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Coagulation performance evaluation of alginate as a natural coagulant for the treatment of turbid water

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ABSTRACT

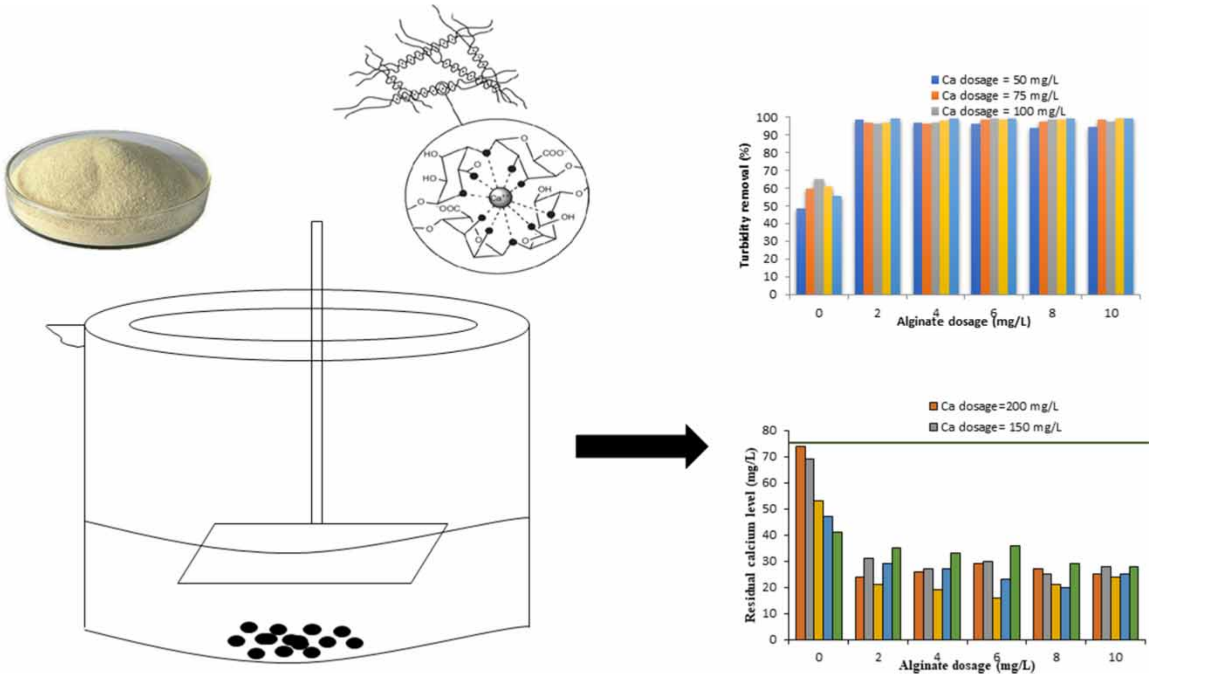
Alginates are quite abundant in nature as they occur both as a structural component in marine brown algae (Phaeophyceae) comprising up to 40% of dry matter and as capsular polysaccharides in soil bacteria. Alginic acid is the only polysaccharide, which naturally contains carboxyl groups in each constituent residue, and possesses various abilities for functional materials. Experiments were carried out for water of turbidity 300 NTU. Alginate as such doesn't act as a coagulant, instead it should be converted to calcium alginate by adding calcium ions. Calcium chloride was used for imparting calcium ions necessary for the reaction. The dosage of calcium was fixed as 50 mg/L, 75 mg/L, 100 mg/L, 150 mg/L, 200 mg/L, and alginate doses between 2 to 10 mg/L. Calcium dosage below 50 mg/L was not sufficient for the formation of the egg-box structure which is responsible for the coagulation and flocculation process. For the mechanism of charge neutralization to take place effectively, calcium should be added first followed by alginate. pH and conductivity of the sample remain constant before and after the treatment. The dosage of alginate required for the treatment is less, so the cost of treatment also will be much less, thus alginate can replace the usage of chemical coagulants like alum.

Key words: alginate, calcium chloride, coagulation, flocculation, turbidity

HIGHLIGHTS

- Alginate, a natural and renewable bio-flocculant, produces no secondary pollution and hence can be used as potential coagulant for drinking water purposes.
- Unlike other coagulants, the pH and conductivity of the treated water do not change.
- The concentration of Ca ions is well within the desirable limit after treatment.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Water is used for several purposes by humans but the level of purity of the water being consumed is very crucial since it has a direct effect on health. Appropriate treatment technology can render this poor water resource into safe potable water; however, conventional technology may not be appropriate for those communities in terms of economics, availability, and operational constraints (Zhao *et al.* 2012; Aneem *et al.* 2021). The high cost of treated water makes most people in the rural communities resort to readily available sources, which are normally of low quality exposing them to waterborne diseases (Gao *et al.* 2005).

The cost of water treatment is increasing and the quality of river water is not stable due to suspended and colloidal particle load caused by land development (Gravastrand *et al.* 2017; Mortadi *et al.* 2020). As a result of the high storms during rainy seasons the turbidity level increases and the need for water treatment chemicals increases as well, which also leads to a high cost of treatment that the water treatment companies cannot sustain. As a result, the drinking water that reaches the consumer is not properly treated. The lack of safe drinking water is a leading cause of morbidity and mortality, especially in local communities where waterborne diseases are prevalent and persistent due to low-quality surface source water. Therefore, it is of great importance to find a natural alternative for water coagulants to treat turbidity (Hopkins *et al.* 1993; Roussy *et al.* 2005; Shan *et al.* 2017; Delelegn *et al.* 2018). Turbidity removal is one of the important steps in the water treatment process and generally is achieved using the coagulation process (Wu *et al.* 2012).

Many coagulants have been widely used in conventional water treatment processes depending on their chemical characteristics. Recent studies have pointed out several serious drawbacks of using the two most common coagulants, aluminum and iron salts, such as Alzheimer's disease (Ng *et al.* 2012; Wu *et al.* 2012; Tang *et al.* 2015), production of large sludge volume, reduction of pH (Amagloh & Benang 2009; Vijayaraghavan & Shanthakumar 2016; Prokopova *et al.* 2021), and low efficiency in coagulation of cold water (Nwaiwu & Bello 2011) . In addition, their application is inappropriate in some developing countries because of the high cost and low availability (Okolo *et al.* 2021). The conventional method of water purification using aluminum sulfate (alum) and calcium hypochlorite puts pressure on the nation's over-burdened financial resources since they are imported, thereby making treated water very expensive in most developing countries and beyond the reach of most rural folks (Dechojarassri *et al.* 2018).

Many studies concerning natural coagulants obtained from marine invertebrates like anthropods, marine brown algae, etc., referred to them as 'polyelectrolytes' even though many of these studies did not conduct indepth chemical characterization to determine their ionic activity (Xiao *et al.* 2010). Natural coagulants are mostly either polysaccharides or proteins. These coagulants offer a broad range of applications such as biocompatibility, biodegradation, biological activity, non-toxicity, being non-allergenic, and having the ability for fiber and film formation (Dechojarassri *et al.* 2018).

Knowledge on the usage of alginates is still at the level of infancy. In recent works done by (Dechojarassri *et al.* 2018) and (Wang *et al.* 2020), alginate has been used as a secondary coagulant along with primary coagulants like aluminum sulfate, ferric chloride, etc. for color removal and DOC removal respectively. Aylin Devrimci *et al.* (2012) investigated the use of alginate for drinking water treatment in the turbidity ranges of 150, 80, and 10 NTU and achieved turbidity removal of up to 98% for 150 NTU turbidity levels. Alginic acid or alginate is a natural polysaccharide obtained from marine brown algae. Alginate can also be produced by certain bacterial strains such as *Azotobacter vinelandii* and *Pseudomonas aeroginosa*. The industrial applications of alginates are linked to the ability to retain water and its gelling, viscosifying, and stabilizing properties (Wang *et al.* 2020). Alginate could be regarded as a true block copolymer composed of homopolymeric regions of M and G, termed M block and G blocks respectively, interspersed with regions of alternating structure (Dechojarassri *et al.* 2018). The chemical structure of alginate is shown in Figure 1.

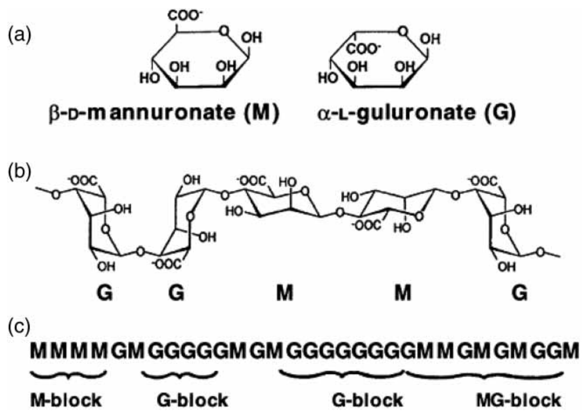


Figure 1 | (a) Alginate monomers; (b) chain conformation; (c) block distribution [22, 23].

The most useful and unique property of alginates is their ability to react with polyvalent metal cations, especially calcium ions to produce strong gels or insoluble polymers (Aneem *et al.* 2021). Alginate forms the so-called 'egg-box' structure with calcium ions. This gel formation property of alginate as well as other specific reactions with metal cations makes alginate an industrially important biopolymer with wide applications (Liu *et al.* 2021). The action of alginate is believed to be by one of the following mechanisms: charge neutralization along with bridging the gap between the particles, or by the formation of calcium alginate gel, which is especially more effective at high calcium concentrations (Yaseen & Scholz 2019; Zhao *et al.* 2020, 2012). Calcium alginate gel combines with particles and captures them at the stage of gel formation or after gel formation. Finally, the flocs are formed by the gel and the particle gets heavy enough and settles down (Devrimci *et al.* 2012; Zhao *et al.* 2012).

The present study intends to throw light on the natural material extracted from algae that is more predominant in Asian countries. This work focused on the turbidity value of 300 NTU, which may occur during floods in storm seasons. The main aim of this work is to check the effectiveness of alginate with a multivalent cation like calcium for the treatment of highly turbid water.

2. MATERIALS AND METHODS

Alginate used in this study belongs to the Lobachemie products and calcium chloride is from Merck and is of analytical grade. Both sodium and alginate were used as such in powder form. The concentration of alginate is varied from 2–10 mg/L, whereas calcium concentration is varied between 50–200 mg/L.

2.1. Preparation of turbid water

Synthetic turbid water was prepared by adding kaolin to the water to obtain the required turbidity. The clay (kaolin) was dried, powdered, and sieved through a 75 μ sieve. The predetermined amount of clay was added to the water and mixed up thoroughly using the flash mixer for uniform dispersion of clay particles for 30 min. The suspension was then allowed to stand for 3 h to allow for complete hydration of the kaolin and after that, the supernatant was taken care of and served as the sample for the experiments. The characteristics of the water used for the study were analyzed according to standard procedures and are given in Table 1.

Table 1 | Characteristics of tap water

S. No	Parameter	Value
1.	pH	7.55
2.	Conductivity	780 μS
3.	Total alkalinity as $CaCO_3$	285 mg/L
4.	Total hardness	245 mg/L
5.	Calcium hardness as $CaCO_3$	120 mg/L
6.	Calcium hardness as Ca^{2+}	48 mg/L
7.	Chlorides	114.9 mg/L

2.2. Coagulation study

Experiments were conducted on 500 mL laboratory-prepared turbid water samples. Each 500 mL synthetic turbid water sample was treated using different doses of calcium and alginate. Alginate in sodium form and calcium as $CaCl_2$ were used for the tests. Calcium and alginate were used in the powdered form throughout the process. Calcium concentrations varied between 50 to 200 mg/L whereas alginate concentrations varied between 2 to 10 mg/L. A standard jar test apparatus was used during the experiments, employing a mixing regime for each sample with the following sequence: 2 min rapid mixing at 160 rpm following the calcium dosing, 2 min rapid mixing at 160 rpm following the alginate dosing, 10 min mixing at 80 rpm and another 10 min slow mixing at 40 rpm and finally 30 min of settling. Turbidity values were measured in the sample after 30 min settling period using a Nephelometric turbidity meter. The operational parameters like the effect of paddle position, different forms of calcium ion, dosing order effect, settling time effect were studied.

3. RESULTS AND DISCUSSIONS

The turbidity of the water may fluctuate depending on the factors such as geological, environmental, structural, temperature, etc. For the experiments, working turbidity of 300 NTU was taken by adding the required amount of kaolin and the experiments were carried out in the laboratory.

3.1 Effect of the positioning of paddle

Fixing up of paddle position plays an important role in the reaction of alginate with colloidal particles. This was studied by fixing the paddle at 2 different positions: 1/3rd from top and 2/3rd from the top, and initial calcium concentration was 50 mg/L and the results are shown in Figure 2.

From Figure 2, it was observed that the effectiveness of alginate was maximum for the paddle position at the top. This is because the alginate is a material with very little density hence it will float on the surface of the water without any contact with colloidal particles. Hence Paddle was fixed at 1/3rd from the top throughout the process to ensure alginate getting mixed up thoroughly with calcium for gel formation.

3.2. Effects of different forms of calcium ion on turbidity removal

The property of the gelling ability of alginate with multivalent cations like calcium had been made use of for the process of coagulation. The concentration was focused also on the counterpart of the supplied calcium ion compound that it should not impart any undesirable characteristics like hardness, and so on, to the water. Experiments were carried out with two different forms of calcium compound (calcium chloride dihydrate and calcium chloride (fused)) with the dosage of 100 mg/L and their performance is shown in Figure 3.

From Figure 3, it is clear that calcium chloride dihydrate gave better results when compared with calcium chloride (fused). This might be due to the reasons that calcium chloride (fused) reacts vigorously in the

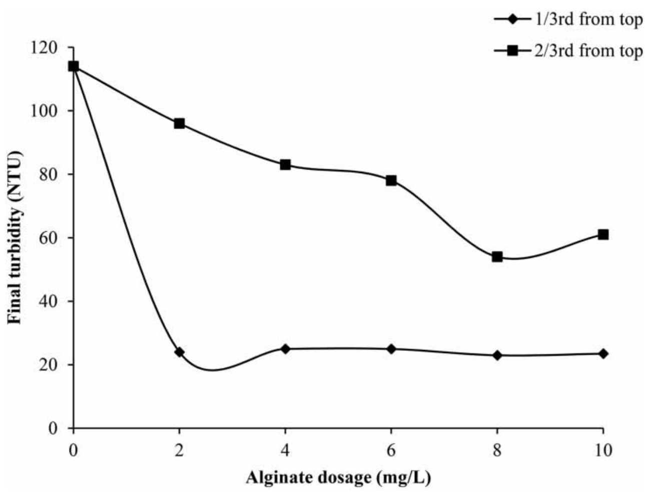


Figure 2 | Fixing up of Paddle position.

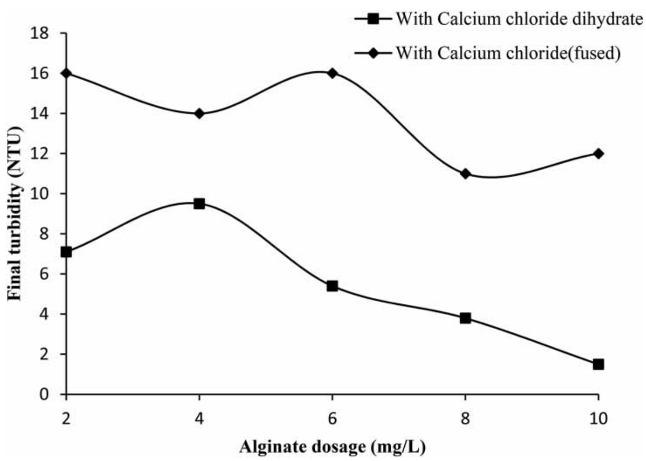


Figure 3 | Selection of form of calcium ion to be dosed.

atmosphere, measurement of the accurate quantity of chemical was quite difficult, and it also possesses handling difficulties. Hence, calcium chloride in the form of calcium chloride dihydrate was used for all the studies.

3.3. Effect of calcium on turbidity removal without alginate

To understand the prospective application of the alginate and to ascertain the effect of alginate, samples were dosed with calcium chloride alone in different concentrations without alginate and the results are tabulated in Table 2.

Table 2 | Effect of calcium on the turbidity removal without alginate

Calcium concentration (mg/L)	Initial turbidity (NTU)	Residual turbidity (NTU)	Turbidity removal rate (%)
200	300	133	55.7
150	300	118	60.7
100	300	106	64.7
75	300	121	59.7
50	300	154	48.7

Table 2 illustrates that calcium alone can remove the colloidal particles. Yet, by itself, calcium cannot decrease the turbidity to the reasonable values required for domestic purposes like drinking, etc. Thus it was anticipated that the use of alginate could improve the efficiency of coagulation. The mechanism of removal when calcium alone was added is supposed to be surface charge neutralization. The reason for the decrease in turbidity removal (Table 2) for higher calcium concentration could be due to the phenomenon of charge reversal.

3.4. Effect of dosing order between calcium and alginate

The dosing order plays an important role in deciding the efficacy of the coagulant, which is depicted in Figure 4. This was done in two sequences with a fixed calcium dosage of 100 mg/L: Dosing calcium first followed by alginate and dosing alginate first followed by calcium; the results are shown in Figure 4.

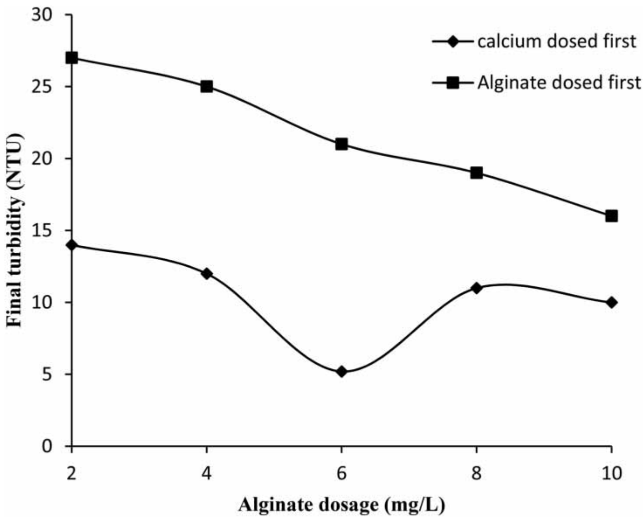


Figure 4 | Effect of dosing order between calcium and alginate.

From Figure 4, it is clear that the addition of calcium first provided good results when compared with the addition of alginate first. This is because the addition of alginate first will make the entire suspension more anionic. Moreover, the repulsive action of coagulant and colloidal particles could retard the calcium-alginate gel formation around the colloidal particles. The addition of calcium first will reduce the repulsive force between the colloidal particle and the coagulant and compression of the double layer will take place. Thus dosing order of calcium first followed by alginate was adopted throughout the process.

3.5. Effect of settling time on turbidity removal

The time taken for settling depends upon the density and shape of floc formed. To optimize this parameter, samples were allowed for the settling period of 10, 20, and 30 min and the final turbidity values are shown in Figure 5.

From Figure 5, it is clear that between 20 min and 30 min, not much change in residual turbidity was noticed. This is due to the reason that during mixing, rapid mixing was done at the higher speed of 160 rpm (larger velocity gradient), which will result in the formation of smaller and dense flocs, which can settle quickly. And since the turbidity taken was 300 NTU, the concentration of particles was more which could facilitate the enmeshment of colloidal particles in the settling flocs. It was observed that at the end of 10 min, only gel formation took place, and flocs formed were floating on the surface of the water. At the end of 20 min, all flocs had settled and observation of clear water was seen. Hence it was presumed that a settling time of 20 min is sufficient for this turbidity.

3.6. Effect of calcium and alginate concentration on turbidity removal

Calcium concentrations were fixed for each set during which alginate concentrations were decreased from 10 to 0 mg/L. Similar experiments were conducted for calcium concentrations of 200, 150, 100, 75, and 50 mg/L for the 300 NTU of initial turbidity to check the effect of calcium and alginate concentration on the effectiveness of coagulation. Immediately after the dosage of alginate, the strands of alginate solidified and denser flocs were formed.

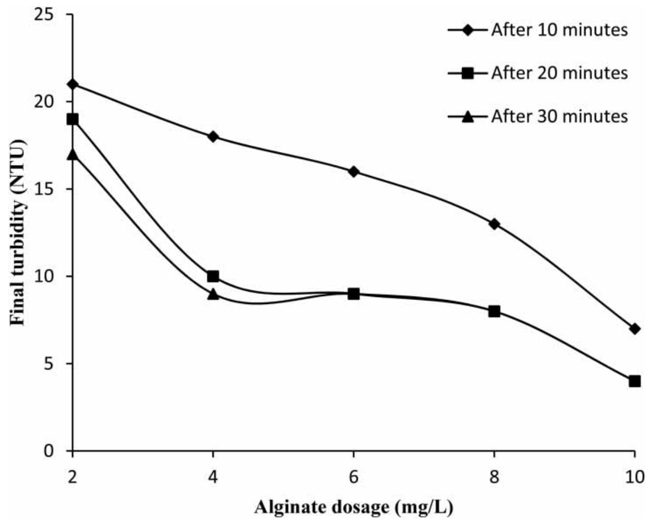


Figure 5 | Effect of settling time on the turbidity removal.

Figure 6 gives a plot of final turbidity values to alginate concentration for different calcium doses. Final turbidity got reduced to a minimum value of 1.8 NTU with an alginate concentration of 10 mg/L and calcium concentration of 200 mg/L, as is clear from Figure 6. For all the calcium dosages except 50 mg/L, the residual turbidity values were observed to be around 10 NTU. This is because the low concentration of positively charged calcium ions was not sufficient to neutralize the colloidal particles. If the material of alginate is to be used for drinking water treatment purposes, residual turbidity less than 5 NTU can be achieved with alginate dosage between 6 to 10 mg/L and calcium concentration greater than 50 mg/L. For 50 mg/L of calcium concentration, filtration can be done after coagulation/settling to achieve residual turbidity less than 5 NTU.

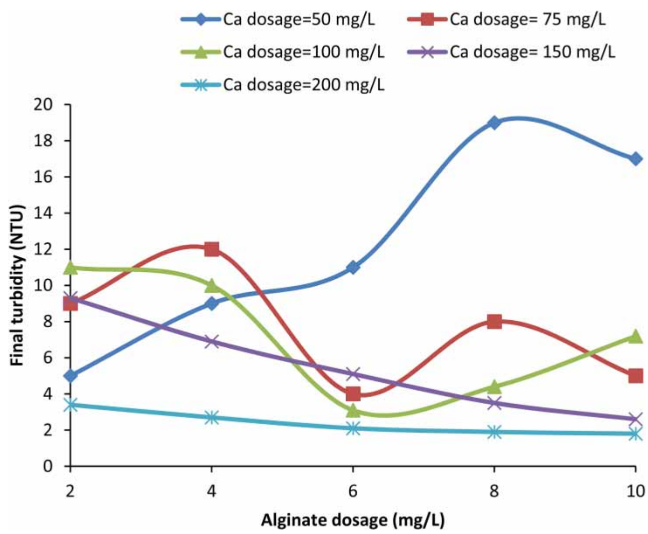


Figure 6 | Effect of change in calcium concentration for varied dosage of alginate on final turbidity.

The calcium-alginate gel possesses a strand-like structure with a lot of attraction sites for cations and finally, egg-box structure formation will take place for capturing colloidal particles. If the dosage of alginate becomes higher or lower, there is a greater possibility of restabilisation of the colloidal particles. The excess alginate gel sites will be further involved in the inter-particle bridge formation. Charge reversal could take place if the calcium ion concentration exceeds the level required for charge neutralization of colloidal particles and gel formation with alginate [10, 28, 29]. Optimization of alginate concentration can be done by selecting the lowest concentration of alginate that will correspond to the reasonable turbidity value. Because the lower the concentration of alginate, the lower will be the cost of treatment. Figure 7 summarizes the turbidity removal efficiency for different dosages of calcium and alginate. For all the studied ranges of alginate and calcium levels, the removal efficiency was observed to be more than 93%.

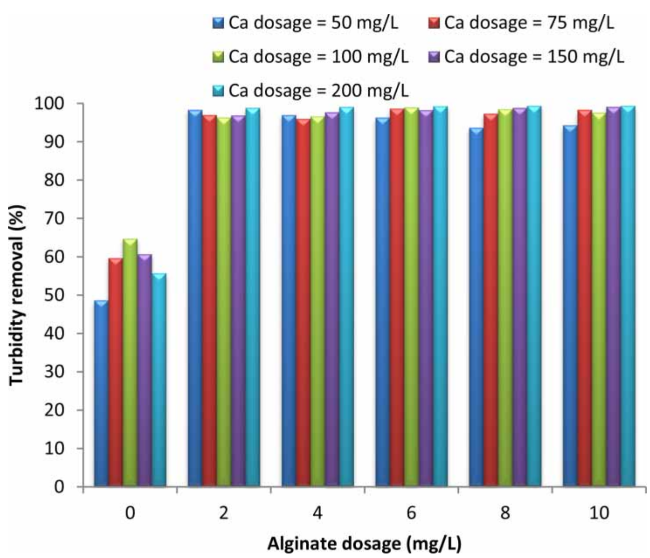


Figure 7 | Turbidity removal efficiency for different calcium dosages.

Generally, a polymeric coagulant doesn't change the pH of the water during treatment [29, 30]. The pH and conductivity of the treated water were checked and are shown in Figure 8. The pH of the sample before adding the coagulant was checked out to be 7.55 and after treatment, a negligible change in pH value was observed. The conductivity of the sample was also tested before and after treatment and it also remained constant, as is clear from Figure 8.

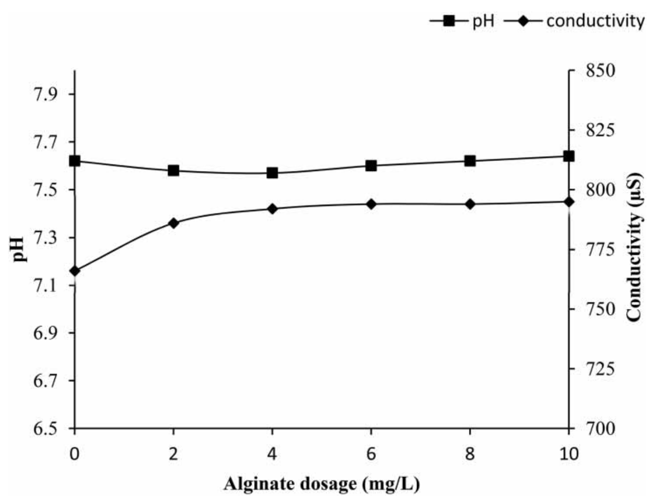


Figure 8 | Determination of pH and conductivity of the treated water.

Since a higher concentration of calcium chloride was added in the process, the fate of calcium was checked in the treated water to ensure the residual calcium levels were within the desirable values. The residual calcium level of water plays a significant role both in domestic as well as industrial purposes (incrustation of pipes). In the case of drinking water treatment, the excess calcium level will deteriorate human health and scale formation will occur in the discharge pipes.

It is evident from Figure 9 that for all the studied ranges of calcium and alginate, the residual calcium levels are within the desirable limit, which proved that alginate could be used for the treatment of water for drinking and industrial purposes.

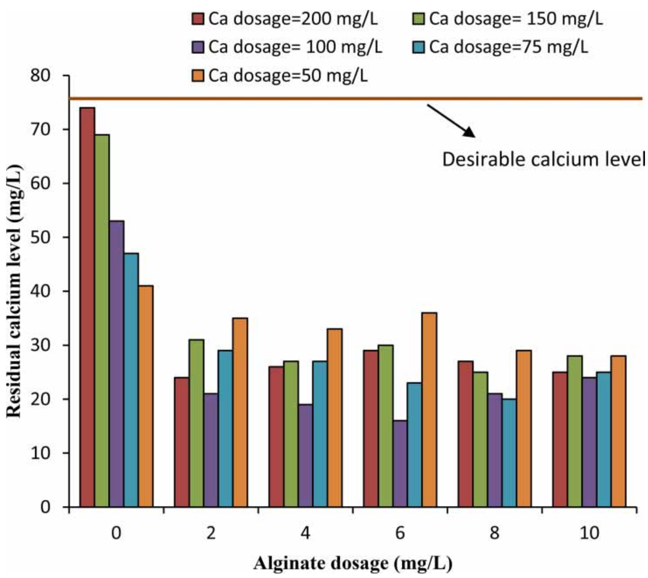


Figure 9 | Residual calcium level in the treated water.

4. CONCLUSION

The effectiveness of the material in treating water for removal of turbidity was investigated and in support of that various related tests had been conducted in the laboratory. Algal alginate is ascertained to be an effective coagulant for the removal of turbidity in water. The effectiveness of alginate mainly depends on the 'egg-box' structure formation that will occur in the presence of divalent ions, mainly calcium ions. This study reveals that alginate, a natural coagulant, has a greater potential for the removal of turbidity and made it suitable for drinking purposes.

Based on the results obtained from this study, it shows that the efficiency of the process is greatly dependent on the position of the paddle for mixing, different forms of the Ca ion, dosing order of Ca and alginate and settling time. Also, alginate proved to be a promising coagulant when compared with chemical coagulants because the latter will change the pH and electrical conductivity of water. The coagulation-flocculation process at alginate dosages of 6–10 mg/L and calcium dosages greater than 50 mg/L resulted in turbidity values less than 5 NTU, which are suitable for drinking water purposes. Under these conditions, removal of greater than 93% is achieved for all the cases and maximum removal of 99.3% is obtained. Concentration of alginate plays an important role in deciding the cost of treatment. For all the studied dosages of calcium and alginate, the residual calcium level was also found to be well within the desirable limit. Natural coagulants are eco-friendly and don't cause any harm to the consumers and thus they can be made use of in all applications of water treatment. Alginate is a potential coagulant if its extraction techniques are feasible and can replace the usual chemical coagulants. For the sustainability assessment of natural coagulants, further research on improving the capability of natural coagulants should be focussed on.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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