



Effect of adsorbents and chemical treatments on the removal of strontium from aqueous solutions

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ABSTRACT

In the present investigation, three different solid wastes namely almond green hull, eggplant hull, and moss were initially treated and used as adsorbents for the adsorption of strontium ion from aqueous solutions. Adsorbent types and chemical treatments are proved to have effective roles on the adsorption of Sr(II) ion. Among the three adsorbents, almond green hull demonstrated strong affinity toward strontium ion in different solutions. The effectiveness of this new adsorbent was studied in batch adsorption mode under a variety of experimental conditions such as: different chemical treatments, various amounts of adsorbent, and initial metal-ion concentration. The optimum doses of adsorbent for the maximum Sr(II) adsorption were found to be 0.2 and 0.3 g for 45 and 102 mg L⁻¹ solutions, respectively. High Sr(II) adsorption efficiencies were achieved only in the first 3 min of adsorbent's contact time. The kinetics of Sr(II) adsorption on almond green hull was also examined and it was observed that it follows the pseudo second-order behavior. Both Langmuir and Freundlich models well predicted the experimental adsorption isotherm data. The maximum adsorption capacity on almond green hull was found to be 116.3 mg g⁻¹. The present study also confirmed that these low cost agriculture byproducts could be used as efficient adsorbents for the removal of strontium from wastewater streams.

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1. Introduction

The disposal of radioactive wastewater from commercial nuclear plants is one of the major problems in nuclear waste management. Some of the important isotopes in the radioactive wastewaters are strontium, cesium, plutonium, americium and technetium [1]. ⁹⁰Sr is one of the products of nuclear fission in radioactive waste effluents resulting from reprocessing of nuclear fuels. Furthermore, strontium has a variety of commercial uses. It is used in certain optical materials and produces the red flame color of pyrotechnic devices such as fireworks and signal flares.

Due to the long life, high solubility, and bio-toxicity of strontium, separation and recovery of this ion from waste solutions needs special attention. During the last decades, researchers have put numerous efforts to develop suitable processes for better separation of radioactive ions from wastewater. Currently, there are many physico-chemical processes for the removal of these ions from waste streams such as: chemical precipitation and flocculation [2], phosphate precipitation [3], Cu or Ni ferrocyanide precipitation [4], ion exchange [5–7], membrane process [8–10], immobiliza-

tion method [11] and adsorption [12]. Among these, adsorption is one of the most effective and economic methods. Adsorption processes have been widely used in water and wastewater related industries. Removal of heavy metals [13,14] and organic pollutants [15] are applications as such. Moreover, various types of adsorbents were utilized for the adsorption of radionucleic wastes and strontium ions including natural and synthetic zeolites [16], moss as a phytosorbent [17], multiwall carbon nanotubes (MWCNT)/iron oxide magnetic composites [18], activated carbon [19], composite of magnetic particle [20], clinoptilolite [21], and kaolinite [22].

In the present work, we have focused on the removal of strontium ion from aqueous solutions using almond green hull, an agricultural waste, as a new adsorbent. In our previous work, we had successfully employed almond green hull for the removal Co(II) ion from aqueous solutions [23]. Almond trees are abundance in the world and almond hull is an agriculture crop residue that cannot be used by animals and it is usually burnt. The annual production of almond (nut with hard shell) is about 108,000 tons in Iran. Almond green hull is estimated to be about 0.25–0.60 wt% of the whole almond fruit depending on the type. Therefore, approximately 36,000–162,000 tons of this waste material is annually generated in the agriculture section that can be used for wastewater treatment. To the best of our knowledge, this material has not been used before for this kind of application. Utilization of almond green hull not only provide a low cost and easily available adsor-

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Nomenclature

q_e	amount adsorbed (Eqs. (1)–(4) and (6))
C_e	equilibrium concentration (Eqs. (1), (4) and (6))
C_0	initial metal ion concentration (Eqs. (1) and (5))
V	volume of the solution (Eq. (1))
W	mass of adsorbent (Eq. (1))
q_t	amount adsorbed at time t (Eqs. (2) and (3))
$k_{2,ads}$	rate constant of the second-order adsorption (Eqs. (2) and (3))
t	adsorption time (Eqs. (2) and (3))
q_m	Langmuir constant related to the maximum adsorption capacity (Eq. (4))
b	Langmuir constant related to the energy or net enthalpy of adsorption (Eqs. (4) and (5))
K_f	Freundlich constants related to adsorption capacity (Eq. (6))
n	Freundlich constants related to adsorption intensity (Eq. (6))

bent for the removal of heavy metals such as Sr(II), but also it would help the environmental protection.

2. Materials and methods

2.1. Materials

Strontium nitrate was obtained from Merck in analytical grade. Stock solutions were prepared by dissolving strontium nitrate in deionized water. Three adsorbents i.e. almond green hull and eggplant hull, as agricultural residue, and moss were used. The collected precursors were washed with distilled water several times to remove water soluble impurities followed by drying in an electrical oven at 180 °C. Dried samples were ground and then the following two different chemical treatments were carried out to extract soluble organic compounds of the adsorbent and also to enhance chelating efficiency:

- Mixing with the solution of hydrogen peroxide (2%vol) + ammonia (2%vol).
- Mixing with the solution of nitric acid (20%).

2.2. Batch adsorption

All adsorption experiments were carried out using adsorbents with the particle sizes less than 88 μm . Adsorption capacities of the prepared adsorbents (treated and untreated samples) for Sr(II) ion were investigated using aqueous solutions of this metal ion. The adsorbate stock solution was prepared by dissolving $\text{Sr}(\text{NO}_3)_2$ in the distilled water. Then it was diluted to obtain standard solutions containing 45 and 102 mg L^{-1} of Sr(II). Batch adsorption studies were carried out with 0.3 g of adsorbent and 50 mL of Sr(II) solution in several cylindrical flasks. The pH of solutions was kept in neutral medium. Flasks containing adsorbent and adsorbate were agitated on a mechanical stirrer with 720 rpm for predetermined time intervals at specified temperatures. At the end of agitation, the suspensions were filtered using microporous filter paper. The Sr(II) ion concentrations in the solutions were determined by an atomic absorption spectrophotometer (Varian, spectra-110-220/880 Australia Pty. Ltd.) equipped with a Zeeman atomizer.

The uptake of the metal ion in each solution was calculated by the difference of initial and final concentrations. All experiments were conducted triplicate and the results reported here are the average values. The data was used to calculate the equilibrium

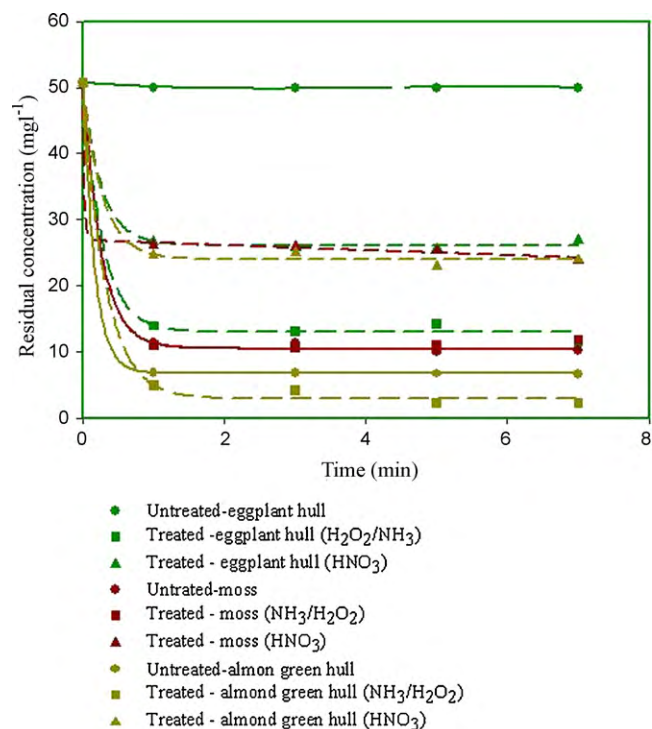


Fig. 1. Effect of chemical treatment on the removal of Sr(II) from aqueous solution (amount of sorbent = 0.3 g; $T = 25^\circ\text{C}$; initial concentration = 51 mg L^{-1}).

metal uptake capacity according to the Eq. (1).

$$q_e = \frac{V(C_0 - C_e)}{W} \quad (1)$$

Where q_e (mg g^{-1}) is the amount of metal adsorption in the adsorbate phase at equilibrium, C_0 and C_e are the initial and equilibrium concentrations of metal ion (mg L^{-1}) in the aqueous solution, V is volume of the solution (L), and W is the mass of adsorbent (g) in the mixture.

3. Results and discussion

3.1. Effect of chemical treatment

Chemical treatment was carried out for extracting soluble organic compounds and enhancing chelating efficiency. This process affects adsorption capability of adsorbent for the removal of strontium ions resulting in some functional groups on the solid surface. The effect of chemical treatment on three different sorbents is shown in Fig. 1. It shows that type of adsorbent is an effective parameter on the removal of Sr(II) ions. As seen in this figure, different adsorbents without any pretreatment have lower adsorption capacities for strontium ions. It is also interesting that for all adsorbents except the untreated-eggplant hull, the amounts of adsorbed Sr(II) are increased rapidly during the first minute of contact period and the process reached to the equilibrium. Namely, after 1 min, removal efficiencies of eggplant hull, moss, and green hull for the untreated, $\text{NH}_3/\text{H}_2\text{O}_2$ treated, and HNO_3 treated samples were 0%, 72%, 47.6%, 78%, 78%, 48.6%, 86.6%, 91%, and 51%, respectively.

Meanwhile, the results suggest that almond green hull treated with hydrogen peroxide (2%vol) and ammonia (2%vol) has higher strontium removal efficiency compared to other adsorbents. This is related to the nature of functional groups created on the solid surface. Generally, chemical treatments attach some functional groups on the adsorbent surfaces that have different affinities toward

Table 1
Surface acidity and basicity of almond green hull treated with H₂O₂ and NH₃.

Phenolic groups (nmol g ⁻¹)	Lactonic groups (nmol g ⁻¹)	Carboxylic groups (mmol g ⁻¹)	Surface basicity (mmol g ⁻¹)
1.26	0.45	0.39	1.165

metal ions in the aqueous solution. Based on these results, in all of the subsequent experiments, we have used this chemically treated almond green hull as adsorbents.

The amounts of surface functional groups on the treated adsorbent were measured by the Boehm's technique. This method can be described as follows: 0.5 g of adsorbent were placed in a series of flasks, each containing 50 mL of 0.05N sodium bicarbonate, sodium carbonate, sodium hydroxide and hydrochloric acid, respectively. Flasks were sealed and shaken for 24 h, after which, the solutions were filtered. Then, 10 mL of each solution was titrated with 0.05N sodium hydroxide and/or hydrochloric acid, depending on the original solution used. The amount of acidic groups on the adsorbent is calculated based on the assumption that NaOH neutralizes carboxylic, lactonic and phenolic groups; Na₂CO₃ neutralizes carboxylic and lactonic groups and NaHCO₃ neutralizes only carboxylic group. The number of surface basic sites is calculated from the amount of HCl reacted with the adsorbent. The reaction between reagents and acidic oxygenated-functional groups on the surface is based on the differences in the acid/base strength. The strength of acidic and basic groups is as follows: carboxylic > lactonic > phenolic.

Results obtained from the Boehm's method applied to treated almond green hull are presented in Table 1. It can be seen that the amounts of acidic and basic groups for this adsorbent are as follows: phenolic > basic > lactonic > carboxylic.

Important properties of solid/liquid systems are influenced by the electric charges on the particle surface. When immersed in an electrolyte solution, a charged particle will be surrounded by ions of opposite sign, so that, from a distance it appears to be electrically neutral. The pH value required to give zero net surface charge [24], designated as the point of zero charge (PZC). Noh and Schwarz [25] proposed an alternative method to the conventional acid/base titration technique. The isoelectric point of the NH₃/H₂O₂ treated sorbent was determined by Noh-Schwarz's mass titration [26,27]. To measure the PZC of the sample, three different initial pH solutions were prepared using HNO₃ and NaOH. The initial pH of 3, 6, or 10.8 was used as the background electrolyte. For each initial pH, six containers were first filled with 100 mL of the solution and then different amounts of samples were added (0.05%, 0.1%, 0.5%, 1%, 5%, and 10% by weight). The equilibrium pH was measured after 24 h. The mass titration curves given for a number of treated sorbents is shown in Fig. 2. This figure illustrates the plot of pH values as a function of mass fraction of the treated sorbent in the solution. As seen in the figure, the asymptotic isoelectric point is about 6. This finding confirms the results of Boehm's tests about the functional groups. According to the Boehm's outcomes, the most functional groups on the surface of the treated sorbent are phenolic and basic groups. Therefore, these functional groups have crucial role for the removal of strontium ions from aqueous solutions.

3.2. Effect of contact time and temperature

Fig. 3 shows the effect of operating time on the adsorption of Sr(II) at different temperatures for a fixed adsorbent (treated-almond green hull) of dose 0.3 g. The initial concentration was kept constant at 45.5 mg L⁻¹. The experimental data indicate that Sr(II) ion adsorption increases rapidly during early contact times and the process achieves almost 90% of its removal efficiency. The short

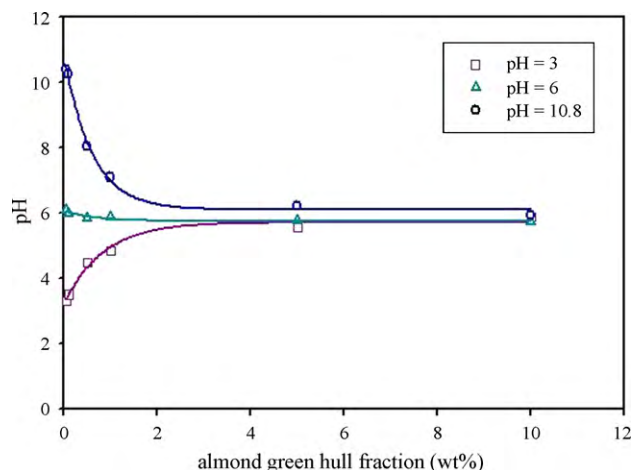


Fig. 2. Mass titration results for almond green hull treated with NH₃/HNO₃.

time needed for adsorption to reach equilibrium state could be attributed to the high adsorption efficiency and readily available adsorbing sites (functional groups) on the adsorbent surface. The results also indicate that there is not a significant difference among the amounts of Sr(II) adsorbed at different temperatures especially after 3 min. This is another reason for the great potential between Sr(II) and almond green hull and independency of the adsorption process to the temperature.

3.3. Effect of initial concentration and adsorbent content

The effect of initial concentration has been investigated using two different concentrations of 45 and 102 mg L⁻¹ at different amounts of sorbent (0.2, 0.3, and 0.4 g) and the results are shown in Fig. 4. It is found that the removal efficiency of Sr(II) was enhanced by increasing the adsorbent/adsorbate ratio, especially at the period of 30 s, but the absolute amount of adsorbed Sr(II) per unit weight of adsorbent declined. Adsorption efficiency and adsorbed amounts of Sr(II) per unit weight of adsorbent at two different initial concentrations and adsorbent dosages of 0.2, 0.3, and 0.4 g at 30 s of elapsed time are presented in Table 2. It is clear that for higher metal concentration, the dose of adsorbent is more critical and it has greater effect on the removal efficiency.

The equilibrium times (about 2 min) are found to be the same for two different concentrations. Similar results are also obtained in

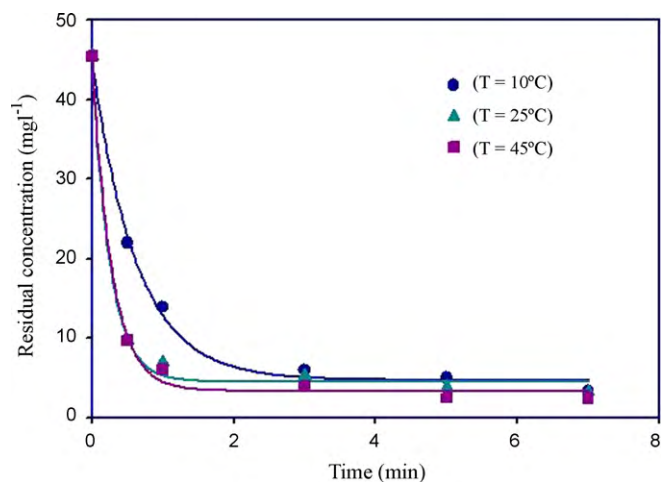


Fig. 3. Effect of contact time on adsorption of Sr(II) at different temperatures (stirring speed = 720 rpm; sorbent amount = 0.3 g; initial concentration = 45.5 mg L⁻¹).

Table 2

Removal efficiency and adsorbed amount of Sr(II) per unit weight of sorbent after 30 s.

Solution initial conc. (mg L ⁻¹)	Sorbent dose (g)	Removal efficiency (%)	Sr(H) adsorbed (mg g ⁻¹)
45	0.2	69.7	7.85
	0.3	78.5	5.89
	0.4	79.7	4.48
102	0.2	45	11.45
	0.3	59.3	10.07
	0.4	67.2	8.56

our previous investigation [23]. The results show that Sr(II) removal is concentration dependent. At higher initial concentrations, the ratio of available adsorption sites to strontium ions is less and the binding sites saturate more rapidly.

3.4. Kinetic study

In the order to investigate the mechanism of adsorption, kinetic models are generally used to test the experimental data. One of the frequently used kinetic models is pseudo-second-order equation. This model is expressed as:

$$\frac{dq}{dt} = k_{2,ads}(q_e - q_t)^2 \tag{2}$$

where $k_{2,ads}$ (g mg⁻¹ min⁻¹) is the rate constant of the second-order adsorption.

$$\frac{t}{q_t} = \frac{1}{k_{2,ads}q_e^2} + \frac{t}{q_e} \tag{3}$$

The equation constants can be determined by plotting t/q_t against t .

The values of different parameters determined from the pseudo-second-order kinetic model along with their corresponding correlation coefficients are presented in Table 3. The correlation coefficients of this model are nearly equal to 1 and the theoretical values of q_e are in good agreement with the experimental data. This finding suggests that the adsorption of strontium ion follows the second-order kinetic model. As reported in Table 3, the strontium adsorption-rate constants ($k_{2,ads}$) increase significantly with the temperature rise, showing that the adsorption is favorable in higher temperatures.

3.5. Adsorption isotherm

The Langmuir and Freundlich equations are commonly used for describing adsorption equilibrium in water and wastewater treatment applications. The linear form of Langmuir isotherm is given by the following equation:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{bq_mC_e} \tag{4}$$

where q_e is the amount of adsorbed material at equilibrium (mg g⁻¹), C_e is the equilibrium concentration (mg L⁻¹), b is the energy or net enthalpy of adsorption constant (L mg⁻¹), and q_m

Table 3

Adsorption rate constants (q_e) and coefficients of correlation (D) from the pseudo-second-order kinetic model at different temperatures.

T (°C)	$k_{2,ads}$ (g mg ⁻¹ min ⁻¹)	Experimental q_e (mg g ⁻¹)	Calculated q_e (mg g ⁻¹)	D (%)	R ²
10	0.65	6.99	7.14	2.14	0.99
25	0.80	7.00	7.19	2.71	0.99
45	1.36	7.09	7.24	2.11	0.99

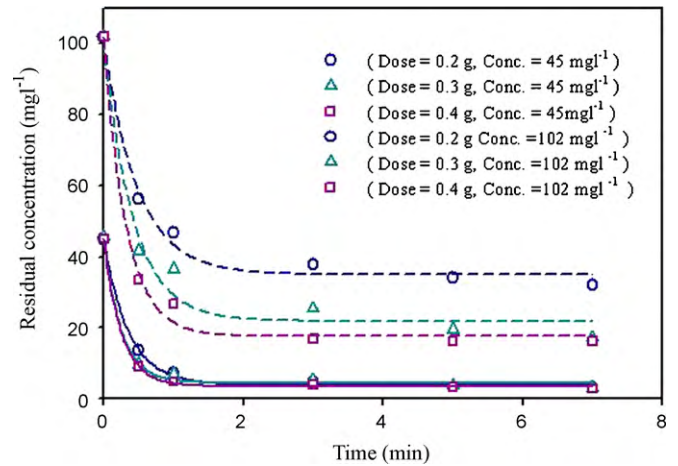


Fig. 4. Effect of initial concentration on adsorption of Sr(II) with different amount of adsorbent (stirring speed = 720 rpm; T = 25 °C; initial concentrations = 45.5 and 102 mg L⁻¹).

Table 4

Freundlich and Langmuir adsorption constants associated to the adsorption isotherms of strontium ion on almond green hull.

Langmuir constants			Freundlich constants		
q_m (mg g ⁻¹)	b (L mg ⁻¹)	R ²	K_f (mg g ⁻¹)	n	R ²
116.30	0.0048	0.99	3.59	1.95	0.99

is the Langmuir constant related to maximum adsorption capacity. When $1/q_e$ was plotted against $1/C_e$, straight lines with slope of $1/bq_m$ were obtained (Fig. 5), indicating that the adsorption process follows the Langmuir isotherm. Constants of the model, b and q_m were calculated and these values are given in Table 4.

The essential characteristics of the Langmuir isotherm which expressed in terms of a dimensionless separation factor (R_L) can be calculated from Eq. (5).

$$R_L = \frac{1}{1 + bC_0} \tag{5}$$

where b is the Langmuir constant and C_0 is the initial metal-ion concentration (mg L⁻¹).

The parameter (R_L) is related to the shape of the isotherm according to the following adsorption characteristics: $R_L > 1$ represents unfavorable adsorption; $R_L = 1$ corresponds to a linear relationship; $0 < R_L < 1$ is favorable adsorption and $R_L = 0$ is irreversible [28].

R_L values presented in Table 5 indicate favorable adsorption of strontium ion for all concentrations at 25 °C.

The adsorption data is also analyzed using Freundlich model. The logarithmic form of Freundlich model is given by following equation:

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \tag{6}$$

where q_e is the amount of adsorbed material at equilibrium (mg g⁻¹), C_e is the equilibrium concentration of the adsorbate (mg L⁻¹) and K_f and n are Freundlich constants related to adsorption capacity and adsorption intensity, respectively. The plot of $\log q_e$ against $\log C_e$ for the adsorption data of strontium is shown in Fig. 6.

Table 5

R_L values for adsorption of Sr(II) ion on almond green hull at 25 °C.

Initial cone. (mg L ⁻¹)	161	229	297	538	635
R_L	0.56	0.47	0.41	0.27	0.24

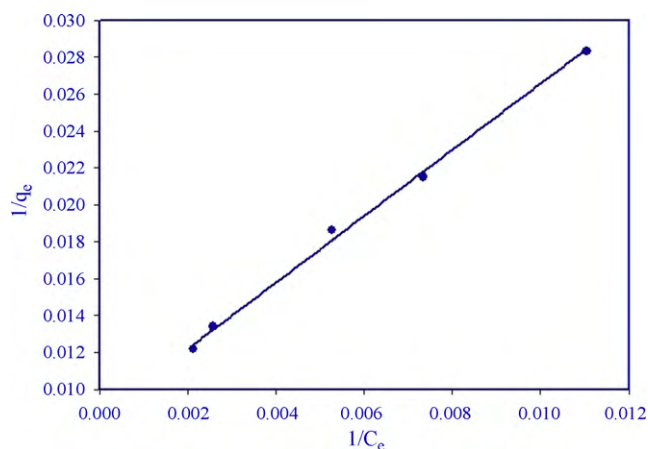


Fig. 5. Langmuir isotherm for adsorption of strontium ion (stirring speed = 720 rpm; $T = 25^\circ\text{C}$).

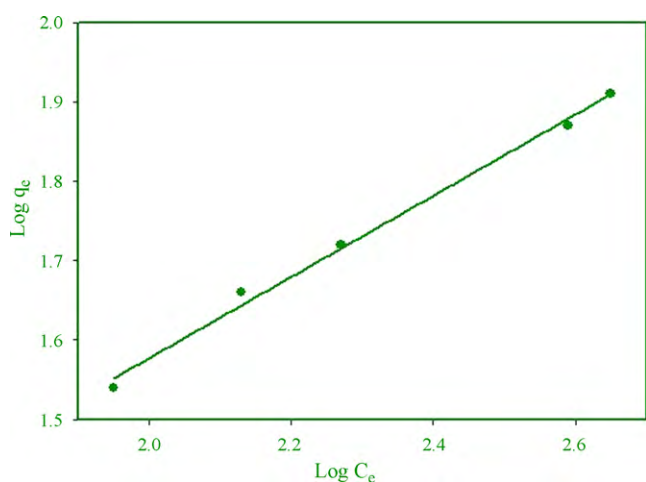


Fig. 6. Freundlich isotherm for adsorption of strontium ion (stirring speed = 720 rpm; $T = 25^\circ\text{C}$).

The Freundlich parameters are also given in Table 4. The sorption parameters were determined at fixed temperature (25°C) for a concentration range of $161\text{--}635\text{ mg L}^{-1}$. All solutions contained a fixed mass of 0.1 g of adsorbent. The good correlation coefficients show that both Langmuir and Freundlich models are suitable to predict the adsorption equilibrium state of strontium ion.

4. Conclusions

The feasibility of using a new adsorbent derived from almond green hull for the rapid removal of strontium ion from aqueous solutions was studied. The following conclusions can be drawn from the present study:

- Efficient and rapid adsorption takes place within only 2 min contact time.
- Batch adsorption studies show that approximately 96% of the strontium ions are removed at 25°C .
- Kinetic study proves that the adsorption of strontium ion follows the pseudo-second-order kinetic model.
- The equilibrium isotherm analysis indicates that the experimental adsorption data of strontium onto the almond green hull can be correlated by both Langmuir and Freundlich isotherms.
- Low cost of this adsorbent with its rapid adsorptive capability offer a promising technique for partial treatment of industrial wastewaters.

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