-550 biochar have relatively high equilibrium adsorption density q_e, the adsorption rates become very fast and the equilibrium times are short. Such short equilibrium times coupled with high adsorption capacity indicate high degree of affinity between Sr^{2+} and RS-550 biochar surface [30].

On the other hand, from R^2 values in Table 4, Elovich model can be accepted as one of characteristics of Sr^{2+} sorption on RS-550 biochar. Elovich equation was also used successfully to describe second-order-kinetic. Elovich equation assuming that chemisorption with highly heterogeneous materials, but does not propose any definite mechanism for adsorbate–adsorbent interaction [31]. Many investigators found that initial rapid adsorption followed by a slow chemisorption is well expressed by Elovich equation, in particular, when the adsorption process follows the pseudo-second-order. According to each studied system, the phenomenon was differently explained, by the presence of sites with different energies on the adsorbent surface, or by the modification of the nature of the bonding between adsorbate and adsorbent.

Table 4
Kinetic parameters of Sr^{2+} adsorption onto RS-550 biochar

Model	Parameters	Value
q_e (exp) (mg/g)		13.0
Pseudo-second-order equation		
	q_e (mg/g)	13.0
	k_2 (g/mg min)	0.01
	h (mg/g min)	1.2
	R^2	0.99
The Elovich equation		
	α (mg/g min)	4.4
	β (g/mg)	0.84
	R^2	0.94

3.6. Effect of temperature

The time dependent uptake of Sr^{2+} adsorption by RS-550 biochar for the Sr^{2+} concentrations of 30 mg/L at different temperatures (25 and 55˚C) at pH 6 is depicted in Fig. 2. As shown, the equilibrium sorption capacity of Sr^{2+}onto RS-550 biochar was favored at higher temperatures and increased with increasing temperature (endothermic adsorption).

On fundamental grounds, an increase of adsorption with temperature indicates an activated chemisorption process, since physisorption is favored at low temperature being basically exothermic processes. In chemisorption, an increase in temperature increases adsorption due to overcoming of the energetic barrier. This means that Sr^{2+} sorption by rice straw biochars involves not only physical but also chemical sorption. In liquid phase ion-adsorption, the adsorption mechanism is more complicated depending on possible interactions between solid surface, solute, and solvent [32]. Factors involved are surface functional groups, properties of ion, and properties of solution [33]. Some of them are affected by temperature such as,

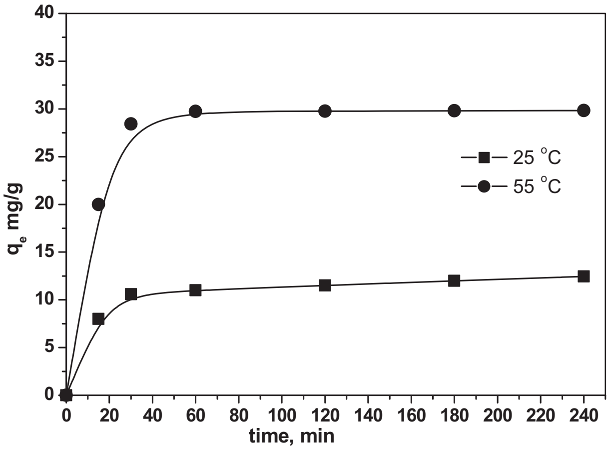

Fig. 2. Effect of contact time at different solution temperatures (25 and 55˚C) on Sr^{2+} sorption by RS-550 biochar.

solubility, ion hydration, hydrophobicity. Their variation can interfere with the effects that temperature can have on adsorption reaction itself that can be chemical or electrostatic [34]. Temperature affects positively some parameters of the system solid-solute-solvent, resulting in an increase in adsorbed amount. For instance, an increase in kinetic energy of adsorbing molecules increases solubility, decreases hydrophobicity, and ion hydration energy. The latter seems to be the factor most affected by temperature. At high temperature, ions are readily dehydrated, and therefore, their sorption becomes more favorable.

3.7. Sorption isotherms

The representation of experimental data in the form of sorption Isotherm is important for understanding of interaction of sorbate with sorbent and also to get a possible insight of the sorption capacity of the sorbent. The sorption data were examined using the Langmuir [35] and Dubinin–Radushkevich (D–R) [36] isotherm models equations.

Langmuir model $$\frac{1}{q_e}=\frac{1}{q_{\max}}+\left(\frac{1}{q_{\max}k_l}\right)\frac{1}{C_e} \quad (7)$$

D–R model $$\ln q_e=\ln q_m-\beta\in^2 \quad (8)$$

where $q_{\max}$ (mg/g) is the sorbent monolayer capacity; k_L(L/mg) is Langmuir constant related to sorption free energy; q_m (mg/g) is the theoretical saturation capacity; and ε is Polanyi potential, $\varepsilon = RT\ \ln\ (1 + (1/C_e))$, where T (K) is the absolute temperature and R (J/mol K) is the gas constant; β (mol^2/kJ^2) is a constant related to the mean free energy of sorption per mole of the sorbate. Langmuir $q_{\max}$ and K_L can be calculated from linear plot of $1/C_e$ vs. $1/q_e \times q_m$ and β of D–R equation can be calculated by plotting ln q_e vs. ε^2. Fig. 3 shows the langmuir and D–R isotherm for Sr^{2+} sorption onto RS-550 biochar. Langmuir and D–R parameters for of Sr^{2+} sorption onto RS-550 biochar being listed in Table 5. Based on the correlation coefficients, the applicability of the isotherms was compared. Higher correlation coefficient (i.e. with values nearly ≈1) showed that D–R model is more suitable for describing the sorption equilibrium of Sr^{2+} onto RS-550 biochar in the studied concentration range. The constant β can gives an indication on mean free energy E (kJ/mol) of adsorbate molecule sorption when it is transferred from solution to the sorbent surface. β can be calculated using the relationship [37]:

$$E=\frac{1}{(2\beta)^{1/2}} \quad (9)$$

The calculated E is 9.8 kJ/mol (Table 5) that corresponds to ion exchange process [38]. This confirms the results obtained from pH effect section.

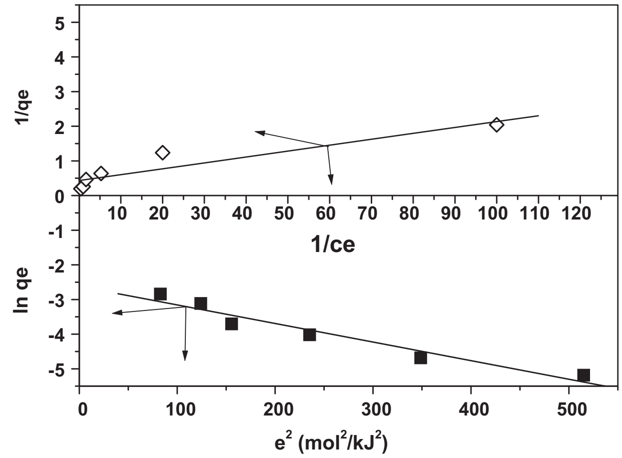

Fig. 3. Langmuir and D–R plot for Sr^{2+} sorption onto RS-550 biochar.

Table 5
Isotherm constants for Sr^{2+} sorption onto RS-550 biochar

Model	Value
Langmuir	
$q_{\max}$ (mg/g)	198
K_L (L/mg)	27.2
R^2	0.94
D–R	
q_m (mg/g)	70.3
β (mol^2/kJ^2)	5.36×10^{-3}
R^2	0.98
E (kJ/mol)	9.84

4. Conclusions

From this study, it can be concluded that biochar prepared from rice straw can effectively remove Sr^{2+} from aqueous solution. Biochar prepared by pyrolysis at 550°C (RS-550 biochar) shows better sorption ability for Sr^{2+} than those prepared at 650 and 750°C. The sorption was found to be strongly dependent on pH, contact time, and sorbent dosage. Optimum sorption of Sr^{2+} on RS-550 biochar occurred at pH of 6. The contact time of 1 h was required to reach the equilibrium. The sorption proceeds according to the pseudo-second-order kinetic model. Ion exchange is probably the major mode of sorption process. The experimental data perfectly fitted the D–R model. The maximum sorption capacity of the biochar for Sr^{2+} was 198 mgg^{-1} in the studied concentration range. Adsorption of Sr^{2+} was on biochar found to be endothermic. The results of the present investigation suggest that rice straw-based biochar can be used as an environmentally benign and low-cost biomaterial for Sr^{2+} removal from aqueous solution.

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