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Comparison of different rice straw based adsorbents for chromium removal from aqueous solutions

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ABSTRACT

Chromium is a well known highly toxic heavy metal and one of the major pollutants in the tannery wastewaters. This research aims at studying the feasibility of using different rice straw-based adsorbents, fine rice straw (FRS), rice straw carbon (RSC), and rice straw activated carbon (RSAC) for removal of chromium from aqueous solution. In batch experiments, effect of operating variables such as contact time, adsorbent dose, pH of solution, and initial chromium concentration have been studied. The best operating conditions were: FRS—equilibrium time 12 h, FRS dose 20 g/l, pH 2; RSC—equilibrium time 8 h, RSC dose 12 g/l, pH 2; RSAC—equilibrium time 4 h, RSAC dose 8 g/l, pH 2. The maximum adsorption achieved at pH 2.0 for all studied adsorbents. The adsorption results for the studied adsorbents were fitted to Langmuir and Freundlich isotherms with correlation coefficient of 0.99, 0.95, respectively, and the maximum adsorption capacity was 7.99, 18.83, and 40.32 mg/g. Desorption of chromium from different rice straw (RS) particles using acid and base treatment exhibited a higher desorption efficiency by more than 85, 95, and 97% for FRS, RSC, and RSAC, respectively. The present study showed that RS could be used as a biosorbent for removal of chromium from aqueous solution with a limited ratio.

Keywords: Adsorbent; Chromium; Heavy metals; Raw agricultural wastes; Rice straw

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1. Introduction

In the last few years, industrialization in many regions has increased the discharge of heavy metals in the environment [1]. Toxic metals can seriously affect humans, plants, and animals which have been involved in causing a large number of afflictions [2]. Therefore, removing heavy metals from contaminated water is a challenging area of research due to the health and environmental aspects [3,4].

Chromium exists in natural water in two main oxidation states, hexavalent chromium (Cr(VI)), and trivalent chromium (Cr(III)). The main industrial sources of chromium pollution are tanneries, electroplating, metal processing, wood preservatives, paint, pigments, textile, dyeing, steel fabrication, and canning industry [5,6]. As per US EPA, Chromium concentration is 0.05 mg/l in drinking water and 0.10 mg/l in surface water [7]. The tanning industry is one of the largest pollutants all over the world. The total number of tanneries in Egypt is more than 300, of which more than 85% adopt the chromium tanning process [8].

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Several systems have been studied for chromium removal from aqueous solutions/wastewater. Such as reduction followed by electrochemical precipitation, chemical precipitation, chemical oxidation–reduction, ultrafiltration, ion exchange, reverse osmosis, solvent extraction, electrodialysis, electrochemical, coagulation, evaporation, and adsorption [5,9,10]. High capital and operational costs are the major drawbacks of these systems [6,11]. The high cost of activated carbon sometimes limits its applicability for heavy metal removal [12–14]. Therefore, the interest of researchers is increasing using alternative adsorbents, which are quite low-cost, easily available, and effective [15]. Many studies have been conducted on the use of agricultural wastes as an absorbent for heavy metals removal [16–22] hazelnut shell.

Agricultural wastes-based adsorbents have been approved as a low-cost and effective for heavy metals removal. In the recent years, several studies have been reported on various low-cost adsorbents such as maple sawdust; sugar industry waste [23]; soya cake [24]; activated carbon derived from fertilizer waste [25]; tea factory waste; Turkish brown coal [26]; moss peat [27]; coconut trees [28]; lignocellulosic residues [29]; rice brand [30]; activated neem leaves [31]; activated tamarind seeds [32]; etc. However, many of these naturally available adsorbents have low chromium adsorption capacity. Several natural or agricultural wastes have been used for the removal of heavy metal from industrial wastewater. In general natural or agricultural waste contains different functional groups like hydroxyl, aldehyde, aliphatic acid, alkene, amide, aromatic nitro, silicate, sulphonate, etc. Thus, there is a need to develop or find innovative low-cost adsorbents with an affinity toward metal ions for the removal of Chromium from aqueous solution which leads to high adsorption capacity [33]. The present study was undertaken to compare different rice straw-based adsorbent for Cr(III) removal.

2. Materials and methods

2.1. Adsorbent preparation and characterization

The raw RS was collected from the local farms in Qalioubia government, north Cairo, Egypt. It was naturally dried and cut to fraction of particles. Then, the fraction of particles was sieved. The fine rice straw (FRS) contains the fraction of RS with particles size of ≤200 μm. The size was selected based on other reported study [34]. The FRS was stored in a desiccated glass bottle. Some of the prepared FRS was used in the preparation of the rice straw carbon (RSC) which was prepared by soaking, washing, and drying the RS in 10% potassium hydroxide solution (KOH) for 24.00 h. Then, it was followed by washing with distilled water to remove free KOH and drying at 105 ± 5°C for 24.00 h. The same previous process (preparation of RSC) was done for preparing the rice straw activated carbon (RSAC). Then, it was followed by washing with distilled water to remove free KOH and drying at 105 ± 5°C for 24.00 h. The characteristics of each FRS, RSC, and RSAC are as shown in Table 1. Fig. 1 shows the fourier transform infrared (FTIR) spectrum for the FRS adsorbent. Fig. 1 indicates antisymmetric stretching vibration, and presence all of hydroxyl, aliphatic, aromatic, carboxylic, and phenolic groups.

2.2. Chromium aqueous solution

Chromium (III) aqueous solution was prepared by dissolving the specific amounts of chromic tri-chloride ($CrCl_3.6H_2O$) in distilled water. It was prepared weekly and stored at 4°C. The selection of the studied Cr(III) concentration was based on the literature and preliminary studies on the common chromium concentration in the tannery wastewater [15]. Cr(III) concentration was determined by atomic absorption spectrophotometer method on spectra AA Varian 10/20 computerized spectrophotometer located in the National Research Center's laboratory. Under the experiment's conditions, there is no way to produce Cr(VI) from oxidation of Cr(III).

2.3. Batch adsorption and isotherm studies

Batch experiments were conducted using a 600 ml Pyrex reactor with 500 ml of the chromium aqueous solution. The mixture was agitated at a speed of 60 rpm using Jar test apparatus (test di cession C6F) for 2 min at room temperature (25 ± 3°C). Thereafter, the necessary amount of absorbent was added to the aqueous solution simultaneously with pH adjustment to the required value using H_2SO_4 [35]. Effects of contact time, initial chromium concentration, pH, and

Table 1
Characteristics of the used adsorbents

	Characteristics		
Adsorbent	Surface area (m^2/g)	Micropore volume (cm^3/g)	Pore size (μm)
FRS	133.20	0.03912	11.75
RSC	198.80	0.08450	6.14
RSAC	315.10	0.15871	4.08

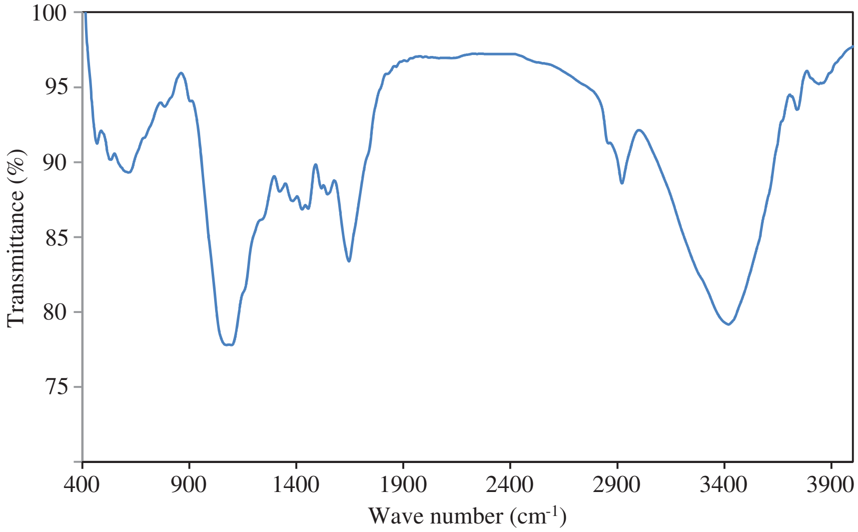

Fig. 1. FTIR spectrum of FRS.

adsorbent dose were studied. Adsorption isotherm was determined by batch equilibrium test at the optimum pH and contact time for adsorption [10]. The experimental data were modeled using Langmuir and Freundlich isotherms.

2.4. Desorption regeneration

Desorption studies are conducted by batch experiments. The 50 g of saturated adsorbent with chromium was first treated with 500 ml of NaOH solution (concentration 32%) for 1 d. After the NaOH treatment, the adsorbent was separated from the solution and washed with distilled water. Washed adsorbent is further regenerated with 500 ml of HCl solution (concentration 30%) for 1 d. After that, the adsorbent was washed with distilled water and dried at room temperature (25 ± 3˚C) for 6 h. Desorption experiments were carried out with different initial concentrations of chromium from 50 to 200 mg/l, while maintaining the adsorbent amount fixed at pH 2 and room temperature (25 ± 3˚C) [36].

3. Results and discussion

3.1. Effect of contact time and initial chromium concentration

To study the effect of contact time and the initial Cr(III) concentration on the adsorption of Cr(III) on FRS, initial Cr(III) concentration were 50, 100, 150, and 200 mg/l; contact time were 0.25, 0.50, 1, 2, 4, 8, 12, 24, and 48 h. Other adsorption conditions were pH 2 and sorbent dose 20 g/l when using FRS, 12 g/l when using RSC, and 8 g/l when using RSAC. The effect of the contact time and the initial Cr(III) concentration on the removal of Cr(III) ion by FRS, RSC, and RSAC are shown in Fig. 2(a)–(c). The decrease of Cr(III) removal with the increase of initial Cr(III) dose could be related to the ratio between the available adsorption sites to Cr(III) molecules which led to the saturation of the sites more rapidly [15]. The results show increase of Cr(III) removal with the increase of the contact time. Increasing contact time further 12 h (FRS), 8 h (RSC), and 4 h (RSAC) did not significantly increase the Cr(III) removal. This could be ascribed to the large concentration gradient between the film and the available pore sites in the initial stages of adsorption of Cr(III) and hence the rate of adsorption of Cr(III) is faster. The rate of adsorption decreases in the later stages of the Cr(III) adsorption could be due to the slow pore diffusion of the solute ion into the bulk of the adsorbent [35]. Based on the results, equilibrium adsorption is attained in 12, 8, and 4 h when using FRS, RSC, and RSAC, respectively. Similar results are also reported by researchers for a variety of adsorption systems [15,35–38].

3.2. Effect of initial pH

Study of the pH effect on the adsorption of Cr(III) on RS, initial pH was varied in the range 1–6. Other operating conditions were: FRS—FRS dose 20 mg/l, contact time 12 h; RSC—RSC dose 12 mg/l, contact time 8 h; RSAC—RSAC dose 8 mg/l, contact time 4 h all with initial Cr(III) concentration 50 mg/l. The

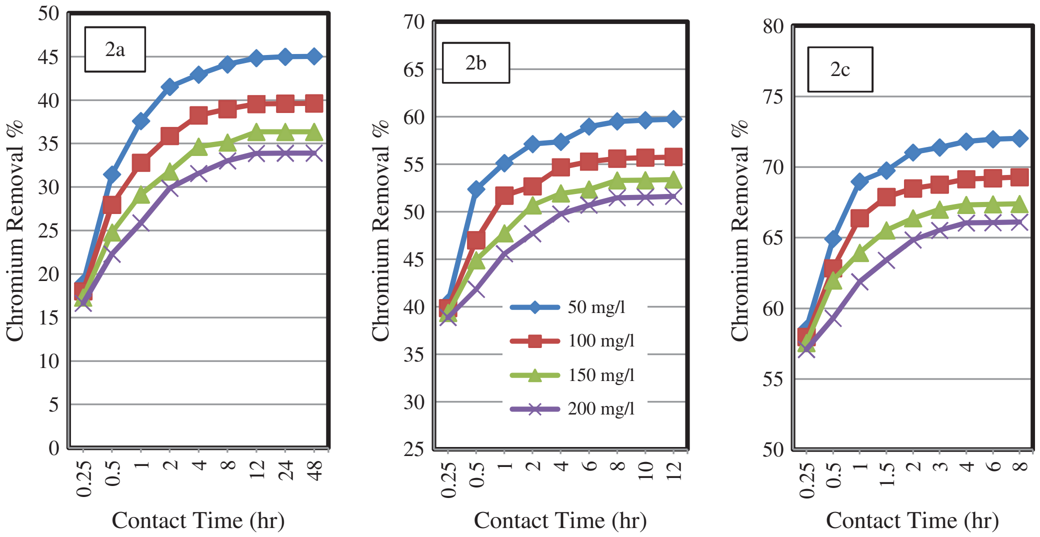

Fig. 2. Effects of contact time and initial Cr(III) concentration on chromium removal.

results of Cr(III) removal in the pH effect experiment for different adsorbents are shown in Fig. 3. As shown in the results, increase of Cr(III) removal with the decrease of pH in the range 2–6 and the maximum adsorption occurred at pH 2. Similar results of the high adsorption of the Cr(III) in the very acidic had been reported in the literature [39–42]. The high adsorption of the Cr(III) on RS low pH value could be ascribed to the high quantity of hydronium ions (H^+) which could promote the adsorption of negatively charged Cr(III) ions [43].

The more positive the surface charge of the biomass, the faster the removal rate of Cr(III) in the rice straw solution, since the binding of anionic Cr(III) ion species is enhanced. The high values of pH as 5 and 6, it were noticed that the Cr(III) settled in the bottom. At higher pH values up to pH 5, the abundance of hydroxyl ions (OH^{-1}) increased hindrance to diffusion of $HCrO_4^{-1}$ anion and at pH 5–6 to diffusion of CrO_4^{-2} anion, thereby reducing adsorption [15].

Similar observations have been reported for adsorption of Cr(III) by RS particles at pH 2 [34]; activated carbon prepared from apricot stone and almond shell at pH 1 [44]; coconut shell at pH 2.5 [45]; coconut shell and fibre at pH 2 [46]; hazelnut shell at pH 2 and 1 [47,48]; used tyre, sawdust, and bituminous coal at pH 2 [49]; *T. arjuna* nuts at pH 1 [50]; and *H. brasiliensis* sawdust at pH 2 [38].

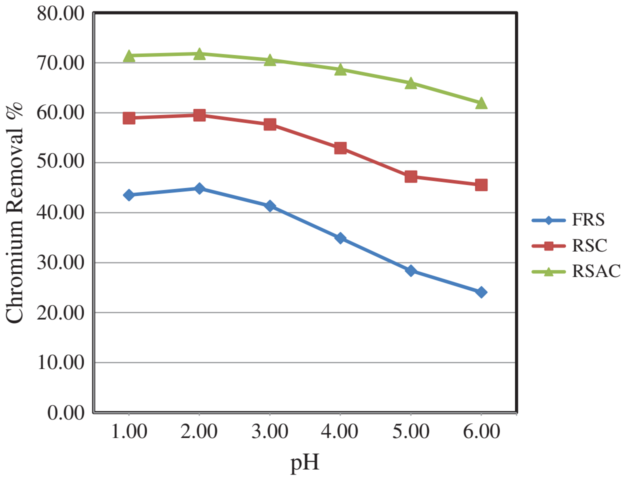

Fig. 3. Effect of pH on chromium removal.

3.3. Effect of adsorbent dose

The effect of the RS dose as a sorbent material on the removal of Cr(III) ion is shown in Fig. 4. The adsorbent doses were in the range of 5–40 g/l for FRS, 4–20 g/l for RSC, and 4–12 g/l for RSAC. The pH was 2 at all. However, the previously determined the equilibrium time of 12 h for FRS, 8 h for RSC, and 4 h for RSAC have been considered. As shown in Fig. 4, the chromium removal is affected by adsorbent dose and adsorbent type. It is increased with the increase of adsorbent dose. At same conditions of dose, pH, and initial concentration RSAC achieved the highest chromium removal more than RSC and FRS. The nature of adsorbent and the available adsorption sites affect the rate of adsorption of Cr(III). The mechanism of solute transfer to the solid includes diffusion through the fluid film around the adsorbent particle and diffusion

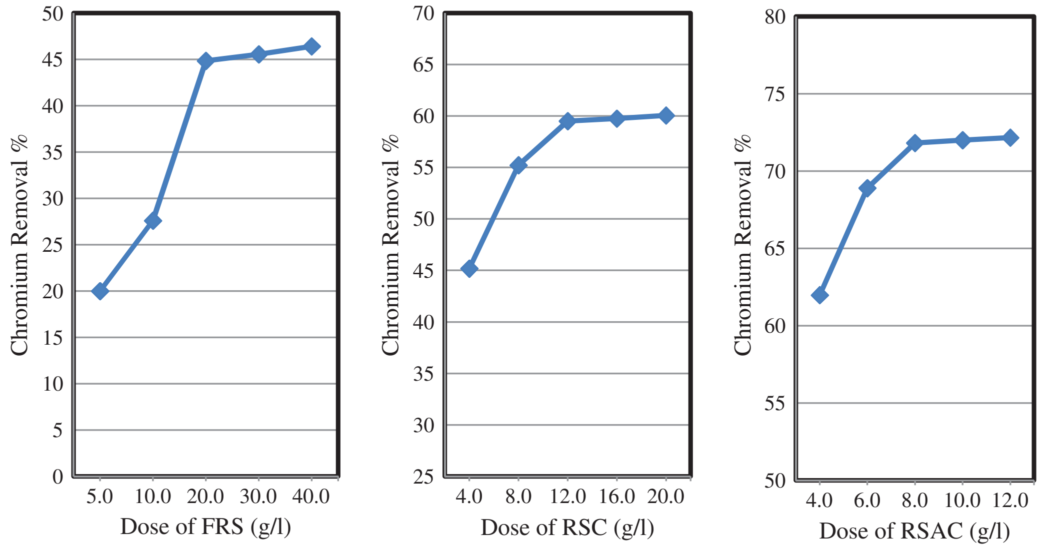

Fig. 4. Effects of adsorbent dose on chromium removal.

through the pores to the internal adsorption sites. Similar results are also reported by researchers for a variety of adsorption systems [15,34–36].

3.4. Adsorption isotherms

Experimental isotherms are useful for describing adsorption capacity to facilitate evaluation of the feasibility of this process for a given application and for the predictive modeling procedures for analysis and design of sorption systems [34]. The Langmuir and Freundlich isotherms are most frequently used to represent the data of sorption from solution. In this study, the adsorption equilibrium of chromium onto RS different particles was modeled using the Langmuir and Freundlich isotherms at different adsorbent doses. The Langmuir adsorption isotherm:

$$q_e = Q_{max} \cdot b \cdot C_e/(1 + b \cdot C_e) \tag{1}$$

In which q_e is the amount of solute adsorbed per unit weight of the solid adsorbent; C_e is the concentration of solute remaining in the solution at equilibrium; Q_{max} is the number of moles of solute adsorbed per

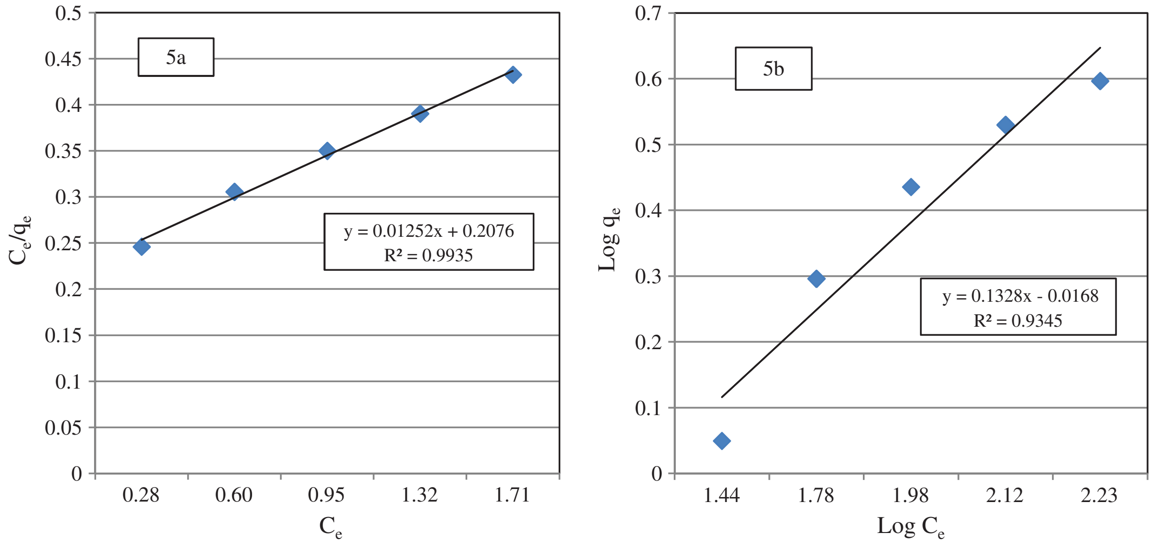

Fig. 5. Langmuir (5a) and Freundlich (5b) isotherm for adsorption of Cr(III) by FRS.

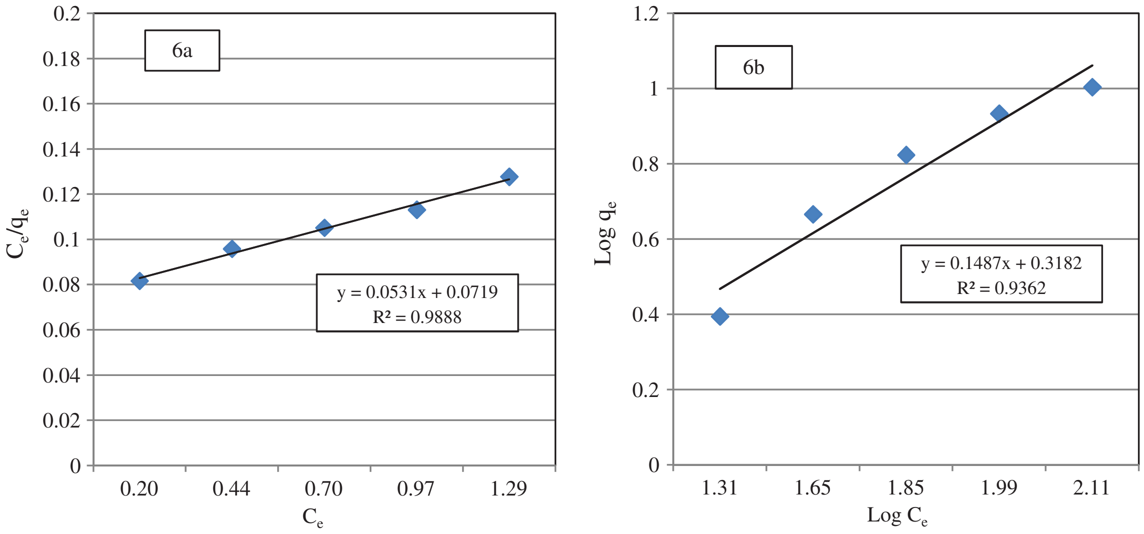

Fig. 6. Langmuir (6a) and Freundlich (6b) isotherm for adsorption of Cr(III) by RSC.

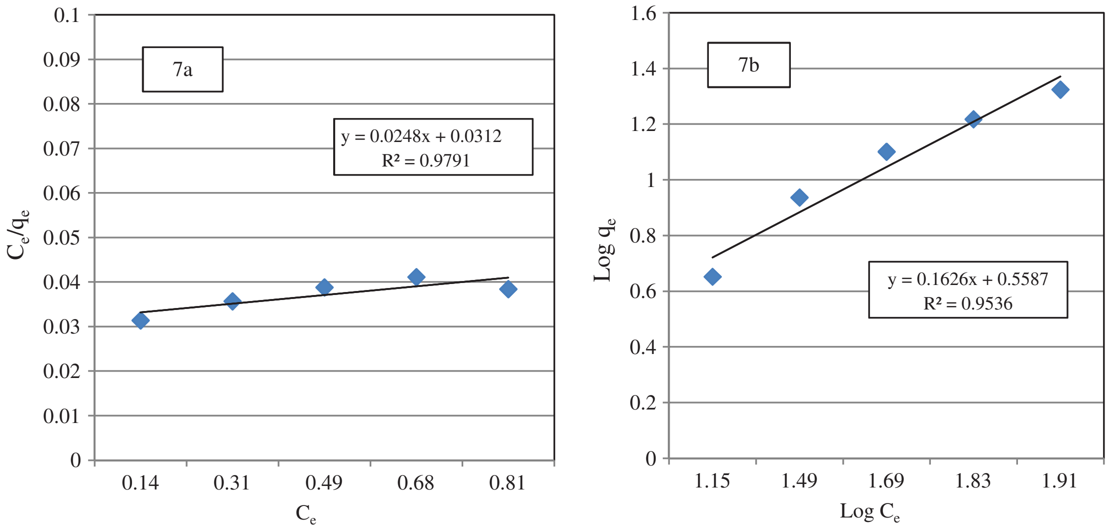

Fig. 7. Langmuir (7a) and Freundlich (7b) isotherm for adsorption of Cr(III) by RSAC.

unit weight of adsorbent in forming a monolayer on the surface (monolayer limiting adsorption capacity); b is constant related to the energy of adsorption.

$$q_e = K_f \cdot C_e^{(1/n)} \tag{2}$$

In which K_f and $1/n$ are the Freundlich constants (adsorption capacity) which were considered to be the relative indicators of adsorption capacity and adsorption intensity.

Table 2
Langmuir and Freundlich isotherm for adsorption of Cr(III)

	Model			
	Langmuir		Freundlich	
Adsorbent	Q_{max}	b	K_f	$1/n$
FRS	7.987	0.603	0.962	0.133
RSC	18.832	0.738	2.081	0.149
RSAC	40.323	0.795	3.620	0.163

Table 3
Comparison of maximum adsorption capacities of chromium in various adsorbents with and without activation

S. No.	Sorbent	Q_{max} (mg/g)	References
1	Almond green hull—biosorbent	2.040	[15]
2	Sawdust—carbon	41.520	[36]
3	Coconut shell—carbon	20.000	[45]
4	Almond shell—activated carbon	2.400	[46]
5	Hazelnut shell—activated carbon	17.700	[50]
6	Almond shell—carbon	10.600	[51]
7	Fine Rice straw—biosorbent	7.987	This study
8	Rice straw—carbon	18.832	This study
9	Rice straw—activated carbon	40.323	This study

The isotherms were fitted to the linear form of each Langmuir equation $[C_e/q_e = 1/(b \cdot Q_{max}) + C_e/Q_{max}]$ and Freundlich equation $[\log q_e = \log K_f + (1/n) \cdot \log C_e]$as shown in Figs. 5–7. Comparatively, the obtained R^2 values were more satisfactory for Langmuir model. Using the slope and the intercept of this line, b and Q_{max} were found as shown in Table 2. For Freundlich model, with using the slope and the intercept of this line, K_f and $1/n$ were found to be as shown in Table 2.

The basic assumption of Langmuir model was based on monolayer coverage of the adsorbate (initial chromium solution) on the surface of adsorbent and which was an indication of the fact that the sorption of chromium onto the RS generates monolayer formation [34]. Comparison of maximum adsorption capacities of chromium was tabulated in Table 3 shows that RS is an abundant low-cost biomaterial which had high chromium uptake capacity that should be seriously considered for the disposal of chromium from water and wastewater.

The adsorption capacity of chromium onto RS was compared with several low-cost adsorbents and they are reported in Table 3. As shown in the Table 3, the adsorption capacity of chromium on different RS is around (7.99–40.32) mg/g at pH 2 and room temperature 25 ± 3˚C. RS in this study possesses reasonable adsorption capacity in comparison with other sorbents.

3.5. Regeneration studies

The saturated adsorbent which contains chromium is not safe for the disposal due to the stringent environmental constraints. It is important and appropriate to propose a method for the regeneration and reuse of adsorbent so as to reduce the load on environment in terms of disposal of polluted adsorbent [36]. In the

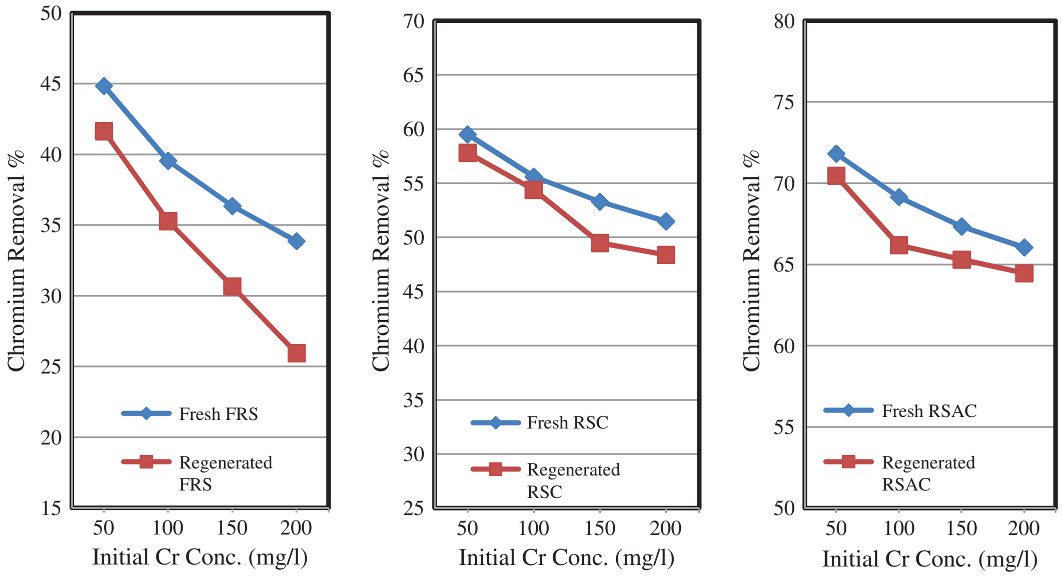

Fig. 8. Comparison for the percentage removal of Cr(III) using fresh and regenerated RS.

present study, RS is regenerated and is used for the removal of chromium at different initial Cr(III) concentration in the range of 50–200 mg/l. Fig. 8 shows the comparison of the percentage removal of chromium using fresh and regenerated different RS. The regenerated RS is again used for the adsorption of Cr(III) using the same initial Cr(III) concentration ranging between 50 and 200 mg/l as used for fresh RS for a contact time 12, 8, and 4 h. and sorbent dose 20, 12, and 8 g/l using FRS, RSC, and RSAC, respectively, at pH 2.

The percentage removal of chromium obtained found to decrease for FRS from 41.64 to 25.95% as compared to the corresponding values for fresh adsorbent ranging from 44.84 to 33.87%, and decrease for RSC from 57.81 to 48.39% as compared to the corresponding values for fresh adsorbent ranging from 59.51 to 51.47%, and decrease for RSAC from 70.46 to 64.46% as compared to the corresponding values for fresh adsorbent ranging from 71.82 to 66.05%. These results using regenerated RS exhibit higher desorption efficiency by more than 85% and up to 97% of the fresh RS for the removal of chromium. Since, Cr(III) adsorption is an example of physical adsorption; it is possible to regenerate the FRS, which can be considered for reuse as indicated by the results obtained in this study. The adsorption of chromium onto the RS is highly pH dependent. Hence, desorption of chromium is accomplished by increasing the solution pH [36].

4. Conclusion

The RS acts as a reasonably good adsorbent for the removal of Cr(III) from aqueous solution. The following conclusions had been listed based on the results of the present study and the scientific information derived from literature:

- The maximum adsorption of chromium occurred at pH 2.
- Both the Langmuir and Freundlich isotherms were suitable for describing the biosorption of chromium.
- Desorption of chromium from RS using acid and base treatment exhibited a higher desorption efficiency by more than 85% and up to 97%.
- The Langmuir and Freundlich adsorption models had been used to represent the experimental data fitted very well to the Langmuir isotherm model.

The present study showed that RS could be used as low-cost sorbent for removal of chromium from aqueous solution with a limited ratio.

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