Coagulation Study on Extracted Algal Alginate from Red Algae as Natural Coagulant for Remediation of Textile Dye Congo Red

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ABSTRACT

In the present study, extracted algal alginate from red algae as natural coagulant was used for removal of textile dye congo red (CR) from water. In developing countries like India, only about 10% of the wastewater being generated is treated, whereas the remaining 90% is discharged into the water bodies as it stands. Color and turbidity are the most common problems in the disposal of wastewater. The removal of color is one of the key challenges in wastewater treatment. For the coagulation process, the synthetic textile wastewater samples had CR concentrations of 50, 100, 150, 200, and 250 mg/L and varying initial pH of 4, 5, and 6. Different dosages of calcium and alginate (1, 2, 3, 4, 5, and 6 g/L) were used to perform the experiments. The obtained results exhibited that the effectiveness of color removal was higher at lower pH 4, and the calcium and alginate dosages are dependent on the CR concentration of the synthetic textile wastewater. These investigations demonstrating the higher efficiency of calcium alginate as a coagulant, where maximum color removal achieved over 95.05%. Increasing alginate dosages and residence times can enhance the performance of coagulation. Dye color is often present in real wastewater and needs to be removed before being reused or discharged to the environment.

1. INTRODUCTION

A basic need for all living organisms is water, but water is unable to be used when it mixes with impurities creating wastewater caused by industrialization and increasing populations. Treatment of synthetic textile wastewater is one of the major problems faced by researchers and industries (Vijayaraghavan and Shanthakumar, 2016; Sivalingam et al., 2019; Sivalingam and Sen, 2019). Textile industries are using various dyes like anionic dyes (alizarin yellow, methyl orange and congo red) and cationic dyes (methylene blue, crystal violet and rhodamine B) for dying and printing purposes. After being applied, these dye colors are generating synthetic wastewater dye solutions that are discharged without any treatment into the environment. The traditional wastewater treatment is aerobic biodegradation which gives low removal efficiency thus the need to employ new techniques. The printing and dyeing industries use an extensive amount of azo dye congo red (CR), which is made from 1-naphthalene sulfonic acid. A high quantity of this dye molecule causes an allergic reaction or possibly causes it to change into a carcinogen similar to Benzidine. Moreover, due to its structural, chemical, and thermal stability and difficult in biodegrading property CR is a persistent substrate in wastewater (Pham et al., 2019; Ajoke et al., 2021).

Commonly, various kinds of wastewater treatments have been used to purify the water such as biological treatment, membrane separation, coagulation and flocculation, oxidation, photo-chemical, ion exchange, and adsorption (Chuah et al., 2005; Assimeddine et al., 2022). However, these processes require higher costs and energy than the coagulation/flocculation which is a quite simple, effective, and economically feasible method. Coagulation technique is as simple as adding some chemicals to the suspended particles to destabilize the solution where the surface of the colloids has been modified and converting all the particles into solid particles. Coagulants increase the size of minute particles in a solution by balancing electrical charges,

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which counteract electrostatic repelling forces that cause repellent electrical charges (destabilized particles), that are typically negative, to be neutralized.

As coagulant aids, natural or synthetic polymers as well as inorganic metals like alum, ferric chloride, lime, etc. are used. For many years alum (aluminum sulfate) has been the most widely used coagulant. The aluminum ions, Al^{3+} that form when the alum is dissolved in water, neutralize the negative charges that the colloidal particles carry and which help to destabilize them. The alum destabilizes the particles through hydrolysis making it a hydrolyzing metal salt coagulant (Nozaic et al., 2001). When the water is slowly swirled the precipitates enmesh the destabilized particles to help them settle down while the Al ions hydrolyze and create $Al(OH)_3$. The main drawbacks of metal salt coagulations are the generally large amounts of chemicals required, simultaneously huge amounts of sludge generated and substantial pH changes throughout the treatment. Another disadvantage of alum is the concern about the suspected role of aluminum in Alzheimer's disease (Huang et al., 2000; Ozacar and Sengil, 2003).

Due to some negative impacts of synthetic polymers on human health, the current research focuses on natural polymeric materials. The natural polymeric materials such as polysaccharides have been suggested because they are easily available, low cost, low molecular weight, and high shear stability. Additional advantages of this natural polyelectrolyte include safety for human health, biodegradability, and a wider effective dose range of flocculation for various colloidal suspensions (Kawamura, 1991; Palapa et al., 2021). Therefore, inorganic coagulants have been suggested to be replaced by natural organic polymers in recent years (Salehizadeh and Shojaosadati, 2001).

Water treatment uses natural coagulants that are taken from specific types of plants, such as *Prosopis Juliflora*, *Cactus latifaria*, and *Moringa oleifera*. Both high and low turbidity water responds favorably to these coagulants. Moreover, chitosan, cationic starches, and extract of cactus, have been used to remove turbidity (Pal et al., 2005; Zhang et al., 2006). These investigations show that natural coagulants are more effective at removing color than inorganic metals and synthetic polymers.

Alginate was produced in this manner from sea brown algae. This polysaccharide is organic. Monomers of α-L guluronic acid (G) and β-D mannuronic acid (M) combine to produce the polymer. The monomers create MM, MG, and GG block structures to create alginate. According to Aylin Devrimci (Devrimci et al., 2012; Ekanayake and Manage, 2022), virtual quantity of these building blocks present in alginate structure shows better performance with metal ions. Alginate is the only polysaccharide, which naturally contains carboxyl groups in each constituent residue, and possesses various abilities for functional materials (Ikeda et al., 2000). Strong gels or insoluble polymers are created by the alginate's reaction with polyvalent metal cations, particularly calcium ions. They are divided into three major classes according to their coloration: brown algae (*Phaeophyceae*), red algae (*Rhodophyceae*), and green algae (*Chlorophyceae*) (Kharkwal et al., 2012). There are 4,000 red species, 1,500 brown species, and roughly 900 kinds of green seaweed that can be found in nature. Alginate is used in a variety of products, including frozen foods, pastry fillings, syrups, bakery goods, dry mixes, meringues, frozen desserts, cooked puddings, salad dressings, flavored meat sauces, chiffons, and dessert gels. The *Chennubhotla*, *Sargassum*, and *Turbinaria* species are used to extract algin.

The main aim of this work is to study an effective coagulation process with extracted alginate as a natural coagulant for removal of highly toxic textile industry dye CR from wastewater. This study further gives sufficient details an effect of different variables such as pH, calcium dose, alginate dose, and dye concentration respectively.

2. METHODOLOGY

2.1 Algae collection

Different species of red (*Kappaphycous allverzii*) and brown algae (*Turbinaria* sp. and *Sargassum* sp.) were taken from coastal water near Mandapam, Tamil Nadu (Vijayaraghavan and Shanthakumar, 2016) (Bay of Bengal), which can be seen in Figure 1. By severing the thallus with a knife close to the rizoid, all of the obtained samples were further processed. Algae were washed in seawater then dried under sunlight and stored in bags in a ventilated area. There are about 900 species of green seaweed among 4,000 red species and 1,500 brown species that can found in nature. The greatest variety of red seaweeds is found in subtropical and tropical waters while brown seaweeds are more common in cooler temperate waters. Seaweeds are marine algae, saltwater dwelling, and simple organisms that fall into the rather outdated general category of plants. Most of them are red (6,000 species), brown (2,000 species) or green (1,200 species).

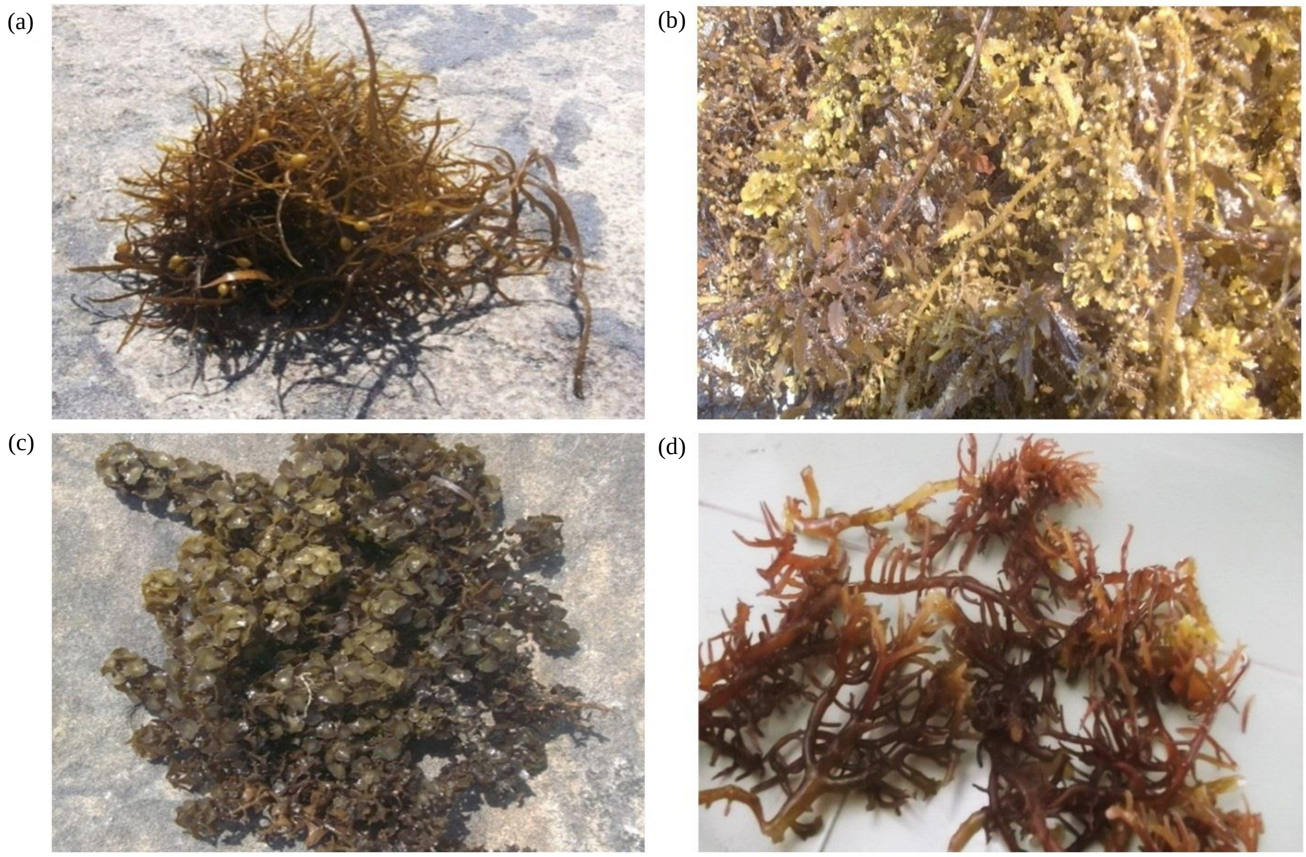


Figure 1. Various species of brown algae, (a) *Sargassum* sp. 1, (b) *Sargassum* sp. 2 and red algae (c) *Turbinaria* sp, (d) *Kaapaphycous allverzii*

2.2 Alginate preparation

Figure 2 shows the schematic diagram for the synthesis of sodium alginate. The collected algae were thoroughly washed by double-distilled water before drying for 30 h at 65 degrees. Alginates removal was done following the method from our previous work (Fenoradosoa et al., 2010; Vijayaraghavan and Shanthakumar, 2016). Twenty five gram aliquots of dried algae were soaked in 800 mL of 2% formaldehyde for 24 h at room temperature rinsed with water. Then, 800 mL of 0.2 M HCl was added and the process was continued for another 24 h. The samples were subsequently given another distilled water wash. Then, alginates were extracted for three hours at 100°C using 2% sodium carbonate. To precipitate the polysaccharides, three liters of 95% ethanol were added after the soluble fraction was filtered out. Sodium alginate was dried at 65°C after being collected and washed twice with 100 mL of acetone, and then transferred to 100 mL of water. Lastly, alginates were precipitated using ethanol (v/v) solution followed by drying at 65°C.

2.3 Dye solution preparation

Congo red dye standard solution (1,000 mg/L) was used to create the necessary concentrations of 50, 100, 150, 200, and 250 mg/L CR in synthetic textile wastewater solutions. The original concentration was made using distilled water and various pH levels of 4, 5, and 6 while mixing for an hour. With a -9.28 mV zeta potential, the CR dye surface is negatively charged. Various doses of calcium and alginates were applied to each 500 mL sample during this experiment.

2.4 UV-Vis spectrophotometer analysis

By measuring the dye sample's absorbance in distilled water at a known standard wavelength (510 nm), UV spectrophotometers can be made uniform. Using a UV spectrophotometer, the absorbance of standard dye solutions (10, 20, 30, mg/L, etc.) was measured. The congo red maximum absorbance wavelength was established at 510 nm. The absorbance of the unidentified sample concentration was then recorded. An equation is provided to determine the concentration of the unknown material and the graph between absorbance and concentration is displayed to confirm the straight line.

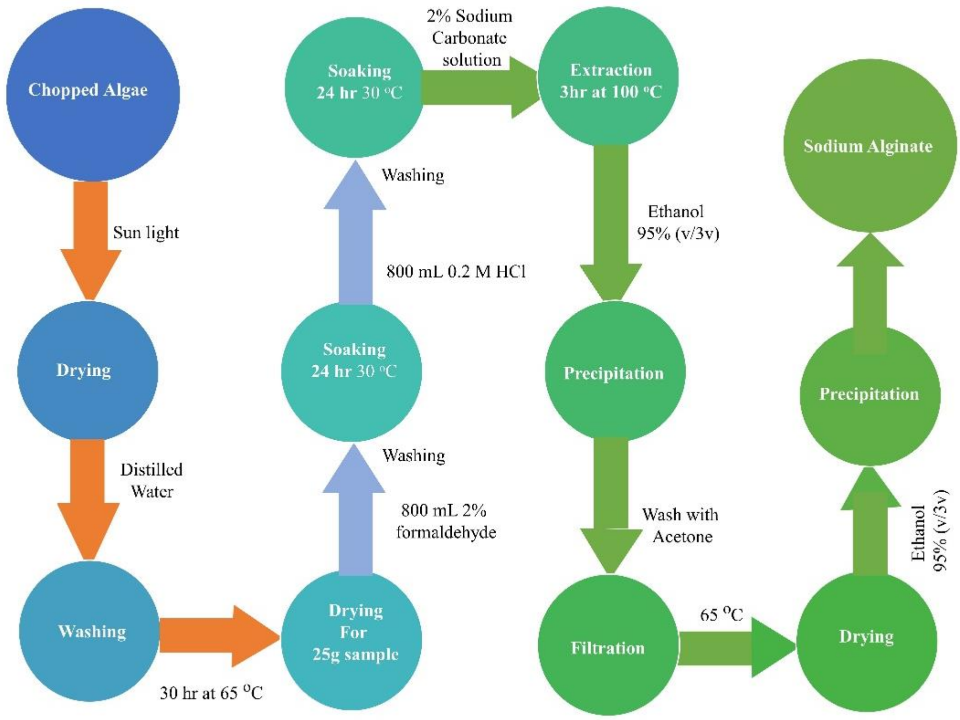


Figure 2. Preparation of sodium alginate

2.5 Experimental procedure

A typical jar test setup was used for the coagulation experiment. As alginate and congo red were both negatively charged substances calcium was introduced first followed by alginate. Colloid/colloid, polymer/colloid, and polymer/polymer pairings repulsive forces are reduced and the double layer is better compressed by the calcium during the coagulation process (Devrimci et al., 2012).

In this study, calcium was employed as CaCl and sodium alginate as alginate. Between 1 and 6 mg/L of calcium were given, and between 1 and 6 mg/L of alginate. Five minutes of gradually increased agitation at 120 rpm for calcium and 5 min of rapid agitation at 120 rpm for alginate, followed by 20 min gradual mixing at 40 rpm and 30 min of settling time were the mixing parameters of each sample used in the tests. Using Whatman filter paper number 42, the collected supernatant following sedimentation was filtered. The filtered solution was analyzed by UV spectrometry at 510 nm of maximum wavelength with the concentrations of 50, 100, 150, 200, and 250 mg/L, which offer improved treatment outcomes. The %CR dye removal was calculated by using equation (1).

$$\text{Dye removal (\%)} = \frac{C_i - C_f}{C_i} \qquad (1)$$

Where C_i and C_f are the initial and final dye concentration respectively.

3. RESULTS AND DISCUSSION

3.1 Yield % of alginate

About 25 g of dry seaweeds were examined to find out how much alginate was produced. Following the extraction method shown in Figure 1, it was noted that the sodium alginate yield contents from *Sargassum* sp. 1, *Sargassum* sp. 2, *Turbinaria* sp., and *Kappaphycus allverzii*, were found to be 10.2, 9.6, 9.8, and 10.5 g, respectively. Figure 3 shows the plot between the type of seaweed used to percentage of sodium alginate yield, which was found to be 40.8, 38.4, 39.2, and 42.0% for *Sargassum* sp. 1, *Sargassum* sp. 2, *Turbinaria* sp., and *Kappaphycous allverzii*, respectively. Based on these findings, it is noticeably clear that the experimental seaweed have the potential to be rich sources of alginate, which can be used as a natural coagulant for color removal.

3.2 Scanning electron microscopy (SEM) analysis

Figure 4 (a) and (b) show the Scanning Electron Microscope images of raw alginate surface and alginate surface after coagulation process. It was observed that the surface of alginate before the coagulation process possess blistering spine flakes and fine perforations. But after coagulation treatment process it was noted that sludge formed was fully lodged with swarms of dye particles on the entire fine porous surface of alginate. Based on the SEM images, it was observed that, when comparing the surface of alginate before and after coagulation test, the surface becomes smooth and no flakes were noted. This morphological change on the surface of the alginate reveals the removal of dye from aqueous solution through coagulation process.

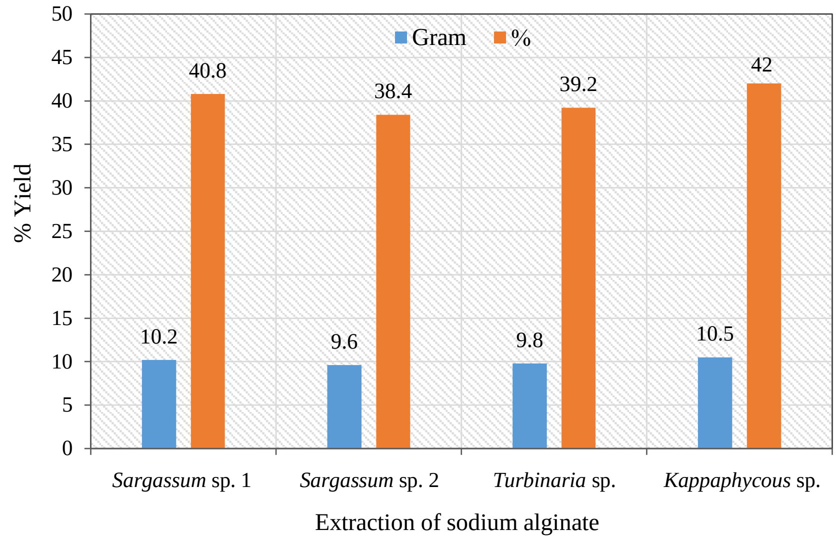
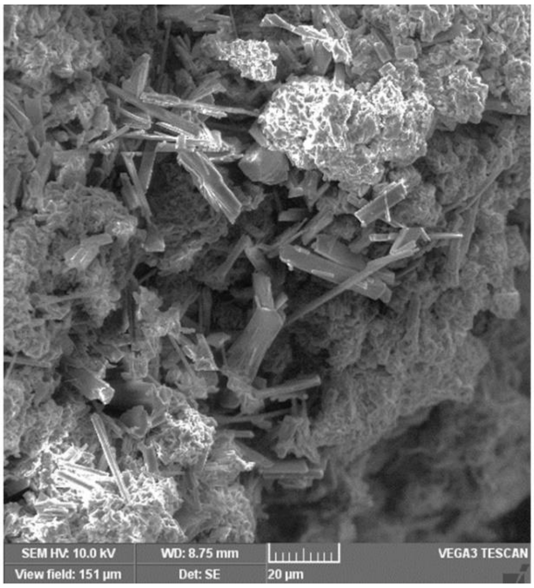


Figure 3. The yield of alginate from brown and red algae species

(a)



(b)

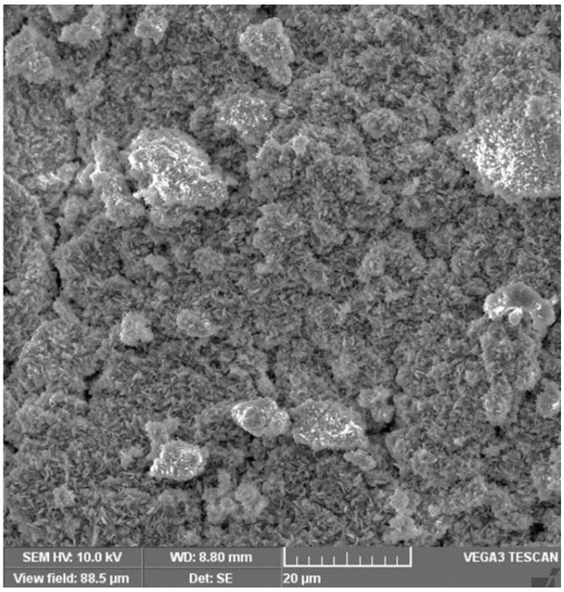


Figure 4. SEM image of (a) raw alginate (b) alginate surface loaded with congo red dye

3.3 Effects of coagulation parameters

The effects of calcium and alginate doses are shown in Figure 5(a)-(f) using synthetic textile wastewater with starting concentrations of 250 and 200 mg/L at pH values of 4, 5, and 6. Because it affects the surface characteristics of the calcium alginate and the ionization/dissociation of the CR dye molecule, the pH of the system has a significant impact on the coagulation process of CR. How dyes are removed from aqueous solutions with different technologies are crucial in wastewater treatment (Ponnusamy and Subramaniam, 2013). In this context, low pH causes an increase in the H^+ ion concentration, and the calcium alginate surface becomes positively charged by absorbing H^+ ions. Because the surface of calcium alginate is positively charged at low pH levels, an extremely strong electrostatic attraction forms between the positively charged calcium alginate surface and the anionic dye molecule that result in a decent amount of dye adsorption (Hoong and Ismail, 2018). More negatively charged sites are present in the systems when the pH increases, while fewer positively charged sites are present (Yupin et al., 2022). Electrostatic repulsion prevents the coagulation of anionic dye molecules at a negatively charged surface location on calcium alginate. In addition, the rivalry between extra OH ions and the anionic dye molecule causes the CR dye to coagulate less readily in an alkaline medium (Kristianto et al., 2019). Figure 5(a)-(c) demonstrates that CR dye removal increases with Calcium dose increases from 1-6 g/L. Moreover, while increasing pH from 4 to 6 the CR dye removal was gradually decreasing. The same phenomena occur during dye concentration decreases from 250 to 200 mg/L as can be seen in Figure 5(a)-(c). In both concentrations the dye removal was 95% at pH 4.

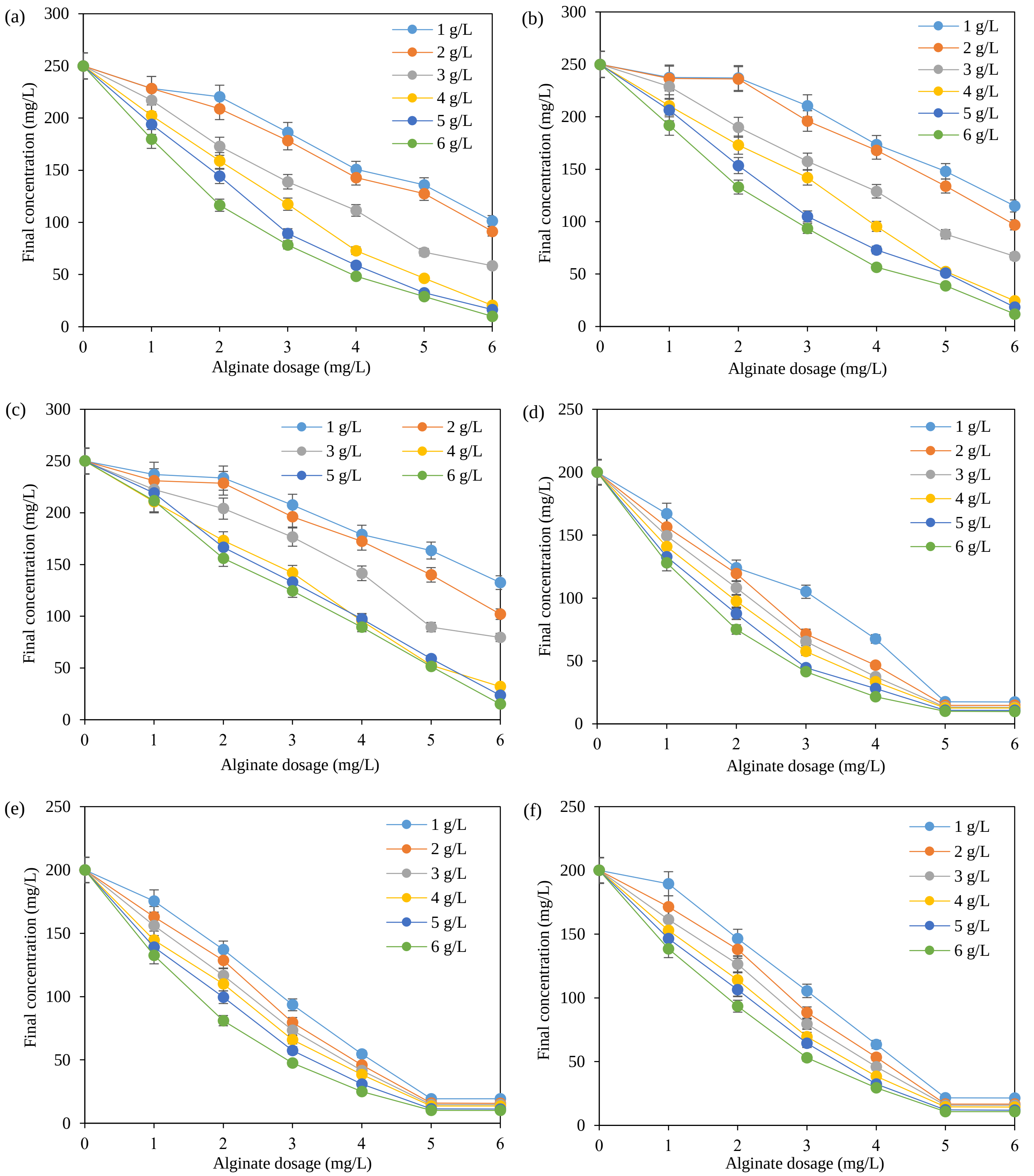


Figure 5. Effect of calcium and alginate dosage in synthetic textile wastewater at initial concentrations of 250 and 200 mg/L at different pH (a) 250 mg/L at 4 pH, (b) 250 mg/L at 5 pH, (c) 250 mg/L at 6 pH, (d) 200 mg/L at 4 pH, (e) 200 mg/L at 5 pH, (f) 200 mg/L at 6 pH

The performance at initial CR concentrations in synthetic textile wastewater of 250 and 200 mg/L is illustrated in Figure 5(a)-(f). This results clearly shows when calcium and alginate were added, the concentration value fell near 10.0 and 9.8 mg/L in 250 and 200 mg/L of synthetic textile waste water, respectively. According to the graph, dye removal efficiency changed as the alginate dose was increased while the calcium dose remained constant. On the other hand, the use of calcium in relation with the dye removal, the efficiency of calcium dose is dependent on alginate dose according to the results. The best dye removal results were obtained using calcium and alginate dosages of 6 and 6 mg/L in synthetic textile wastewater of 250 mg/L, respectively, and calcium and alginate dosages of 6 and 5 mg/L in synthetic textile wastewater of 200 mg/L. At low calcium dosages, calcium alginate generates an inappropriate gel.

When the synthetic textile waste water sample was negatively charged, the low calcium concentration was not enough to neutralize samples so the dyes remained stable. A dose of 6 mg/L of calcium dosage requires 6 and 5 mg/L of alginate dosage in 250 and 200 mg/L of synthetic textile waste water, respectively, to achieve required concentration levels. For 6 mg/L of calcium dosage with 6 and 5 mg/L of alginate dosage the percentage of dye removal values were calculated and removal rates were over 95%. These result show, in high initial concentration, the calcium alginate act as very effective coagulant in different combinations of calcium and alginate doses.

Figure 6(a)-(f) showing the results for initial concentration of synthetic textile waste water at 150 and 100 mg/L for different calcium and alginate doses. From the graphs, low final concentrations near 4.6 and 7.6 mg/L were achieved at 150 and 100 mg/L, respectively, in synthetic textile waste water for 6 mg/L of calcium dosage at different alginate doses studied. The alginate doses level at 150 and 100 mg/L were smaller than alginate dosage level at 250 and 200 mg/L. The optimum alginate dosage range at 250 and 200 mg/L was 6 and 5 mg/L for 6 mg/L of calcium dosage, respectively. On the other hand, this optimum alginate range at 150 and 100 mg/L was 4 and 3 mg/L for 5 mg/L of calcium dosage, respectively. The percentage dye removal values in 150 and 100 mg/L were over 95 % for 5 mg/L of calcium dosage. For low calcium dosage the percentage dye removal values were below 90% in both 150 and 100 mg/L. In low calcium dosage dye removal was lower but still varied depending on the different alginate dosages studied. These results show that at medium initial concentrations, the calcium alginate acts as very effective coagulant in different combination of calcium and alginate doses (Assimeddine et al., 2022).

The final concentration value for an initial concentration of 50 mg/L synthetic textile wastewater at various calcium and alginate doses is displayed in Figure 7(a)-(c). The performance at lower concentration increased when compared to the samples of higher concentration. The optimum concentration of alginate was 2 mg/L for various mg/L of calcium dosage for synthetic textile waste water samples. The final concentration values achieved as near 2.5 mg/L, with various calcium dosage. The efficiency of dye removal ranged from 94 to 96% depending on the calcium and alginate dose.

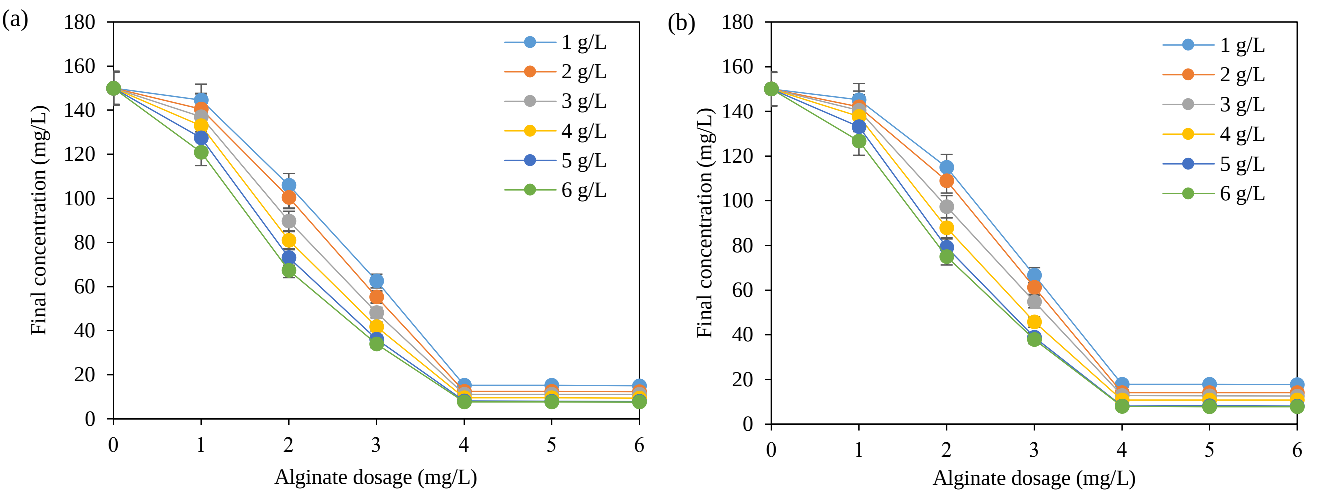


Figure 6. Effect of calcium and alginate dosage on synthetic textile wastewater at initial concentrations 150 and 100 mg/L at different pH (a) 150 mg/L at 4 pH, (b) 150 mg/L at 5 pH, (c) 150 mg/L at 6 pH, (d) 100 mg/L at 4 pH, (d) 100 mg/L at 5 pH, (d) 100 mg/L at 6 pH

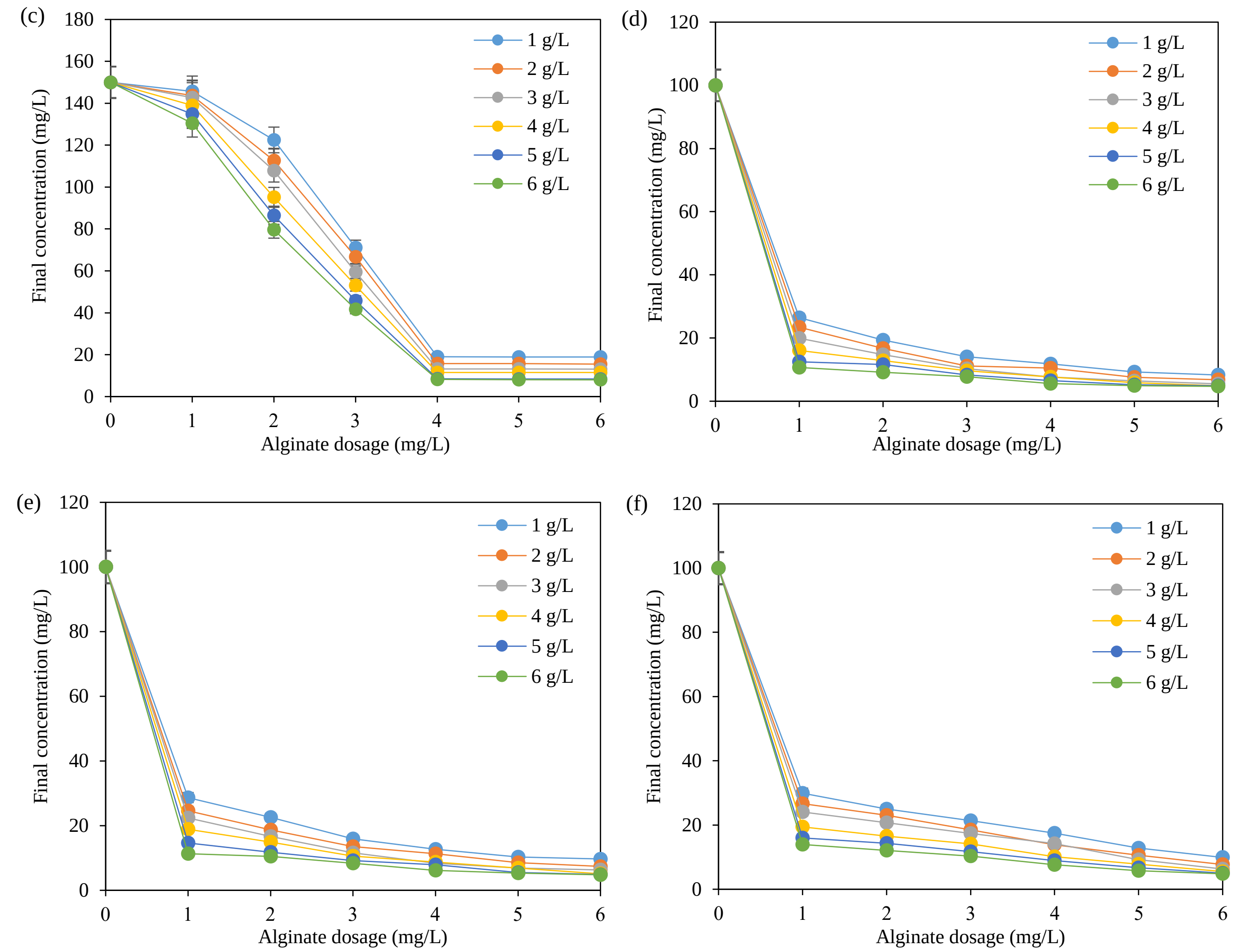


Figure 6. Effect of calcium and alginate dosage on synthetic textile wastewater at initial concentrations 150 and 100 mg/L at different pH (a) 150 mg/L at 4 pH, (b) 150 mg/L at 5 pH, (c) 150 mg/L at 6 pH, (d) 100 mg/L at 4 pH, (d) 100 mg/L at 5 pH, (d) 100 mg/L at 6 pH (cont.)

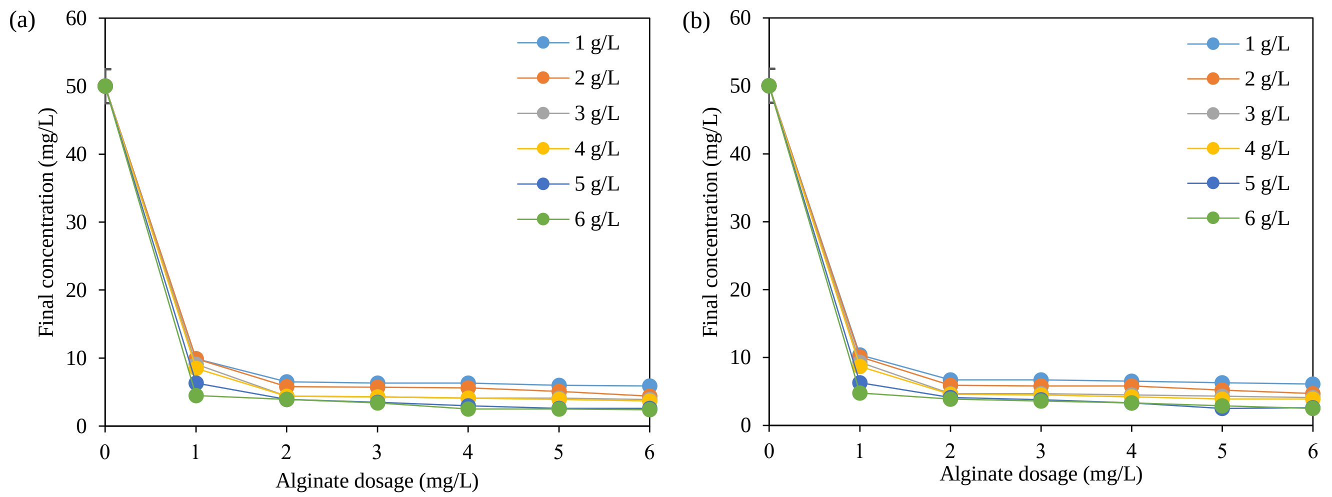


Figure 7. Effect of calcium and alginate dosage on synthetic textile wastewater at initial concentrations of 50 mg/L at different pH (a) 4 pH, (b) 5 pH, (c) 6 pH.

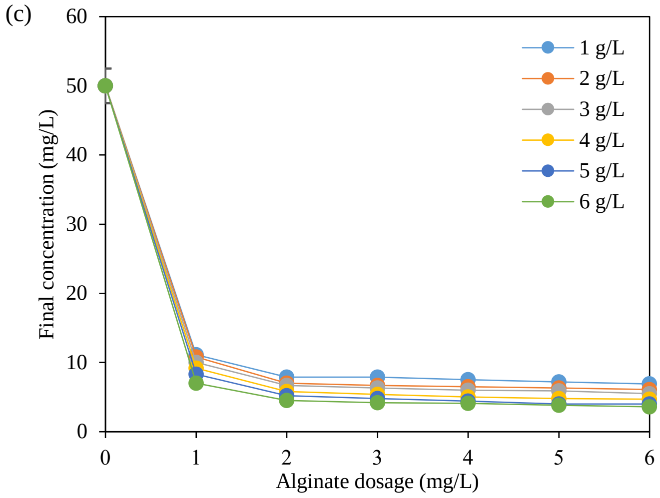


Figure 7. Effect of calcium and alginate dosage on synthetic textile wastewater at initial concentrations of 50 mg/L at different pH (a) 4 pH, (b) 5 pH, (c) 6 pH (cont.)

An increase in the dye removal efficiency was due to the absence of enough particles to constitute nuclei for the flocs to form. The calcium alginate gel formation is the main mechanism, because during or after the stage of gel formation it combines and captures particles to form particles heavy enough to settle down. However, if the flocs were not heavy particles it was not enough to settle down and separate. Calcium alginate gels might have also contributed to some extra turbidity. With these observations low concentration samples may not make floc formation, so using higher viscosity alginates gave better results than lower viscosity alginates in final concentration values. When the alginate molecular weight is higher it increases viscosity and dye removal efficiency. Molecular weight increase is directly related to the polymer chain length. So when it increases, the polymer's bridging ability increases (Palapa et al., 2021).

This kind of intraparticle bridging mechanism helps a lot to form big flocs, so that the settling of particles will be more after adding the suitable coagulant. The other factors like double layer compression, sweep flocculation and charge neutralization may influence a little. However, this bio coagulant, capable of producing a gel, will follow the intraparticle bridging mechanism commonly known as the Egg box model (Ekanayake and Manage, 2022). The presence of carboxyl groups with double bonds increases the affinity of the dye particle towards the coagulant.

Table 1. Overall best performance conditions for congo red dye removal

Initial concentration (mg/L)	pH	Alginate dosage (mg/L)	Calcium dosage (mg/L)	Final concentration (mg/L)	% of dye removal
50	4	2	4	3.9	92.20
	5	2	5	4.1	91.80
	6	2	6	4.5	91.00
100	4	3	5	8.3	91.70
	5	3	4	9.2	90.80
	6	3	6	10.3	89.70
150	4	4	5	8.1	94.60
	5	4	5	8.1	94.60
	6	4	5	8.5	94.33
200	4	5	6	9.9	95.05
	5	5	5	11.4	94.30
	6	5	6	10.8	94.60
250	4	6	6	10	95.00
	5	6	6	12	94.00
	6	6	6	15	92.50

These results have demonstrated explicitly that calcium alginate can function as a coagulant for the removal of color from synthetic textile effluent. Alginate and calcium both failed to reach the desired concentration levels when used alone. Therefore, their combined effect is thought to be necessary. A positively charged substance, such as calcium, must be added to the system due to the alginic acid polysaccharides and the utilized particles negative surface charges. According to Davis et al. (2003), the monomeric units of mannuronic acids and guluronic acids have values of 3.38 and 3.65, respectively. Alginic acid must therefore approach and bind to the surface of calcium ions. As a result, the initial stage in the calcium alginate dye removal technique is surface charge neutralization. Therefore, adding calcium before alginate improved system function. According to Simpson et al. (2004), the action of alginate is the production of calcium alginate gel, or it's held to bridge the gap between the particles by one of two methods.

The main reason to choose combinations for study is because calcium alginate dosage could often attain low concentration values. Table 1 lists the lowest concentrations that were attained together with the corresponding concentrations of calcium and alginate. At different calcium and alginate doses for samples with beginning calcium concentrations of 250, 200, 150, 100, and 50 mg/L, low target values could be achieved. These studies collectively demonstrated that the calcium coagulates more effectively.

3.4 Coagulation kinetics

Kinetic study helps to determine the rate of dye molecules removal form aqueous solution. In this view first order and second order kinetic model has been studied for CR dye removal from textile effluent by alginate as natural coagulant. The following equations are used to verify the first order (Equation 2) and second order (Equation 3) kinetic, respectively.

$$\log\left(\frac{C_i}{C_o}\right) = -kt \tag{2}$$

$$\frac{1}{C_o} - \frac{1}{C_i} = k't \tag{3}$$

Where C_i is initial dye concentration, C_0 is the Concentration of dye after time (minutes), and k is first order and k' second order rate constant (min^{-1}).

From Figure 8(a) and (b) can be seen that the first order and second order kinetics to the experimental data, respectively. These kinetic data show that the intercept for first order does not obey the kinetic equation. Therefore, the coagulation process for removal of CR dye using alginate followed a second order model (Nnaji et al., 2014; Vijayaraghavan et al., 2018).

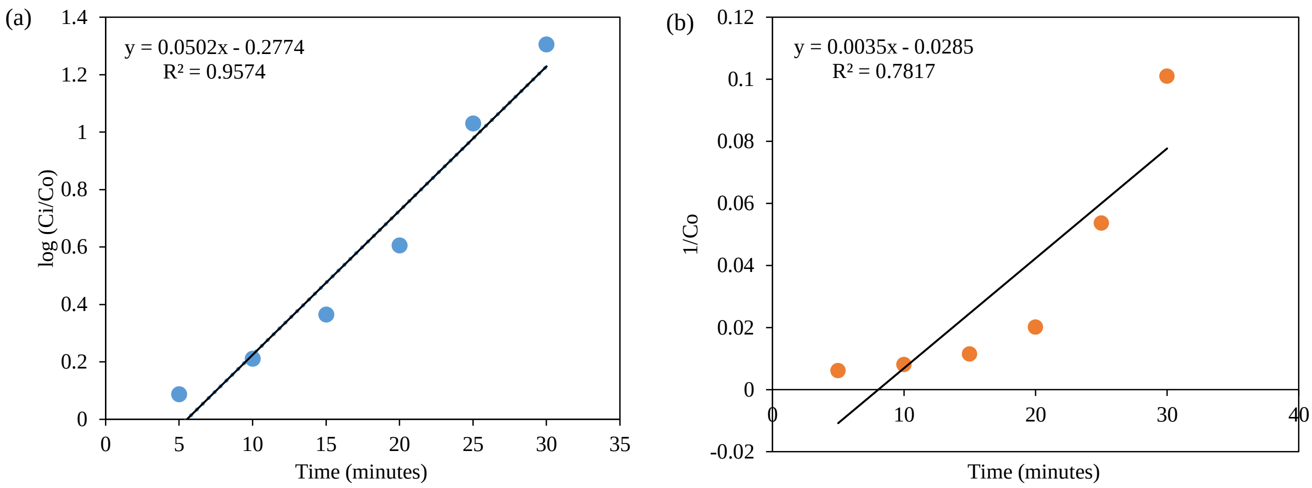


Figure 8. Kinetics study of CR dye (a) first order and (b) second order

4. CONCLUSION

The current study concluded that CR dye can be removed from aqueous solution using natural coagulant obtained from coastal water. The procured alginate worked effectively as coagulant and achieved maximum percentage removal by varying calcium and alginate doses in synthetic textile wastewater at varying concentrations (50-250 mg/L) and pH (4, 5, 6) levels. When compared to low pH at the same concentration of synthetic textile wastewater, hydroxyl group decomposition at high pH resulted in low coagulation process efficiency. In comparison with other decolorization processes, this technique attained results in a short time. The maximum removal percentage of 95.05% was achieved by the operating conditions of pH 4, calcium and alginate dosage of 6.0 g/L. The calcium alginate acted well as a coagulant and removed congo red dye from the aqueous solution effectively, which is present in textile wastewater. Our future study will recover or degrade the dye from sludge using solvent extraction. Once the dye is recovered or degraded, the treated contents can be used as a manure/fertilizer. But the limitations involved in the dye recovery process from the sludge should be large. In the present process, the quantity of sludge formed is very small amount, hence the dye recovery process can be feasible only if the process is scaled up to large quantity.

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CONFLICT OF INTEREST

The authors hereby declare that they do not have any conflict of interest.

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