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Performance study on algal alginate as natural coagulant for the removal of Congo red dye

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ABSTRACT

Coagulation is an important step in the physicochemical treatment of wastewater. Due to the disadvantage associated with the commonly used inorganic metal salts and other synthetic polymers, in recent years, research on the use of natural organic polymers as coagulant is gaining importance. In this study, the coagulation potential of alginate extracted from brown algae, *Sargassum* sp. for the removal of Congo red dye from aqueous solution has been identified. The yield of alginate extraction was found to be 40.8%. The extracted alginate was characterized by Fourier transform infrared spectroscopy and Scanning electron microscopy techniques. The effect of initial pH (4–6), alginate dose (10–60 mg/L), calcium dose (1–6 g/L), and initial dye concentration (50–250 mg/L) on dye removal have been investigated. It is inferred from the study that the maximum removal of dye (96%) was achieved with increasing alginate and calcium dose for increasing dye concentration, at pH 4. The obtained results were compared with the literature on Congo red dye removal using various adsorbents and coagulants.

Keywords: Brown algae; Alginate; Coagulation; Congo red dye

1. Introduction

Among various industrial sectors, textile industries are one of the most common and essential which generate a large volume of wastewater with varying physicochemical characteristics. The variation in physicochemical characteristics is due to many ingredients used in the process, due to which an overwhelming continuous effort is carried out in order to identify appropriate technologies to treat textile industry wastewater [1]. The conventional wastewater treatment involves aerobic biodegradation; however, due to their low efficiency, the present treatment method is based on physical and chemical processes. The various types of wastewater treatment carried out for textile industry wastewater include coagulation/flocculation, oxidation, membrane separation, ion exchange, photochemical, adsorption, biological treatment method, etc [2].

Among the various methods, coagulation is the one of the effective method used for removal of dyes present in the wastewater. Coagulation is the process in which chemicals are added to change the surface of the colloids and make them agglomerate together to settle down. Generally, inorganic metals (such as

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alum, ferric chloride, lime, etc.) and polymers (natural or synthetic polymer) are used as coagulant aid. Alum (aluminum sulfate) is the most commonly used coagulant for many years. However, the requirement of high quantities of chemicals, production of large volume of sludge, and the considerable change in pH during the treatment are the main disadvantages of coagulation by metal salts [3]. The concern about the suspected role of aluminum in Alzheimer's disease is another drawback of alum [4–6]. In addition, the use of synthetic polymers in coagulation has few negative impacts on human health and hence, the research focus on natural polymeric materials (as coagulants) gains importance in recent years.

The natural polymeric materials such as polysaccharides have been suggested due to its easy availability, low cost, low-molecular weight, and high shear stability. Further, the biodegradability, effectiveness for various colloidal suspensions, and human health safety are the additional advantages [6,7]. The performance of plant-based coagulants extracted from naturally available materials such as *Prosopis juliflora*, *Cactus latifaria*, *Moringa oleifera*, etc. is significant in removal of turbidity. In addition, researchers [8–10] have studied cactus extracts, chitosan, and cationic starches as alternative coagulants and reported that these materials have coagulation and flocculation capacity for turbidity removal. The use of low-cost alternative treatment technologies and materials is vital for the treatment of wastewater [11].

Among the naturally available materials (used as coagulants), the alginate extracted from algae also have significant potential as coagulant for treatment of wastewater. Macro-algae can be classified as red algae (Rhodophyta), brown algae (Phaeophyta), or green algae (Chlorophyta) depending on their nutrient and chemical composition [12]. The marine brown algae are the main source of alginic acid or alginate, a natural polysaccharide. The ability of alginate to react with polyvalent metal cations, especially calcium ions to produce strong gels or insoluble polymers made to choose this as a coagulant for wastewater treatment [13–15]. The monomers present in the alginate extracted from the brown algae are D-mannuronic acid and L-guluronic acid [16]. The carboxyl groups present in the alginate can accept any functional group and forms gel structure [17]. Previously, researchers studied algal alginate as coagulant for wastewater treatment. However, removal of Congo red dye from aqueous solution using alginate extracted from brown algae, *Sargassum* sp. has not yet been studied. The objectives of this study are to ascertain (i) the alginate yield from *Sargassum* sp. and (ii) the coagulation potential of extracted algal alginate for removal of Congo red dye.

2. Materials and methods

2.1. Algae collection

The brown algae, *Sargassum* sp. was collected from the coastal waters (Bay of Bengal) in Mandapam, Tamil Nadu. The samples were collected by cutting the thallus with a knife near the rhizoid. The collected samples were washed with seawater in the site and stored in bags with ventilation before transporting it to the laboratory for further processing.

2.2. Alginate preparation

The algae samples received in the laboratory were washed abundantly with tap water and dried in the sunlight for three hours. Later, the samples were dried for 30 h at 65˚C in hot air oven. The alginate industry extraction protocol is divided into five steps: acidification, alkaline extraction, solid/liquid separation, precipitation, and drying [18]. The procedure reported by Fenoradosoa et al. [19] was followed to extract the alginate from the algae. Twenty five gram of dried algae was soaked in 800 mL of 2% formaldehyde for 24 h at room temperature and washed with water. Later, 800 mL of 0.2 M HCl was added to the sample and left for 24 h. The samples were washed again with distilled water and the alginates were extracted with 2% sodium carbonate at 100˚C. The soluble fraction was collected by filtration and polysaccharides were precipitated by three volumes of ethanol 95%. Alginate collected was washed twice by 100 mL of acetone, dried at 65˚C, and dissolved in 100 mL of distilled water. It was then precipitated again with ethanol (v/3v) and dried at 65˚C. The procedure adopted for alginate extraction is presented as flow sheet in Fig. 1. It was reported that the sodium alginate extracted from the *Sargassum* sp. has a high-molecular weight polysaccharide of weight-average molecular weight and number-average molecular weight as 5.528×10^5 and 3.852×10^5 g mol^{-1}, respectively [19]. The PKa values of the monomeric units of alginate, D-mannuronic acid, and L-guluronic acid were reported as 3.38 and 3.65, respectively [20]. Zeta potential showing surface charge of alginate monomeric units was reported as −60 mV for 500 M and −50 mV for 500 G [21]. Charge density, θ of alginate gel with calcium chloride (for M/G = 1) was reported as -9.00×10^{-3} mol dm^{-3} [22].

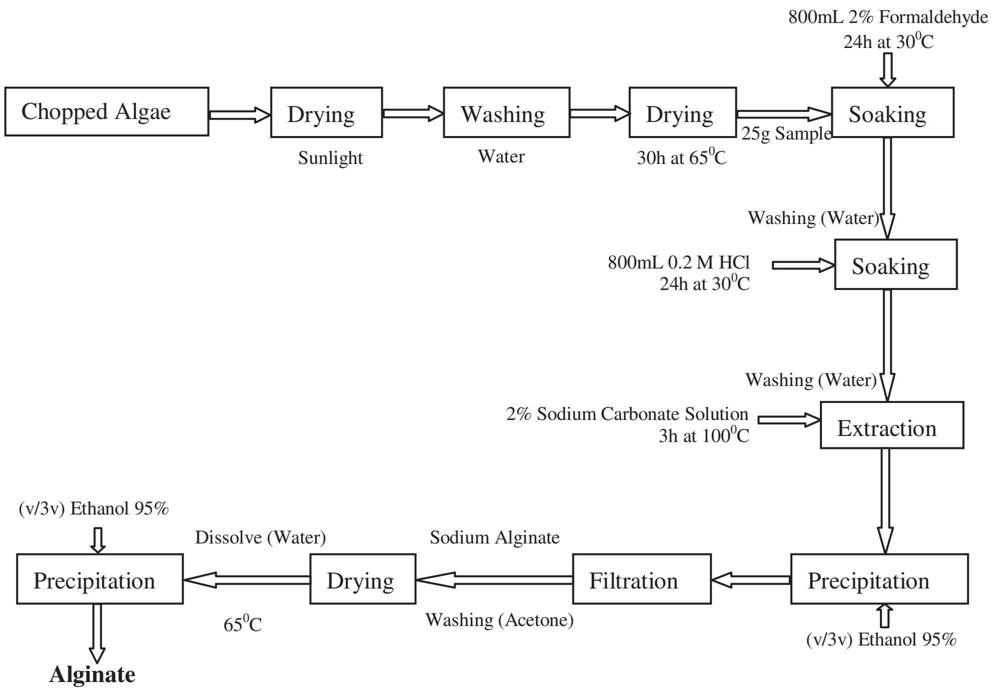

Fig. 1. Schematic flow diagram of sodium alginate preparation.

2.3. Sample preparation

The Congo red dye used in the study is the sodium salt of benzidinediazo-bis-1-naphthylamine-4-sulfonic acid having a chemical formula $C_{32}H_{22}N_6Na_2O_6S_2$ and molecular mass: 696.66 g/mol. Stock solution (1,000 mg/L) was prepared by dissolving appropriate quantity of Congo red dye in double distilled water. The standard solutions were made from the stock solution to obtain desired concentration of dye solution. The concentration of Congo red dye was determined by using a UV-spectrophotometer (Systronics-119, India) at a wavelength corresponding to maximum absorbance of 510 nm; HCl (0.1 N) and NaOH (0.1 N) were used to adjust the pH of the solution.

2.4. Characterization of alginate

The alginate extracted from the brown algae *Sargassum* sp. was characterized using Fourier transform infrared (FTIR) spectroscopy and scanning electron microscopy (SEM). The FTIR spectroscopy (Thermo Nicolet, AVATAR 330) was employed to determine the functional groups present in the alginate. FTIR was done both for raw and dye-loaded samples. The infrared spectrum of alginate was recorded as KBr disks in the range of 4,000–400 cm^{-1}. Scanning electron microscopy (TESCAN-VEGA3, Czech Republic) was used to characterize the surface structure and morphology of the raw and dye-loaded samples of alginate.

2.5. Experimental procedure

Batch scale studies were carried out using alginate extracted from *Sargassum* sp. for removal of Congo red dye from aqueous solutions. Conventional jar test apparatus (Dolphin CIC-304, Sciensil India) was employed and all the experiments were performed in 1,000 mL beakers containing 500 mL of dye solution by following the standard procedure. Calcium as calcium chloride and alginate as extracted algal sodium alginate were used for the experiment. The calcium dose varied between 1 and 6 g/L, whereas the alginate dose varied between 10 and 60 mg/L. Calcium was added first and then alginate was added for all the experiments. The initially added calcium chloride produces Ca^{2+} ions, which act as a binding material between the cross linkages of functional groups in the alginate polymer chain [23,24]. The experiment for each sample was carried out in the following order: 5 min rapid mixing at 100 rpm for calcium dosing, 5 min rapid mixing at 100 rpm for alginate dosing, followed by 20 min slow mixing at 40 rpm, and 30 min for settling. The supernatant after sedimentation was filtered using Whatman No. 42 filter paper. The filtrate was analyzed using UV-spectrophotometer. The experiments were carried out for different initial dye concentrations of 50, 100, 150, 200, and 250 mg/L.

3. Results and discussion

3.1. Alginate yield

To determine alginate yield of brown algae *Sargassum* sp. 25 g of dried brown algae sample was analyzed. The experiments were conducted in four trials and the results obtained were in the range of 39.9–42.3% with an average yield of 40.8%. The variation of alginate yield during each trial could be due to fact that a fraction of alginate powder is lost during washing and filtration processes. The alginate yield indicates the potential application of seaweed for the production of natural coagulant.

3.2. SEM

The SEM images (of various magnifications) of alginate from *Sargassum* sp. are presented in Fig. 2. It can be noted from the figure that the raw alginate (Fig. 2(a) and (b)) contain strands of fibers with numerous pores, whereas after coagulation test (Fig. 2(c) and (d)) the pores were filled with dye and calcium present in the sample. Further, it can be noted from Fig. 2(c) and (d), that the dye uptake on surface of alginate as a gel, formed due to the mechanism described in Section 1 (Introduction). The morphological change in surface of the alginate reveals the removal of dye from aqueous solution through coagulation.

3.3. FTIR analysis

The FTIR spectrum of raw alginate is presented in Fig. 3(a). A broadband stretch at 3,130 cm^{-1} was assigned to hydrogen bonded O–H group. A sharp and strong absorption band at 1,612 cm^{-1} represents the C–C stretch. The bend 1,460 cm^{-1} characterizes the methylene C–H stretch. The band measured at 1,089 cm^{-1} may be due to the appearance of C–C–H and O–C–H. A major band in the region 1,697 cm^{-1} indicates the presence of C=O group and this confirms the nature of alginate. Fig. 3(b) represents the FTIR spectrum of sludge containing alginate and Congo red dye. The wave number 1,633 cm^{-1} was assigned to the open-chain azo (–N=N–) group which is due to the presence of Congo red dye in the sludge [8,19].

3.4. Effect of pH

The pH is the influencing parameter in the coagulation process. The pH of the system influences on the surface charge of coagulants and also the stabilization of the suspension [23]. The variations in removal of Congo red dye from aqueous solution at different pH conditions are depicted in Fig. 4(a). It can be noted from the figure that the maximum percentage dye removal was achieved at acidic conditions, particularly at pH 4. The zeta potential value of alginate was reported as −60 and −50 mV for the M & G monomeric units showed that the alginate surface had a negative charge [21]. However, due to the mechanism that at low pH, the concentration of H^+ ions is more, and the alginate surface obtains positive charge by absorbing H^+ ions. This condition leads to strong electrostatic attraction between the positively charged alginate surface and the anionic Congo red dye molecule. Due to this electrostatic attraction, the dye molecules adhere on the alginate surface and settled down as sludge. When the pH increases, the number of positively charged ions decreases, which does not favor the coagulation of anionic dye molecules due to the electrostatic repulsion. The obtained results are consistent with the results reported in the literature [23,25,26]. As pH 4 was obtained as optimum, all the remaining experiments were carried out at pH 4.

3.5. Effect of calcium and alginate dose

The effect of calcium and alginate dose on dye removal was studied by varying the initial dye concentration (50–250 mg/L) and the results are presented in Fig. 4(b)–(f). It can be noted from the figures that the percentage dye removal increases with increase in alginate dose for all calcium dose levels studied. Further, it can be noted that for low initial dye concentrations (i.e. 50 and 100 mg/L), the dye removal is significant even if the calcium and alginate dose are less. However, when the initial dye concentration increases the low calcium dose is not effective in dye removal. The maximum dye removal (96%) for higher initial dye concentration studied (i.e. 250 mg/L) was achieved at 6 g/L of calcium dose and 60 mg/L alginate dose. It was observed during the experiments that the calcium alginate gel formation was not appropriate at low calcium doses.

The Congo red dye and the alginate are negatively charged and hence they need a positively charged metal cation, like calcium to neutralize their surface charges. The divalent calcium metal cation acted as a cross linkers in the alginate chain which forms a stable gel structure [24,27].

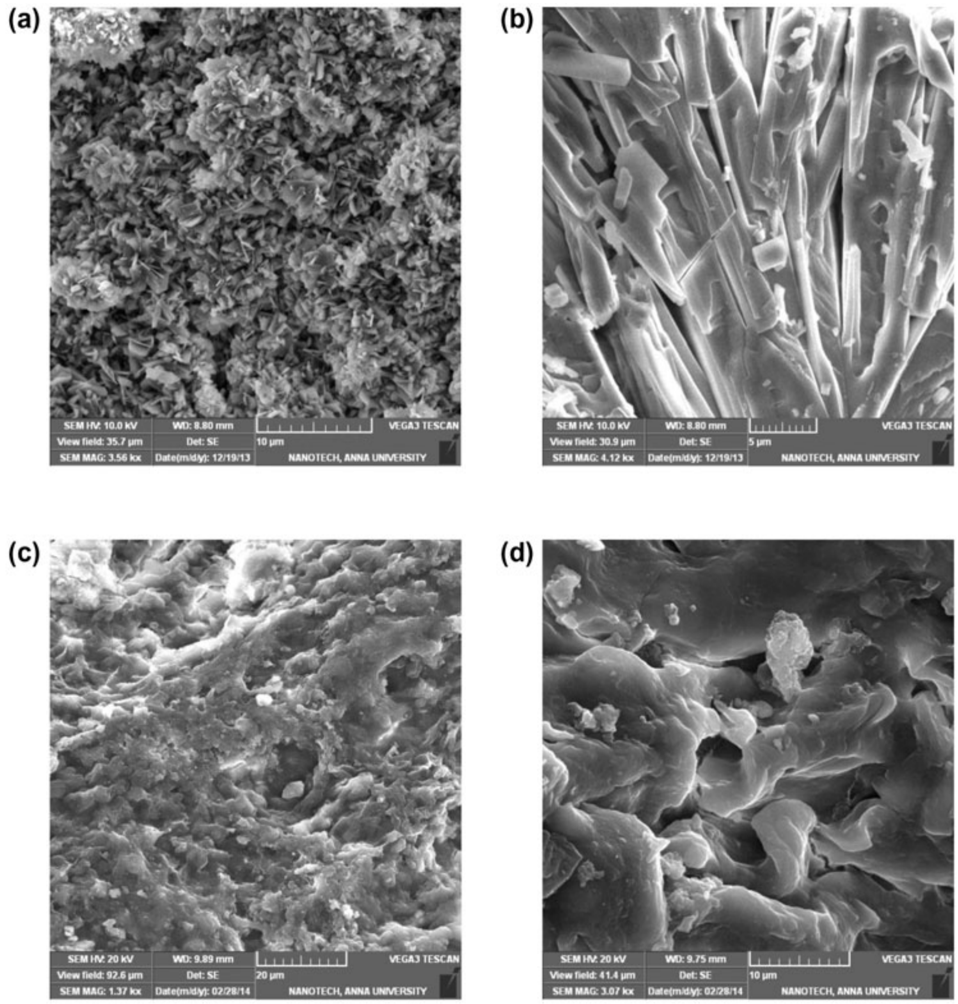

Fig. 2. SEM images of alginate (i) raw (a and b) and (ii) after coagulation (c and d).

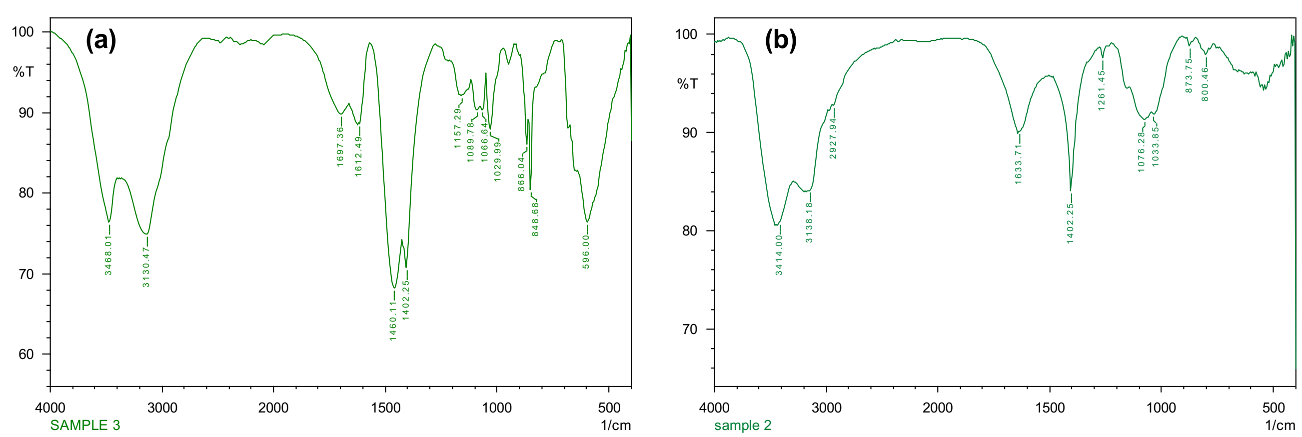

Fig. 3. FTIR spectra of raw (a) and dye-loaded alginate (b).

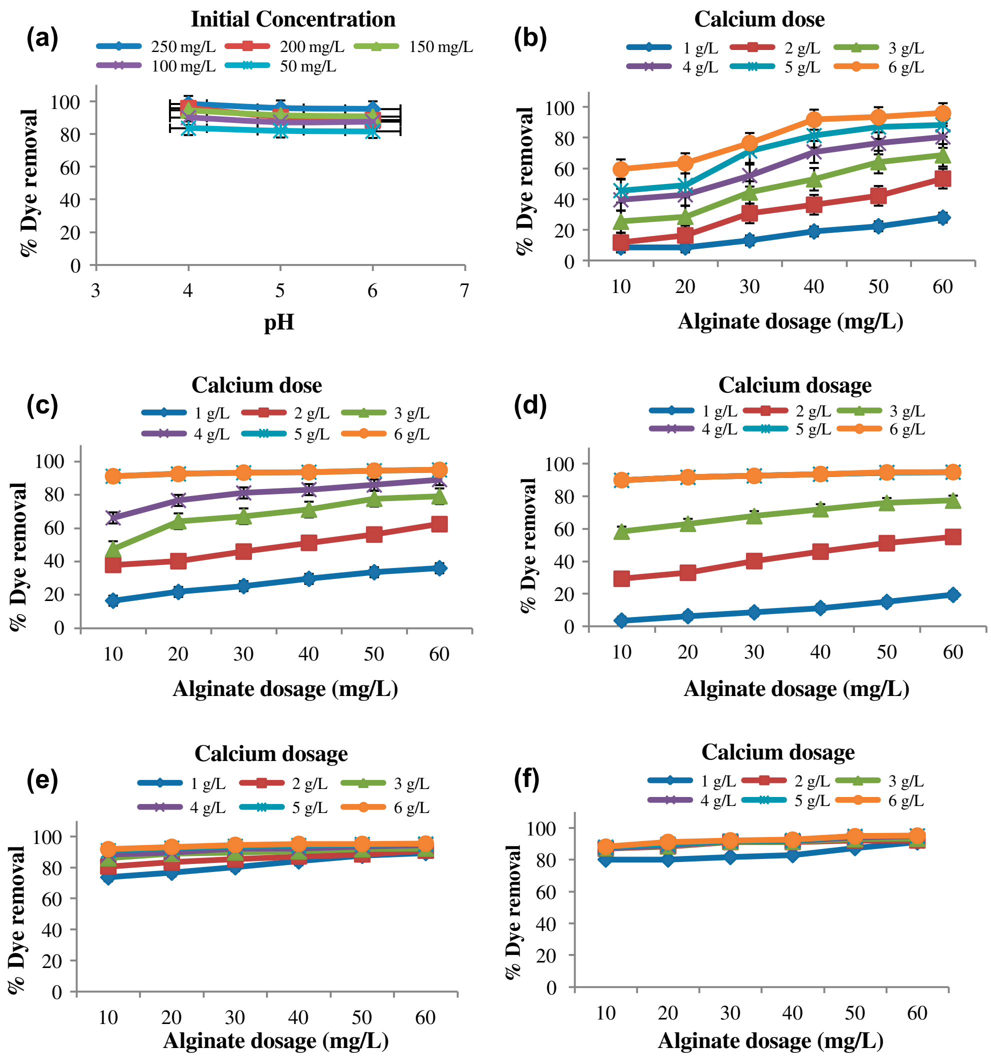

Fig. 4. Effect of (a) pH and effect of calcium and alginate dose on initial dye concentration, (b) 250 mg/L, (c) 200 mg/L, (d) 150 mg/L, (e) 100 mg/L, and (f) 50 mg/L.

The optimum conditions for Congo red dye removal at various initial dye concentrations are presented in Table 1. Further, the obtained results were compared with the literature on Congo red dye removal using various adsorbents and coagulants, and presented in Table 2. It can be noted from Table 2 that the results obtained in the present study are quite comparable with the results reported in the literature.

Table 1
Optimum conditions for Congo red dye removal

Initial dye conc. (mg/L)	Alginate dose (mg/L)	Calcium dose (g/L)	Final dye conc. (mg/L)	% Dye removal
50	20	4	3.9	92.2
100	30	5	8.3	91.7
150	40	5	8.1	94.6
200	50	6	9.9	95.5
250	60	6	10	96

Table 2
Comparison of various adsorbents/coagulants used for removal of Congo red dye

S. No	Process	Adsorbent/coagulant used	Initial dye conc. (mg/L)	Percentage color removal	References
1	Coagulation	Alum	20	99	[28]
		Magnesium chloride and calcium hydroxide	20	99	
		Ferrous sulfate and calcium hydroxide	20	90	
2	Adsorption	Chitosan	50	98	[29]
3	Coagulation	$MgCl_2$	200	79.3	[30]
		$FeSO_4$	200	78.9	
4	Coagulation	Lime	200	92	[31]
		Alum	200	99	
	Adsorption	Activated charcoal	200	99	
5	Adsorption	Rice husk	1	93.4	[32]
6	Adsorption	Papaya seed	50	79	[33]
7	Adsorption	Silver nanoparticles	2	88	[34]
8	Photo catalytic reaction	Chitosan/nano-CdS	20	98	[35]
9	Coagulation	Poly dimethylaminoethyl methacrylate	400	90.6	[36]
10	Adsorption	*Leucaena leucocephala* seed pods	10	91	[37]
11	Adsorption	*Martynia annua*, L. Seeds	50	84	[38]
12	Phytoremediation	*Chara Vulgaris*	10	95	[39]
13	Adsorption	Cashew nut shell	20	94	[40]
14	Biosorption	Tendu leaf	250	75	[41]
15	Adsorption	Coir pith	5	92.8	[42]
		Rice husk	5	89.7	
		Bagasse	5	86.5	
16	Coagulation	Algal alginate	50	92.2	*Present study*
			100	91.7	
			150	94.6	
			200	95.5	
			250	96	

4. Conclusion

The coagulation potential of alginate extracted from brown algae, *Sargassum* sp. for the removal of Congo red dye from aqueous solution has been investigated. The performance of alginate was highly dependent on the calcium dose and initial dye concentration. Due to the surface property of alginate and calcium, it is highly suitable for anionic dye. The percentage dye removal increases with increase in alginate dose for all calcium dose levels studied. At low initial dye concentrations, the dye removal is significant even with the less amount of calcium and alginate dose. The maximum dye removal of 96% was achieved for 250 mg/L initial dye concentration at 6 g/L of calcium dose and 60 mg/L alginate dose. Based on the investigation, it is apparent that the alginate extracted from brown algae, *Sargassum* sp. can be used as a potential coagulant for the removal of Congo red dye from aqueous solution.

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