

Removal of Congo red dye from aqueous solutions by a low-cost adsorbent: activated carbon prepared from Aloe vera leaves shell

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Abstract

Background: Synthetic dyes have several harmful effects on human health as well as aquatic life. In this study, activated carbon (AV-AC), based on Aloe vera leaf shells, was used as a novel agricultural adsorbent, one that is low-cost and available for the removal of Congo red (CR) as a carcinogenic dye from aqueous solutions.

Methods: In the batch system, the influence of different parameters like contact time, pH, adsorbent dosage, and initial CR concentration were examined on the dye removal from liquid medium. The experimental data were fitted by pseudo-first-order and pseudo-second-order kinetics, and also Langmuir and Freundlich isotherms models.

Results: The optimum contact time and pH for the uptake of CR were obtained at 20 minutes and acidic pH of 2. The maximum uptake capacity of CR dye by AV-AC was 1850 mg/g. The results showed that the experimental data were well-fitted by the pseudo-second-order kinetic model ($R^2 > 0.99$) and Freundlich isotherm model ($R^2 > 0.99$).

Conclusion: According to the results of our study, the AV-AC is a low-cost, non-toxic, and effective adsorbent for the uptake of CR dye from aqueous media.

Keywords: Aloe Vera, Congo red, Kinetic, Charcoal

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Introduction

Water pollution caused by discharge of wastewater as a result of industrial activities has been considered one of the grave environmental issues in the world, especially in developing countries (1-3). The discharge of many organic and inorganic pollutants into water media by different industries, as a global environmental problem, has stimulated worldwide attention because of their harmful effects on the environment and also human health (4,5). One of the most important of these pollutants is dye substances. Various industries generate large volumes of dye containing effluents, such as the textiles, paper, rubber, leather, cosmetics, and food industries. The disposal of these coloured wastewaters without acceptable treatment

into aquatic bodies will adversely influence the natural aquatic environment as well as have carcinogenic and mutagenic effects on human beings (6,7). Thus, removal of these dyes is essential before these are discharged into the water bodies (8). Among different kinds of anionic dyes, Congo red (CR) (4-aminonaphthalene-1-sulphonic acid) is a benzidine-based anionic diazo dye, which is generated by some industries, including textiles, printing and dyeing, paper, rubber and plastic industries. It is toxic for many organisms and is a doubted carcinogen and mutagen dye (6). CR is very stable to biological degradation; it has a complex aromatic structure (5,9,10). Several physical, chemical and microbial methods, including membrane separation, ion exchange, biological



degradation, coagulation-flocculation, electrochemical techniques, chemical oxidation, and adsorption, have been used to remove CR from coloured water (11,12). Among these processes, the adsorption technique is current due to its simplicity, high sorption capacity, environmental friendliness, non-toxicity, and availability of a wide range of sorbents. Adsorption onto activated carbon because of its high surface area and high uptake capacity has been commonly used as an adsorbent for the remove of organic pollutants, but it is high-priced, and is hard to regenerate (2,13). In addition to activated carbon, some adsorbents—such as montmorillonite (14), rice husk (15), bentonite (16), rice hull ash (17), *Azadirachta indica* leaf (18), Jujuba seeds (19), red mud (20), polypyrrole–polyaniline nanofibres (21), anion exchange membrane (22), and ball-milled sugarcane bagasse (23)—have been employed for the removal of CR dye. Today, more attention is being given to the use of low-cost adsorbents. Aloe vera grows in warm tropical areas such as the United States, Africa, Australia, South America, India and Iran (2). The original material of Aloe vera is used in the production of latex substance and drugs, but the Aloe vera leaves shell are by-products of the agricultural industries (2,3,24). Omid-Khaniabadi et al used the Aloe vera leaf wastes as a novel adsorbent for the adsorption of pollutants such as 4-chlorophenol (2), methylene blue (24), aniline and methyl orange (6) from aqueous solutions. In all the abovementioned contaminants, the results obtained were of interest for their removal. In this study, Aloe vera leaves shells were used as a cheap adsorbent to produce the activated carbon for CR dye uptake from synthetic wastewater.

Materials and Methods

Materials

Aloe vera leaves were collected from suburban farms of Ahvaz city in Khuzestan province, Iran. The chemical substances—which included CR dye, H_2SO_4 and NaOH—were acquired from Merck Co, Germany. The molecular structure of CR dye is presented in Figure 1. Dye stock solution (1000 mg/L) was prepared by dissolving 1 g of CR dye in 1 L deionized water and different concentrations were made by dilution of the stock solution. A rotary shaker (Behdad-Rotomix model) was used to mix the suspension of dye and AV-AC at 150 rpm.

Preparation of adsorbent

All the waste was carefully washed by deionized water to eliminate the pollutants and impurities. It was then dried in an oven at $105^\circ C$ for 24 hours. The dried shells

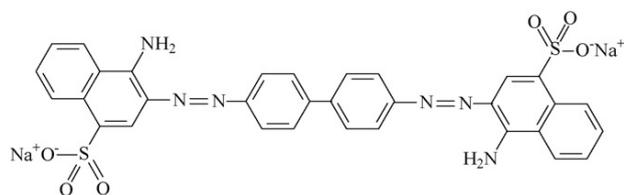


Figure 1. Molecular structure of Congo red (CR).

were crushed through a laboratory mill and were then sieved. The particle sizes with a range of 300–500 μm were carbonized in a furnace at $550^\circ C$ for 20 minutes in a without-air condition and prepared activated carbon was again crushed and sieved (mesh no=40) for the uptake experiments.

Characterization and analysis

For elemental analysis of AV-AC samples, a Heraeus Elemental Analyser (Jobin-Yvon Ultima ICP-AES) was used. The surface morphology of the adsorbent was characterized (before and after CR uptake) by a scanning electron microscope (SEM, Jeol Model Jsm-T330). The remaining concentration of CR in the solution was measured through an UV-Vis spectrophotometer (PG Instrument Limited Model) at a maximum absorbance wavelength of 495 nm (λ max).

Zero point of charge (pH_{zpc}) of the adsorbent was determined by preparing 50 mL 0.01 M NaCl solution in a 100 mL Erlenmeyer flask. The initial pH in each flask was adjusted in the range of 2 to 12 by 0.1 N H_2SO_4 or NaOH solutions. In each flask containing NaCl solution, 0.1 g of sorbent was poured, and then the suspensions were mixed for 24 hours by a rotary shaker. After mixing stage, the final pH of solutions was determined to identify pH_{zpc}.

Adsorption experiments

The batch adsorption experiments were conducted to identify the effects of various factors including contact time (0–100 minutes), pH (2–12), adsorbent dosage (0.5–5 g/L), and adsorbate content (100–500 mg/L) on the sorption of CR by using AV-AC. In all, 44 samples were taken, and the results analysed and the figures drawn in Excel software. All the experiments were performed at laboratory temperature ($25^\circ C$) and were then shaken at 150 rpm in a 250 mL Erlenmeyer flask. After agitation time, the suspension was filtered through a fibreglass paper (0.45 μm) and the absorbance of the clear sample was analysed to determine the remaining CR. The amounts of CR adsorbed on AC-AV and the sorption percentage (%) were computed by the following equations:

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

$$Sorption(\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (2)$$

where q_e (mg/g) is the uptake capacity the AV-AC. C_0 and C_e are respectively the initial and equilibrium dye concentrations in the solution (mg/L). V and m are respectively the volume of solution (L) and adsorbent mass (mg).

Results

Characterization

The elemental analysis of chemical composition of the AV-AC illustrated that Ca, O, K, and Mg are the majors that formed 92.7% of the weight (wt. %) of the adsorbent.

Na (5.92%) and Cl (1.35%) are the minors comprising the adsorbent (2,3). Figure 2A and 2B respectively explain the morphology of the surface of activated carbon before and after the CR uptake.

Effect of contact time

The effects of different contact times on the sorption of CR dye by Aloe vera leaves shell-based activated carbon (AV-AC) at the time intervals of 0–100 minutes in 100 mL dye solution and 1 g/L of AV-AC at the initial pH of solution were investigated. The results are shown in Figure 3A.

Kinetics study

The experimental data of CR dye by AV-AC was fitted by three adsorption kinetics, including pseudo-first-order, pseudo-second-order, and intraparticle diffusion models to obtain better understanding of the sorption process. The pseudo-second-order kinetic model and parameters of the uptake of CR sorption by AV-AC are illustrated in Figure 3B and Table 1 respectively. As can be seen in Table 1, the highest correlation coefficient (R^2) of CR adsorption was related to the pseudo-second-order kinetic. Therefore, the adsorption process of CR dye by AV-AC was followed by the pseudo-second-order kinetic model.

Effect of solution pH and adsorbent dosage

The effect of solution pH (2–12) on the uptake of CR (initial concentration = 100 mg/L) by AV-AC has been shown in Figure 4A. As is obvious, by increment the solution pH, from 2 to 12, the uptake capacity of the adsorbent was decreased. The maximum uptake of CR (100 mg/g) by AV-AC was obtained in pH 2. Thus, pH 2 has been selected as the optimum pH for the next stages. As seen in Figure 4A, the isoelectric point of the activated carbon from natural adsorbent was observed at pH of 11.3. There were positive charges on the AV-AC surface at the lower pH values of the isoelectric point, which will promote a reaction with CR as an azo dye. The effect of different adsorbent dosages of AV-AC in an initial CR concentration (100 mg/L) at 25°C was

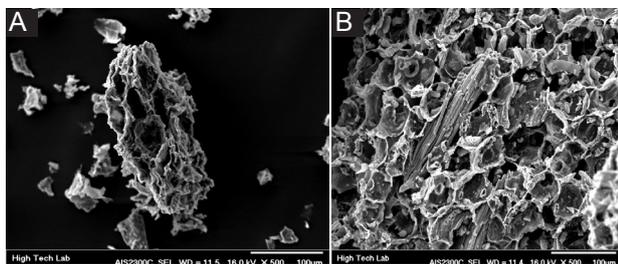


Figure 2. SEM image the AV-AC (A) before and (B) after the uptake process of Congo red (CR).

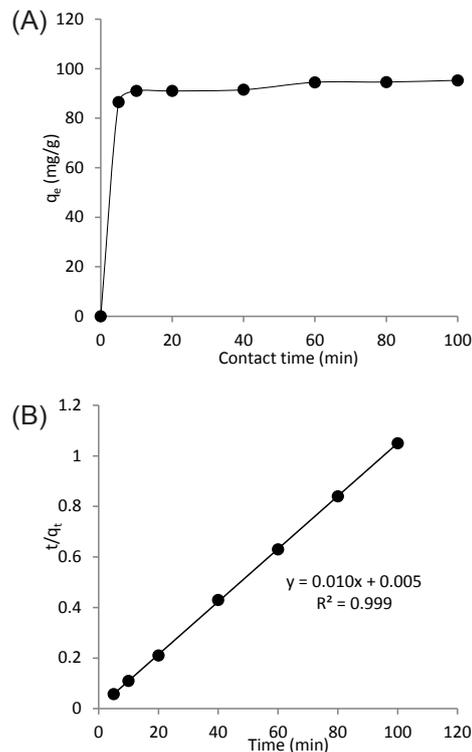


Figure 3. (A) The effect of contact time on the sorption of Congo red (CR) dye by AV-AC (CR con. = 100 mg/L, pH = 7 and adsorbent dosage = 1 g/L) and (B) pseudo-second-order kinetic model.

examined. The results are explained in Figure 4B.

Effect of initial dye concentration

The influence of initial content (100 to 500 mg/L) on the uptake capacity of CR by AV-AC was evaluated. As shown in Figure 5A, the adsorption value of AV-AC at the beginning was quickly increased by the rise in CR concentration in the dye solution.

Isotherm study

Two uptake isotherm models, including Langmuir and Freundlich, were applied to fit the experimental data of CR dye removal by AV-AC.

As can be seen in Figure 5B, the amount of K_f and n were obtained from the cut-off and gradient of liner plotting of $\ln q_e$ vs. $\ln C_e$ respectively. The uptake isotherm parameters of the Langmuir and Freundlich models are presented in Table 2. As is obvious, the value of the correlation coefficient (R^2) of the Langmuir ($R^2 > 0.96$) model was lower than Freundlich ($R^2 > 0.99$) isotherm model.

Discussion

As seen in Figure 2A, before the uptake of CR dye, the surface morphology of adsorbent had irregular cavities

Table 1. Calculated kinetic parameters of Congo red (CR) dye sorption onto AV-AC

Adsorbate	Pseudo-first-order			Pseudo-second-order			Intraparticle diffusion		
	q_e (mg/g)	K_1 (1/min)	R^2	q_e (mg/g)	K_2 (g/mg.min)	R^2	C (mg/g)	K (mg/g min ^{1/2})	R^2
Congo red	1.03	2×10^{-4}	0.84	100	2.4×10^{-3}	0.99	2.52	8×10^{-2}	0.97

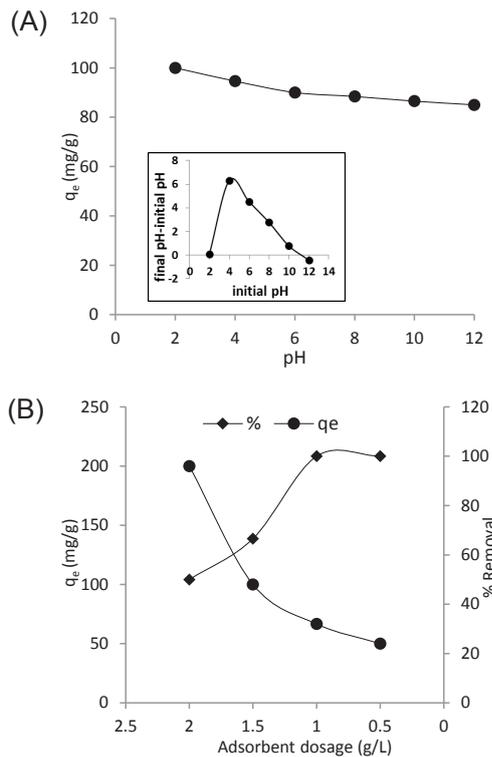


Figure 4. (A) The effect of pH on the uptake of CR dye by AV-AC (contact time=20 min, CR solution con.=100 mg/L and activated carbon dosage=1 g/L) and (B) the effect of adsorbent dosage on the sorption (contact time=20 min, CR solution con.=100 mg/L and pH=2).

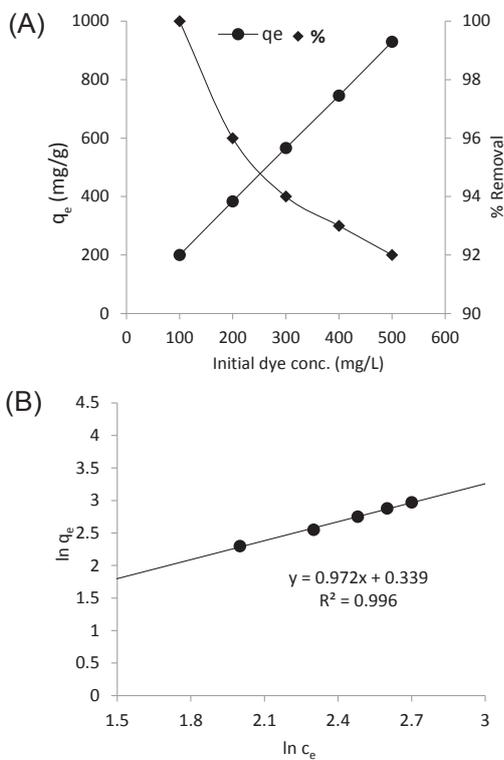


Figure 5. (A) The effect of initial concentration on the sorption of CR dye by AV-AC (contact time=20 min, pH=2 and activated carbon dosage=0.5 g/L) and (B) Freundlich isotherm model.

via fine open holes. After adsorption, as can be seen in Figure 2B, a smoother configuration, regular surface, and expanded pores were formed, after the uptake of adsorbate onto activated carbon (3). The expansion of the pores can be because of the uptake of CR onto the sorbent surface that filled these pores.

The contact time of the uptake process is an important parameter in the context of the economic aspects of the treatment of polluted water (3). The plot of contact time illustrates that the sorption of CR dye was rapidly increased at the first of the uptake process due to the availability of more vacant sites at the adsorbent surface and, with increases in contact time, these vacant sites were filled by CR. Thus, adsorption capacity was gradually increased. The equilibrium was acquired at contact time of 20 minutes ($q_e = 91 \text{ mg/g}$) and then slowly reached a fixed state during the residual time up to 100 minutes. Thus, the contact time of 20 minutes was chosen as the optimum for the subsequent stages.

Table 3 illustrates the uptake capacity and equilibrium time of CR uptake by various adsorbents. As can be seen, the comparison of low-cost adsorbent (AV-AC) with other sorbents of CR illustrates that this adsorbent has a high uptake capacity (91.0 mg/g) and its sorption is approximately 3 to 5 times higher than that of chitosan (19.15 mg/g), valoria bryopsis (10.52 mg/g), and pinus brutia (35.3 mg/g), obtained from other studies. This may be due to the unique characteristics of AV-AC (2).

The pseudo-first-order kinetic is expressed by Eq. (3):

$$\ln(q_e - q_t) = \ln q_e - K_1 t \quad (3)$$

where q_e (mg/g) and q_t (mg/g) are respectively the amounts of CR dye adsorbed onto the AV-AC surface at the equilibrium and at t (min). The parameters of K_1 (uptake rate constant, 1/min) and q_e were determined from linear plotting of $\ln(q_e - q_t)$ against t (min), which were acquired from the gradient and cut-off of the plot respectively (2). The experimental data of the CR adsorption were also analysed by the pseudo-second-order kinetic. This sorption model can be expressed by the Eq. (4):

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

where q_e and q_t have similar definitions to the parameters of pseudo-first-order kinetic mentioned above. Kinetic rate constant (K_2 , g/mg.min) and adsorption capacity at equilibrium state (q_e , mg/g) were obtained from the intercept and slope of t/q_t against t (min) respectively. The intraparticle diffusion model is explained by Eq. (5):

$$q_t = k_{id} \sqrt{t} + C \quad (5)$$

where K_{id} (g/mg.min^{1/2}) is the kinetic model constant rate and C (mg/g) is associated with the thickness of the boundary layer. The amounts of C and K_{id} are achieved from the intercept and slope of the liner plot of q_t versus \sqrt{t} respectively.

Cheng et al illustrated the pseudo-second-order kinetic well followed the uptake of CR dye via activated carbon/

Table 2. Calculated isotherm parameters of Congo red (CR) dye sorption onto AV-AC

Adsorbate	Langmuir			Freundlich		
	Q_m (mg/g)	b (L/mg)	R^2	K_f	n	R^2
Congo red	1850	8.5×10^{-4}	0.96	4.6×10^{-2}	1.02	0.99

Table 3. Comparison of various adsorbents in respect of sorption of Congo red (CR) dye from aqueous media

Adsorbent	C_0 (mg/L)	Equilibrium time (min)	q_e (mg/g)	Ref.
EBTAC	50	22h	21.73	(22)
CB-CTAB	20	4h	19.15	(25)
Valoria bryopsis	30	180	10.52	(26)
Pinus brutia	100	120	35.3	(27)
Maghemite nanoparticles	20	30	20.0	(28)
Eichhornia crassipes	104.45	90	4.97	(29)
PPy-PANI NFs	50	120	50.50	(30)
AV-AC	100	20	91.0	This study

surfactant than other kinetics (5). In addition, according to the amount of C (mg/g) for the uptake of CR, the liner plot of this kinetic model did not pass through the origin. Thus, the intraparticle diffusion model is not the step of rate limiting in the adsorption of CR dye by AV-AC.

The solution pH changes the surface properties of the adsorbent and the level of adsorbate ionization (2,3). Furthermore, the uptake of adsorbate is affected by the solution pH due to the sorption of H^+ and OH^- ions in the liquid (10,25). This outcome can be due to the change in the surface charge of the activated carbon. The amounts of positive charges onto the adsorbent surface were enhanced through decrease in initial solution pH, subsequently causing increase in the sorption capacity in more acidic solution pH. The electrostatic repulsions between the negatively charged AV-AC and anionic dye of CR were increased in the basic pH.

It was identified that with the increase in the AV-AC dosage from 0.5 to 2 g/L, the adsorption capacity was reduced from 200 to 50 mg/g. Decrease in the sorption amounts of upper dosages may be due to the inaccessibility of the dye molecules that cannot cover all the surface sites of the adsorbent. In other words, a huge number of the active sites on the AV-AC surface cannot attain saturation state at higher dosages of the activated carbon. So, dose of 0.5 g/L of AV-AC was chosen as the optimum dose for the subsequent stages of the sorption process.

The quantity of CR adsorbed was incremented from 200 to 929.88 mg/g by the increase in the adsorbate content from 100 to 500 mg/L. This finding can be due to the fact that the adsorbed pollutant created van der Waal's driving force to overcome the mass transmission resistance of CR dye to the active surface sites of the AV-AC. Thus, the above-mentioned power increase through an increase in the adsorbate content, therefore, caused an enhancement of CR sorption capacity.

Adsorption isotherms provide helpful requirements to identify the uptake mechanism and characteristics of the sorbent surface for the design of sorption systems (26,27). The Langmuir model assumes that the monolayer uptake

occurred onto the homogeneous surface of the AV-AC. This model is widely used to the uptake of pollutants from aqueous media. The Langmuir isotherm model is exhibited by Eq. (6):

$$\frac{C_e}{q_e} = \frac{C_e}{Q_m} + \frac{1}{bQ_m} \quad (6)$$

where q_e (mg/g) and C_e (mg/L) are the uptake capacity of the adsorbent and initial CR concentration at equilibrium time respectively. The values of Q_m (mg/g, the maximum uptake capacity of monolayer sorbent) and b (L/mg, constant rate of Langmuir model) were obtained from the gradient and cut-off of liner plotting of C_e/q_e versus C_e respectively. The Freundlich isotherm model is generally used for multilayer adsorption on heterogeneous surface of the adsorbent. This uptake model can be illustrated by the following equation:

$$\ln q_e = \ln k_f + \frac{1}{n} \ln C_e \quad (7)$$

where K_f (L/g) and n are the constant rates of isotherm model respectively associated with the uptake capacity and adsorption intensity.

It can be found that the experimental data of CR dye onto AV-AC were well-fitted to the Freundlich isotherm model. In general, the adsorption bonds between activated carbon and adsorbed CR dye were suitably powerful, since the n value achieved from the Freundlich model was further than that one. Rouf et al showed that the experimental data of CR dye by chitosan were well followed through the Freundlich isotherm model (28).

Comparison with other studies

The amount of maximum monolayer uptake capacity of CR dye by AV-AC was obtained at 1850 mg/g. This amount has been compared with Q_m achieved from the other studies for the removal of CR. Table 4 shows the maximum uptake capacity from the Langmuir model by various sorbents. It can be noted that the maximum of CR uptake capacity of the AV-AC is noticeably further than

Table 4. Comparison of the maximum uptake capacity of AV-AC with that of other adsorbents

Adsorbents	Q _m (mg/g)	Ref.
Eichhornia crassipes	1.58	(29)
Activated carbon (laboratory grade)	1.88	(34)
Cashew nut shell	5.18	(35)
Kaolin (clay materials)	5.44	(36)
Activated carbon prepared from coir pith	6.70	(37)
Acid activated red mud	7.08	(20)
Bagasse fly ash	11.89	(34)
Montmorillonite	12.70	(14)
Cattail root	38.79	(9)
Ball-milled sugarcane bagasse	39.8	(23)
Neem leaf powder	41.20	(38)
Magnetic core–manganese oxide shell	42.0	(39)
Chitosan/montmorillonite nanocomposite	54.52	(14)
Chitosan beads modified with CTAB	94.39	(25)
Ca-bentonite	107.41	(40)
Mesoporous activated carbon	189.0	(41)
Maghemite nanoparticles	208.33	(28)
Cone of Pinus brutia biomass	329.6	(27)
CTAB modified chitosan beads	352.2	(42)
AC/DDAC	769.23	(5)
AV-AC	1850	This study

the maximum capacity of other sorbents.

Conclusion

In this study, CR was removed from aqueous media by Aloe vera leaf shells-based activated carbon (AV-AC). The influence of different parameters like contact time, pH, adsorbent dosage, and CR content were evaluated on the uptake capacity of the adsorbent. The optimum uptake capacity of the AV-AC (91.0 mg/g) was achieved within 20 minutes at pH of 2. The maximum monolayer adsorption capacity was obtained equal to 1850 mg/g. The results showed that the experimental data were well-followed from the second-order-kinetic and Freundlich isotherm models. This natural adsorbent has several advantages, such as low cost, environmental friendliness, high uptake capacity, and non-toxicity. Thus, it can be considered an effective adsorbent on the removal of CR dye from aqueous solutions.

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Ethical issues

The authors certify that all data collected during the study are presented in this manuscript, and no data from the study has been or will be published separately.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

Study concept, design, and critical revision of the

manuscript for important intellectual content: YOK; drafting of the manuscript and advisor, and performing experiments: YOK, HB, MJM, MS, ShS, and SeS.

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