

Oxidized jute as a valuable adsorbent for Congo Red from an aqueous solution

Journal of Engineered Fibers and Fabrics

Volume 17: 1–9

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DOI: 10.1177/15589250221101380

journals.sagepub.com/home/jeff



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Abstract

The raw jute fabric was oxidized with 0.2% or 0.4% NaIO₄ for 60 or 120 min to obtain valuable adsorbents for the textile dye Congo Red (CR). Batch adsorption experiments were carried out as a function of solution pH, contact time, initial CR concentration, and temperature. At an initial CR concentration of 25 mg/L, the raw jute possessed the lowest adsorption due to the lowest availability and content of carboxyl and aldehyde groups capable of binding dye. The fabrics oxidized for 60 min with 0.2% NaIO₄ and 120 min with 0.4% NaIO₄ reached equilibrium adsorption after 240 and 330 min, respectively. The adsorption of CR onto studied fabrics followed the pseudo-second-order model indicating that the chemisorption process is primarily represented. Within the studied range of CR concentrations (25–100 mg/L), its adsorption onto fabric oxidized for 120 min with 0.4% NaIO₄ obeys the Langmuir isotherm model (i.e. adsorption occurs at specific, energetically equivalent adsorbent sites with monolayer coverage of CR over a homogeneous surface), wherein the calculated maximal adsorption capacity is 12.863 mg/g. A thermodynamic study revealed the spontaneous and endothermic character of CR adsorption onto jute fabric. Based on the recorded ATR-FTIR spectra before and after CR adsorption, it is suggested that repulsion, hydrogen bonds, and π - π stacking interactions are involved in the binding of CR onto jute fabrics.

Keywords

Jute, oxidation, adsorbent, Congo Red, adsorption mechanism

Date received: 3 March 2022; accepted: 2 May 2022

Introduction

Synthetic dyes are widely used in many different industries such as textile, paper, leather, wood, plastic, cement, glass, *etc.* which results in their release into nearby streams and rivers.¹ Among different industries using synthetic dyes, the textile industry is considered the most polluting due to the production of significantly high quantities of colored effluents. For example, effluents from dyeing cellulosic fibers contain between 5% and 30% residual direct dye.² Besides the intensive color, such effluents contain solids and salinity, have variable pH, temperature, and chemical oxygen demand (COD).³ Between different classes of dyes, anthraquinone, indigoid, *etc.*, azo dyes, having one or more azo groups (–N=N–) in their chemical structure, are one of

the most important synthetic dyes produced worldwide.⁴ Apart from the fact that the discharge of effluents containing azo dyes causes environmental pollution, their degradation by-products have a toxic and mutagenic impact on

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aquatic organisms. Moreover, they could aggravate photosynthesis by disabling the adsorption of sunlight, thus affecting the ecological balance of water bodies.⁵

All the above-discussed results in considerable attention on the methods used for decoloration of azo dye-contaminated effluents. Ozonation, sonication, flocculation, oxidation and electrolysis, ion exchange, and membrane filtration possess several drawbacks such as high operating energy and costs, difficulties in operation, and formation of sludge (i.e. pollutant transformation from one to another phase resulting in a new kind of pollution). Since magnetic nanoparticles have a large specific surface area and an ultra-high surface energy, they are used as adsorbents for dyes.^{6,7} Nowadays, many researchers are focusing on adsorption as the simplest, most effective, and economical way to decoloration of textile effluents through the utilization of different adsorbents from waste sources.⁸

It has to be noted that as a result of the growing demand for both goods and services, the production of solid waste rise day by day and becoming a major environmental issue that implies the need for its cost-effective disposal. Namely, jute cultivation has risen steadily over the years hitting 2.69 million t in 2020.⁹ Consequently, the production of packaging (sackings, hessian cloth, *etc.*) and other diverse textiles (carpet backings, home textiles) generates a considerable amount of solid waste in the form of fabric.¹⁰

In the light of all above mentioned, this study is focused on finding valuable adsorbents for water-soluble and widely used azo dye Congo Red (CR). The removal of this dye by adsorption is highly preferable over the other methods since its degradation leads to the production of carcinogenic amines.¹¹ Periodate oxidation lead to the conversion of cellulose hydroxyl groups on C2 and C3 atoms to aldehydes, wherein the ring cleavage occurred and 2,3-dialdehyde cellulose was formed.¹² In the current investigation, the waste jute fabric was oxidized with sodium periodate under different experimental conditions in order to obtain more efficient adsorbents for CR.

To the best of our knowledge, the adsorption of CR was previously studied onto differently modified lignocellulosic-based materials, whereby the applied modifications are too complicated and cost-effective. Moreover, some of them included treatments with strong reduction agents or pyrolysis at high temperatures raising the question regarding the adsorbent cost-effectiveness. Besides the fact that a very simple oxidation protocol (in mild conditions) was used in the current study, the fast regeneration of sodium periodate solution after the treatment is possible.¹³

Experimental

Materials

To obtain jute fabrics with high adsorption efficiency for CR, the raw jute fabric (J) was oxidized with 0.2 or 0.4%

Table 1. Adsorbents' labels and oxidation conditions.

Fabric labels	NaIO ₄ concentration, %	Oxidation time, min
J	/	/
J0.2/60	0.2	60
J0.2/120		120
J0.4/60	0.4	60
J0.4/120		120

sodium periodate (NaIO₄) solution (material to liquid ratio of 1:50) for 60 or 120 min. First of all, appropriate amounts of NaIO₄ were dissolved in acetic buffer (pH 4.0). Then, the raw jute fabric samples were immersed in the obtained NaIO₄ solutions with a constant stirring in the dark at room temperature. When the oxidation was completed, the jute fabrics were washed with ice-cold distilled water several times to remove the oxidant and dried at room temperature for 72 h. Their labels along with the oxidation conditions are listed in Table 1.

Adsorption of CR

The adsorption experiments were carried out by varying the solution pH, contact time, initial CR concentration, and temperature, while the obtained results were modeled with widely used kinetic and isotherm models.

About 0.5 g of jute fabric were immersed in 100 mL of freshly prepared CR solution at a pH of 10.00 containing 15 g/L NaCl and constantly shaken. The detailed discussion regarding the optimization of solution pH is given in the Supplemental Material, section 1. The concentration of CR in aqueous solution was determined based on the UV-Vis (Shimadzu 1700 spectrophotometer) absorbance spectra at $\lambda_{\max} = 486$ nm. The mass of dye adsorbed per gram adsorbent (mg/g) was calculated according to equation (1).

$$q = \frac{c_0 - c_t}{m} \cdot V \quad (1)$$

where: c_0 and c_t (mg/L) are the CR concentrations in the aqueous solution before and after a defined period of adsorption, respectively, V (L) is solution volume, and m (g) is the fabric mass.

Adsorption kinetics. The adsorption kinetic experiments were performed at an initial CR concentration of 25 mg/L for up to 420 min at a temperature of 25°C. The pseudo-first¹⁴ (equation (2)) and pseudo-second-order¹⁵ (equation (3)) were used to investigate the adsorption mechanism of CR onto jute fabrics.

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (2)$$

$$\frac{t}{q_t} = \frac{1}{k_2 \cdot q_e^2} + \left(\frac{1}{q_e} \right) \cdot t \quad (3)$$

where: q_e and q_t (mg/g) are the amounts of CR adsorbed per gram adsorbent at equilibrium and at time t (min), respectively, k_1 (1/min) is a pseudo-first-order rate constant, k_2 (g/mgmin) is a pseudo-second-order rate constant.

Adsorption isotherms. The isotherm experiments were carried out by varying the initial concentration of CR (25, 50, 75, and 100 mg/L), while the contact time of 330 min and temperature of 25°C remained constant. Langmuir¹⁶ (equation (4)) and Freundlich¹⁷ (equation (5)) isotherm models were used for modeling the equilibrium adsorption data and determination of the maximal adsorption capacity of jute fabrics.

$$\frac{C_e}{q_e} = \left(\frac{1}{q_m \cdot K_L} \right) + \left(\frac{1}{q_m} \right) \cdot C_e \quad (4)$$

$$\ln q_e = \ln K_f + \frac{1}{n} \cdot \ln C_e \quad (5)$$

where: q_e and q_m (mg/g) are equilibrium and a maximal adsorbed CR per gram adsorbent, respectively, K_L (L/mg) is Langmuir constant, C_e (mg/L) is equilibrium CR concentration in the solution, K_f is Freundlich constant (mg/g) (L/mg)^{-1/n}, $1/n$ is constant related to the fabric surface heterogeneity.

Thermodynamic parameters of adsorption. The thermodynamic experiments were performed by varying the temperature (25°C, 35°C, and 45°C), while the contact time of 330 min and initial CR concentration of 25 mg/L keep constant. Based on the obtained results, the thermodynamic parameters standard Gibbs free energy (ΔG°), standard enthalpy (ΔH°), and standard entropy (ΔS°) were calculated using equations (6) and (7).

$$\Delta G^\circ = -R \cdot T \cdot \ln(K_{eq}) \quad (6)$$

$$\Delta G^\circ = \Delta H^\circ - T \cdot \Delta S^\circ \quad (7)$$

where: R is the universal gas constant (8.314 J/mol K), T is the process temperature (K), K_{eq} is the process equilibrium constant calculated as the ratio between the amount of CR adsorbed q_e (mg/g) and the residual CR concentration in the solution C_e (mg/mL) at the equilibrium (equation (8)).

$$K_{eq} = \frac{q_e}{C_e} \quad (8)$$

The expression of the equilibrium constant quantifies the distribution of the pollutant between the solution and the adsorbed phase. By combining equations (6) and (7), the equation (9) was obtained:

$$\ln(K_{eq}) = -\frac{\Delta H^\circ}{RT} + \frac{\Delta S^\circ}{R} \quad (9)$$

The K_{eq} and ΔG° were calculated for each studied temperature, while the values of ΔH° and ΔS° were estimated from the slope and intercept of $\ln(K_{eq})$ versus $1/T$ plot, respectively.

Contents of carboxyl and aldehyde groups and characterization of fabric surface chemistry

The contents of carboxyl and aldehyde groups were determined using the calcium-acetate method previously described by Ivanovska et al.¹⁸ A sample (0.5 g) was treated with 100 mL of 0.01 M HCl solution for 1 h followed by washing with distilled water. In the next step, 50 mL of distilled water and 30 mL of 0.25 M CH₃COOCa solution were added and the mixture was stirred for 2 h. Thereafter, 30 mL portions of the liquid were titrated with 0.01 M NaOH solution, using phenolphthalein as an indicator. The content of carboxyl groups was calculated according to equation (10):

$$COOH = \frac{80}{30} \cdot 0,01M \cdot V(NaOH) \left[\frac{\text{mmol}}{\text{g}} \text{ jute} \right] \quad (10)$$

where: 0.01 M is a concentration of NaOH, $V(NaOH)$ is the volume of NaOH solution used for titration (mL), and m is the weight of an absolutely dry wood waste sample (g).

In order to selectively oxidize the fabric aldehyde groups to carboxyl groups, 1 g of a sample was added to a mixture containing 0.905 g of NaClO₂, 10 mL of 5 M CH₃COOH solution, and 50 mL of distilled water. Oxidation was carried out by mixture stirring at room temperature for 48 h, and thereafter, the fabric was thoroughly washed with distilled water and acetone. Further, the previously described calcium-acetate method for determining the content of carboxyl groups was applied. The content of aldehyde groups was calculated by subtracting the content of carboxyl groups determined in the starting sample from that of the chlorite oxidized one.

The ATR-FTIR spectroscopy was employed to characterize the jute fabrics and to identify the interaction between them and CR. The transmittance spectra were obtained using Nicolet™ iS™ 10 FT-IR (Thermo Fisher 2 SCIENTIFIC) spectrometer with Smart iTR™ Attenuated Total Reflectance (ATR) Sampling accessory. The spectra were recorded in the range of 4000–450 cm⁻¹ with 32 scans per spectrum.

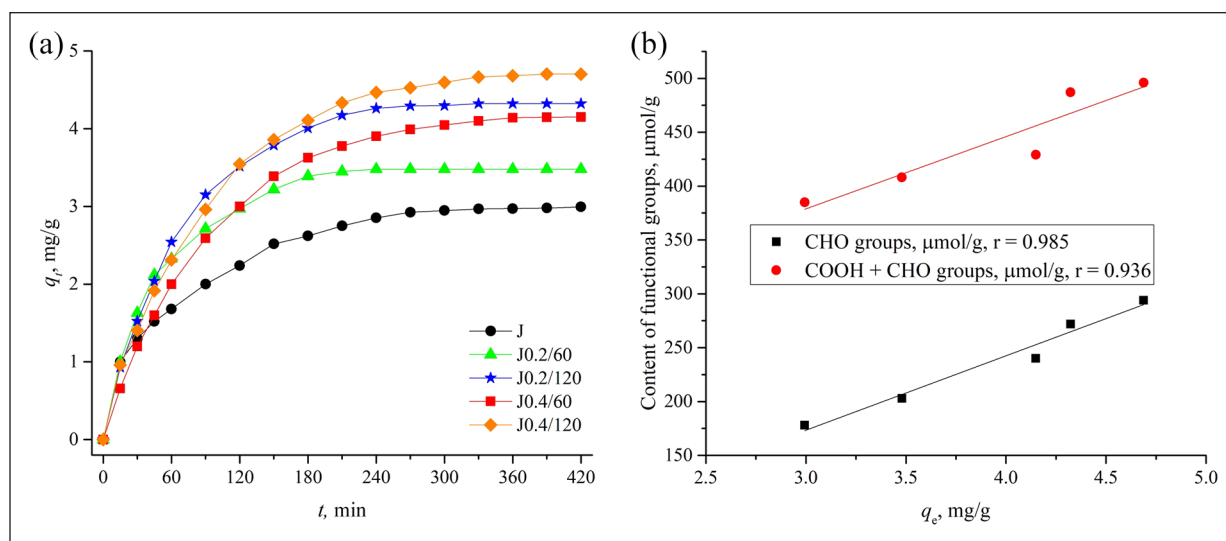


Figure 1. (a) Adsorption of CR (q_t) onto jute fabrics as a function of contact time (t) and (b) linear correlation between q_e and contents of carboxyl (COOH) and aldehyde (CHO) groups.

Results and discussion

The influence of contact time on the adsorption of CR

The necessity to investigate the effect of contact time on the adsorption of CR comes from the economic point of view, which requires the most effective dye adsorption for the shortest time. In this study, the kinetic experiments were carried out by varying the contact time from 15 up to 420 min while remaining the initial solution pH, initial CR concentration, and temperature constant (i.e. 10.00, 25 mg/L, and 25°C, respectively). The results given in Figure 1(a) showed that the lowest equilibrium adsorption capacity (2.99 mg/g) possessed untreated jute fabric (J) which could be ascribed to the lowest availability and content of its functional groups (Figure 1(b)) capable of binding CR. Among oxidized fabrics, J0.2/60 treated under the mildest conditions (0.2% NaIO₄ for 60 min) reached equilibrium adsorption of 3.48 mg/g after 240 min of contact time. Furthermore, 90 min longer adsorption time resulted in the highest adsorption potential (4.66 mg/g) of fabric J0.4/120 due to the highest contents of functional groups, Figure 1. The last one was also confirmed by the ATR-FTIR spectroscopy, Supplemental Material, section 2.

The data presented in Figure 1(a) were further modeled using pseudo-first and pseudo-second-order kinetic models to evaluate the rate of CR uptake from an aqueous solution, which controls the mechanism of dye adsorption. According to the plots and R^2 values (Figure 2), the adsorption of CR onto all studied jute fabrics undoubtedly followed the pseudo-second-order model ($R^2 \geq 0.995$) indicating that the chemisorption process is primarily represented.¹⁹

Panda et al.²⁰ reported that in batch experiments under rapid stirring conditions, transportation of CR from the

solution phase into the adsorbent pores may also be considered a rate-controlling stage. In the current study, fabrics oxidized with 0.4% NaIO₄ have higher adsorption rates than those oxidized with 0.2% NaIO₄, which is clearly seen from their lower pseudo-second constant values (k_2 , Table 2).

In the already published data, the adsorptions of CR onto NaOH,^{21,22} and tannin modified²³ jute fibers as well as onto jute stick powder²⁰ were also best fitted with the pseudo-second-order model.

Effect of initial CR concentration on the fabric adsorption efficiency

The initial dye concentration in the solution plays a pivotal part in the content of dye adsorbed by the adsorbent. The adsorption capacities of oxidized jute fabrics for CR present at different initial concentrations (25, 50, 75, and 100 mg/L) were investigated after a contact time of 330 min at 25°C. From the results presented in Figure 3, it is evident that with increasing the initial concentration of CR from 25 to 100 mg/L, the equilibrium adsorption (q_e) increased by about 1.5–2.6 times. This behavior indicated that the driving force for CR adsorption is the gradient of concentration between the aqueous phase and the solid one, which overcomes the resistance to mass transfer between the two phases; the higher the gradient, the more favored the process.²⁴

Keeping in mind that, independently on initial concentration, fabric J0.4/120 possessed the highest adsorption capacity for CR (Figure 3), the equilibrium between the CR, which gets adsorbed on the active sites of J0.4/120, and the one that remains in the aqueous solution was modeled by Langmuir and Freundlich isotherms, Figure 4.

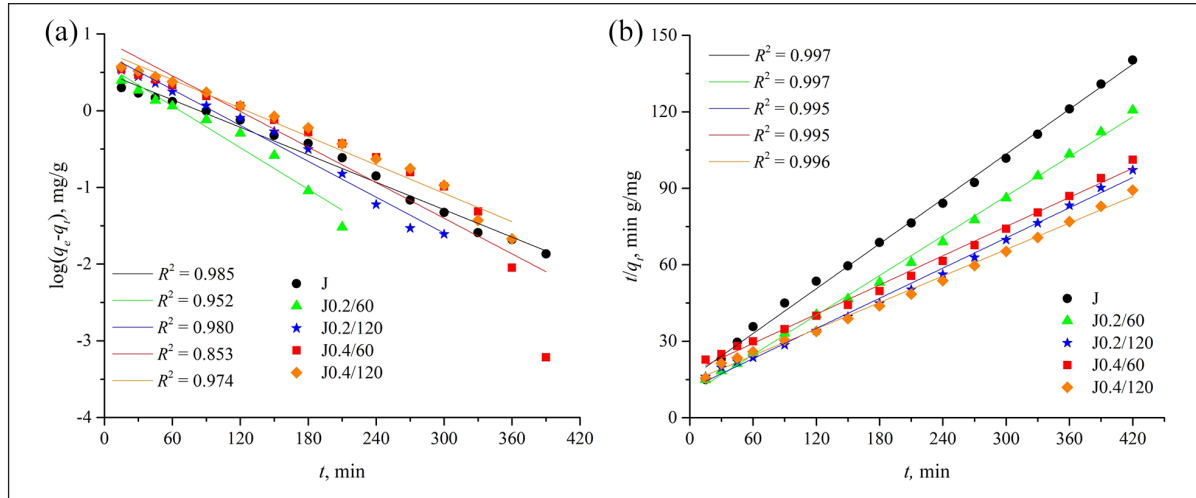


Figure 2. Linear plots of the (a) pseudo-first and (b) pseudo-second-order kinetics for the adsorption of CR onto jute fabrics and the corresponding coefficients of determination (R^2).

Table 2. Kinetic parameters obtained by the pseudo-first and pseudo-second-order kinetic models for CR ($c_0 = 25$ mg/L) adsorption onto jute fabrics.

Kinetic models	Jute fabrics					
	Kinetic parameters	J	J0.2/60	J0.2/120	J0.4/60	J0.4/120
Pseudo-first	$q_{e,cal}$, mg/g	3.160	4.037	5.553	8.318	5.956
	k_1 , 1/min	0.014	0.021	0.018	0.018	0.014
Pseudo-second	$q_{e,cal}$, mg/g	3.409	3.857	5.070	5.241	5.772
	k_2 , g/mgmin	0.006	0.007	0.003	0.002	0.002

These commonly used isotherms are of fundamental importance for adsorption system designing for practical applications. The linearity of the c_e/q_e versus c_e plot along with a very high coefficient of determination ($R^2=0.998$) prove that the CR adsorption onto J0.4/120 obeys the Langmuir isotherm model. According to this model, the adsorption occurs at specific, energetically equivalent sites of the J0.4/120 with monolayer coverage of the CR over a homogeneous fabric surface.^{21,23} The values of maximal adsorption capacity (q_m) and Langmuir constant (K_L) were obtained from the slope and intercept of linear Langmuir isotherm and presented in Figure 4(a). Namely, the theoretical monolayer saturation capacity of fabric J0.4/120 is 12.863 mg/g.

Similarly, the isotherm adsorption data of CR onto NaOH²¹ and tannin modified²³ jute fibers and jute stick powder²⁰ obeys the Langmuir model.

Thermodynamic studies

It is well known that adsorption experimental parameters such as solution pH, time, pollutant concentration, and temperature significantly affect the overall adsorption process. Since the effect of the first three parameters on the CR adsorption was already discussed, in this section, the adsorption experiments for J0.4/120 were conducted at

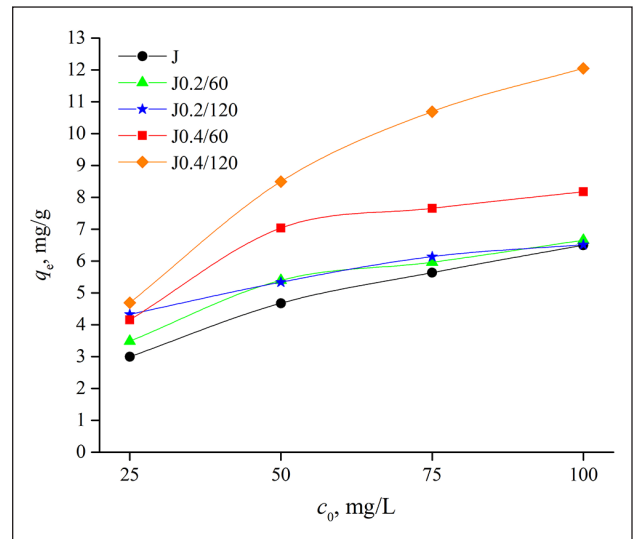


Figure 3. Equilibrium adsorption (q_e) as a function of initial concentration (c_0) of CR.

three different temperatures (25°C, 35°C, and 45°C, i.e. 298.15, 308.15, and 318.15 K), while keeping the contact time of 330 min and initial CR concentration of 25 mg/L constant. The results presented in Figure 5 pointed out that

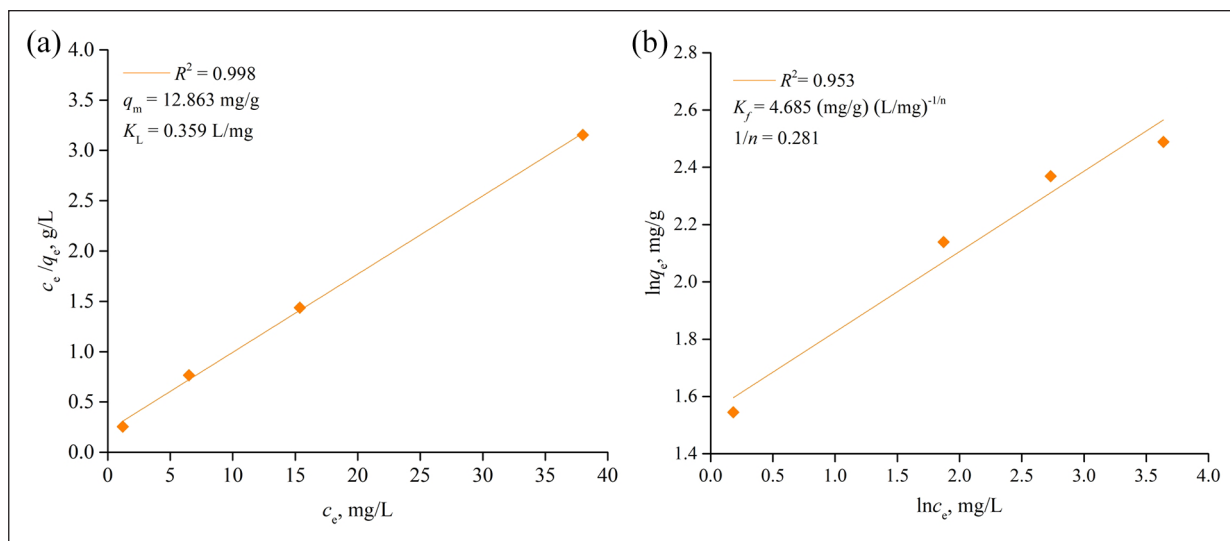


Figure 4. (a) Langmuir and (b) Freundlich adsorption isotherms and the linear fit of the experimental data for CR adsorption onto jute fabrics.

with increasing the temperature from 25 to 35, the adsorption of CR remained almost unchanged. However, with further increasing the temperature to 45°C, the CR adsorption onto fabric J0.4/120 increased by about 10%.

Data obtained from thermodynamic adsorption experiments (Figure 5) were used to calculate the main thermodynamic parameters (i.e. standard Gibbs free energy, standard enthalpy ΔH° , and standard entropy ΔS°) of CR adsorption onto J0.4/120. The positive $\ln K_{eq}$ values in a combination with the negative ΔG° and positive ΔS° calculated for all considered temperatures, indicated that the CR adsorption onto selected fabric is spontaneous.²⁵ Moreover, the positive ΔH° values prove that the adsorption process is of endothermic character.

Adsorption mechanism

Having in mind the previously discussed facts that oxidized jute fabrics possessed better adsorption capacities with respect to untreated and that the highest capacity possessed fabric J0.4/120 (the longest oxidation with a higher NaIO_4 concentration), it is clear that aldehyde and carboxyl groups play a significant role in the adsorption of CR onto oxidized jute fabrics, Figure 1. In order to reveal the adsorption mechanism, the ATR-FTIR spectra of J0.4/120 before and after adsorption of CR (c_0 of 50 and 100 mg/L) were considered, Figure 5. Namely, upon CR adsorption on the jute fabric, the intensities of all bands decreased whereas higher dye concentration induced a larger decrement. A band at 3331 cm^{-1} ascribed to non-oxidized OH stretching vibrations loses sharpness after adsorption of CR indicating the involvement of these groups in the binding. On the other hand, stretching vibrations of C-H groups located at 2917 cm^{-1} are broadening, while the bands at 2850 cm^{-1}

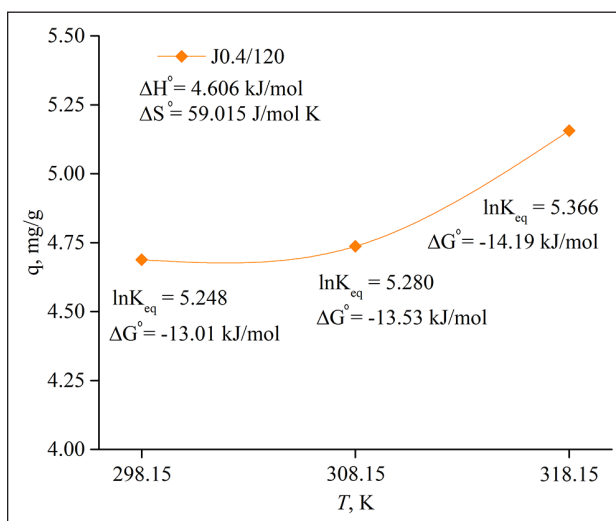


Figure 5. The effect of temperature (T) on the CR adsorption (q) onto J0.4/120.

(ascribed to dominant C-H vibrations of aldehyde groups) and 1639 cm^{-1} (corresponding to C=C aromatic vibrations)²⁶ disappear after CR adsorption.

Taking into account that in the performed adsorption experiments, the pH value of CR solution was adjusted to 10.0, it is clear that both CR and jute fabric surface are negatively charged. Namely, the negative charge of the oxidized jute originates from carboxylate groups that repulse negatively charged CR sulfonate groups, dictating the orientation of the molecule whereby the dye sulfonate groups are oriented away from the fabric surface. In such a way, amino groups are directed toward the fabric surface and can form hydrogen bonds with its surface functional

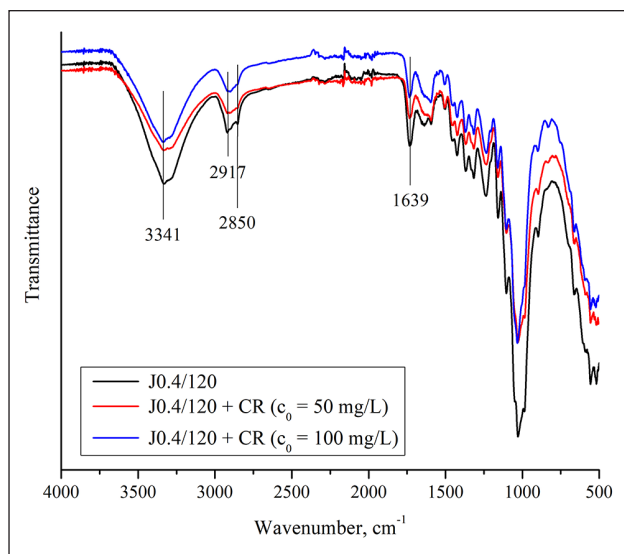


Figure 6. ATR-FTIR spectra of J0.4/120 before and after adsorption of CR.

groups (aldehyde, carboxylate, and hydroxyl), Figure 6. Due to the peculiarity of the CR molecule and its high propensity to form hydrophobic interactions via aromatic rings,^{27–29} it is reasonable to assume that the established hydrogen bonds are strengthened through the π - π stacking interactions with the lignin aromatic rings, Figure 7.⁸ Also, the hydrogen bonds between CR azo groups and remaining OH groups unsusceptible to oxidation, should not be neglected in an overall putative binding mechanism. Moreover, the reason behind the better adsorption of the oxidized jute fabrics compared to the untreated may be the higher availability and flexibility of the aldehyde groups caused by the ring cleavage that occurred due to the sodium periodate oxidation.

Comparison of the adsorption capacity of jute with those of lignocellulosic fiber-based adsorbents

Table 3 compares the adsorption capacities of jute fabrics for CR with the previously reported efficiencies of lignocellulosic fiber-based adsorbents. After careful consideration of the listed results, it can be concluded that the adsorption capacities of J0.4/60 and J0.4/120 are comparable, or somewhat higher than those obtained for tannin-modified jute fibers,²³ jute processing waste,³⁰ and jute stick powder.²⁰ However, the complicated and long procedure (8.5 h) comprising treatment with a strong reduction agent (NaBH_4) for the preparation of tannin-modified jute fibers should not be neglected.

Especially higher adsorption capacity (33.69 mg/g) of NaOH modified jute fibers²² compared to J0.4/120 could

be explained by about three times higher adsorbent dosage as well as initial concentration of CR. Furthermore, Mandal et al.³² prepared kenaf-based activated carbon by pyrolysis at high temperature, that is, 1000°C for 2 h, which, from the economic point of view makes this adsorbent incomparable with those of the current study, Table 3.

Conclusion

The study proposed a simple route for obtaining valuable adsorbents for the textile dye Congo Red (CR). Namely, oxidation of raw jute fabric with 0.2% or 0.4% NaIO_4 for 60 or 120 min provides sufficient availability and content of carboxyl and aldehyde groups necessary for CR adsorption. The higher availability and content of such groups promote the higher adsorption capacities of oxidized fabrics. The obtained results demonstrated that at an initial CR concentration of 25 mg/L, the fabrics oxidized for 60 min with 0.2% NaIO_4 and 120 min with 0.4% NaIO_4 reached equilibrium adsorption after 240 and 330 min, respectively. The adsorption of CR onto studied jute fabrics followed the pseudo-second-order model pointing out that the chemisorption process is primarily represented. With increasing the initial concentration of CR from 25 up to 100 mg/L, the equilibrium adsorption increased by about 1.5–2.6 times. Within the studied range of CR concentrations, its adsorption onto fabric oxidized for 120 min with 0.4% NaIO_4 obeys the Langmuir isotherm model, that is, the adsorption occurs at specific, energetically equivalent sites of the J0.4/120 with monolayer coverage of the CR over a homogeneous fabric surface. The calculated maximal adsorption capacity of the mentioned fabric is 12.863 mg/g, which is comparable, or somewhat higher than those of other lignocellulosic fiber-based adsorbents reported in the literature. A thermodynamic study revealed the spontaneous and endothermic character of CR adsorption onto jute. The proposed adsorption mechanism suggests that the repulsion between fabric carboxylate and CR sulfonate groups dictates the orientation of the dye molecules allowing amino groups to form hydrogen bonds with fabric surface functional groups. The adsorption is reinforced by stacking π - π interactions between CR aromatic rings and lignin aromatic moieties. The lifecycle of oxidized jute fabrics with adsorbed CR could be extended by their further utilization as home textiles (i.e. decorative fabrics or carpet backing), for the production of bio-based composites which will be used as building materials, or carbonized and used as a great alternative to graphene and lithium batteries.

Declaration of conflicting interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

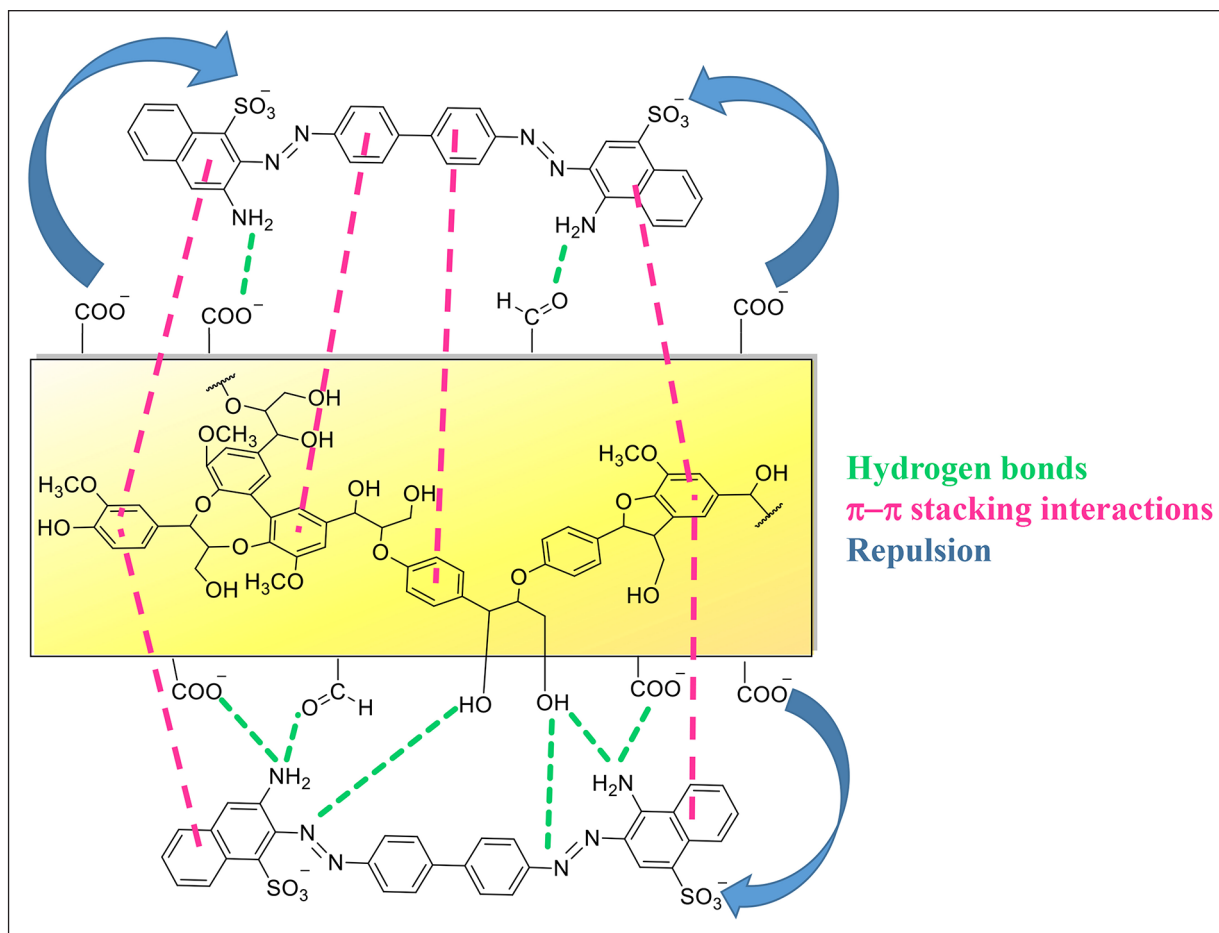


Figure 7. Proposed mechanism of CR adsorption.

Table 3. Comparison between the different lignocellulosic fibers' adsorption capacities for CR and findings of the current study.

Lignocellulosic fiber-based adsorbent	Adsorbent dosage, g/L	pH	Contact time, min	c_0 , mg/L	q_{ex} , mg/g
Na-OH modified jute fibers ²²	14.52	7.2	N/A	150	33.69
Tannin – modified jute fibers ²³	6	3	30	50	~7.5
Flax ³¹	2	6.5-6.7	80	25	~8.5
Jute stick powder ²⁰	0.05	7.0	180	N/A	~4.75
Kenaf-based activated carbon ³²	0.05	4.0	120	20	12.661
Jute processing waste ³⁰	N/A	7.8	20	50	~7
J					4.671
J0.2/60					5.393
J0.2/120	5	10.0	330	50	5.335
J0.4/60					7.033
J0.4/120					8.493

Funding

The author(s) disclosed receipt of the following financial support for the research, authorship, and/or publication of this article: This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (Contract No. 451-03-68/2022-14/200287 and 451-03-68/2022-14/200135). A.I., and M.K. would also like to acknowledge

financial support from the Science Fund of the Republic of Serbia, #7726976, Integrated Strategy for Rehabilitation of Disturbed Land Surfaces and Control of Air Pollution -RECAP.

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Supplemental material

Supplemental material for this article is available online.

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