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Assessment of feasibility of natural coagulants in turbidity removal and modeling of coagulation process

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ABSTRACT

In this research, experiments were carried out in the laboratory to investigate the coagulation/flocculation behavior of natural coagulants, such as chitosan, *Moringa oleifera*, and cactus mucilage, for the treatment of synthetic turbid water of 314.4-NTU turbidity. A series of experiments were performed with varying the dosage of the coagulants. The result showed that the optimum dosages were 1.5, 80, and 40 mg/l for chitosan, *M. oleifera*, and cactus, respectively. IR spectral study demonstrates that the functional groups $COOH^-$ and OH^- were responsible for the coagulation activity. The existence of such functional groups along the chain of charged particles and polygalaturonic acid implies that chemisorption between charged particles and $COOH^-$ may occur. The presence of OH^- groups along its polymeric chain also infers possible intermolecular interactions which may distort the linearity of the chain. The study clearly indicates that the coagulation activity can be modeled by the Langmuir and Freundlich models. Experimental data fitted very well to both the models, which was proved from the R^2 values which are almost nearer to unity.

Keywords: Coagulation; Dose; Turbidity removal; Modeling; Chitosan; *Moringa oleifera*; Cactus

1. Introduction

Water is considered as a national resource of utmost importance. Water is vital to ensure the population's well-being and quality of life and to preserve the productivity of the agricultural sector. The quality of river or reservoir water is commonly characterized by the content of suspended solids (SS), colloidal particles, natural organic matter (NOM), and other soluble, mostly inorganic, compounds present in different concentrations. Therefore, when the river or reservoir water is intended for human consumption, an appropriate treatment process is usually considered as necessary to meet the respective drinking water standards. The production of potable water from most raw water sources usually entails the use of a flocculation/coagulation stage to remove turbidity in the form of suspended and colloidal material. One of the most important steps during the conventional treatment process is the coagulation/flocculation process, which serves mainly for the removal of SS (including colloidal microparticles) and NOM [1].

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Nearly all the colloidal impurities in natural waters are negatively charged, and hence these systems may be stable as a result of mutual electrical repulsions [2]. According to the coagulation and flocculation theory, colloidal destabilization can be achieved by adding cations that interact specifically with the negative colloids and reduce (or neutralize) their charge [3]. Alum is the most widely used coagulant in water treatment because of its proven performance and cost effectiveness. The use of alum as a coagulant in the treatment of water increases the aluminum concentration in treated water [4–6]. A high concentration of aluminum is also of concern because of its adverse effects on health. Aluminum intake in the body has been linked to several neuropathological diseases including percentile dementia and Alzheimer's disease [7–9].

A polyelectrolyte, in concurrence with a metal coagulant, improves coagulation by accelerating the process of coagulation. The coagulant aid reduces the necessity of alum and improves the physical characteristics of flocs. Most of the naturally occurring polyelectrolytes are of plant origin and the coagulants are derived from the seeds, leaves, pieces of bark or sap, roots, and fruit extracts of trees and plants. Many of the researchers throughout the world have performed studies on various natural coagulants and flocculants [10–18]. The natural macromolecular compounds derived from the cactus species and the low-cost anionic polysaccharide and polyelectrolyte derived from *Strychnos potatorum* are capable of reducing turbidity of water through flocculation process [19]. Natural coagulants of plant origin have been used for water purification for many centuries. *S. potatorum* (Nirmali seeds) was used as a clarifier between the fourteenth and fifteenth centuries. Sen et al. reported that seeds of the Nirmali tree were used to clarify turbid river water about 4,000 years ago in India [20].

Moringa oleifera powder has been reported to have the capability of reducing low and high turbidity in the water surface [21–28]. *M. oleifera* was used as a natural coagulant in a full-scale treatment trial at the water treatment works in Malawi. Turbidity values as high as 270–380 NTU were reduced to around 04 NTU, which are within the WHO [29] guidelines value, with the addition of the powder. A study by Zhang et al. on the performance of cactus as a coagulant in water treatment indicated that the coagulation performance of cactus was very much effective for turbidity removal; residual turbidity of less than 5 NTU could be obtained with initial turbidities from 20 to 200 NTU [15].

Natural coagulants have been reported to have several other advantages compared to synthetic coagulants, such as alum and ferric chloride, in that they produce much lower sludge volume and are safe for humans. Ghebremichael et al. investigated that the sludge produced from *M. oleifera* coagulated turbid water is only 20–30% that of alum [30]. Litherland [31], Katayon et al. [32], and Sanghi et al. [33] showed that the residue of alum in water may be carcinogenic. Natural coagulants are biodegradable and cost effective for developing countries since they can be locally grown and have a wider effective dosage range for flocculation of various colloidal suspensions [34].

The objective of the present study is to investigate the coagulation/flocculation potential of natural coagulants, such as chitosan, *M. oleifera*, and cactus mucilage, to remove the turbidity from synthetic turbid water prepared from local clay and to study the applicability of Langmuir and Freundlich models for the coagulation process.

2. Materials and methods

The materials used in this study are chitosan, seeds of *M. oleifera*, and Cactus Opuntia pads. Chitosan powder was purchased from Everest Edward, India sea foods, Thoppumpady Cochin, Kerala, *M. oleifera* seeds were collected/purchased from local farmers and the Cactus Opuntia pads were collected from the rural area of Kopargaon. The entire chemicals used in this study were purchased from Vijay Chemicals, Shrirampur Dist., Ahmednagar. The preparation of synthetic turbid water and coagulant powders is elucidated below.

2.1. Preparation of turbid water

Locally available natural clay was used to prepare synthetic turbid water by soaking the clay for 24 h in tap water and then blending it for 10 min. The suspension was washed through a 75 micron sieve. This was kept as a stock suspension for the preparation of different turbidities, such as low, medium, and high. A portion of the stock suspension was diluted with tap water and after 30 min of settling in a container, the supernatant was carefully decanted and desired turbidities of 34.2 NTU (low), 146.8 NTU (medium), and 314.4 NTU (high) were obtained by diluting it with tap water [17,18].

2.2. Preparation of chitosan solution

Chitosan (deacetylated chitin; poly-[1-4]-β-glucosamine) with 85.84% deacetylation prepared from crab shells was used for the preparation of coagulant solution. 100 mg of chitosan powder was weighted and mixed with 10 ml of 0.1 M HCl solution. Mixer was kept aside for about an hour to dissolve the chitosan powder. The dissolution process was slow and some amount of chitosan remained in the form of a thin gel even after this time. It was diluted to 100 ml with distilled water to obtain a solution containing 1.0 mg chitosan per milliliter of solution. The solution was prepared fresh before each set of experiments [35,36].

2.3. Preparation of *M. oleifera*

Pods and shells were removed manually and kernels were grounded in a domestic blender and sieved through 600-μm stainless steel sieve as per the methodology used by the pervious researchers [37,38]. The powder was then weighed and dissolved in distilled water to make a 500 mg/l solution. The solution was stirred for 30 min using a magnetic stirrer, and finally filtrated through a Whatman filter paper no. 40. A fresh solution was prepared every day in order to avoid aging effects.

2.4. Preparation of cactus coagulant

Cactus Opuntia pads were collected from rural area of Kopargaon, washed thoroughly with tap water, and stored in the refrigerator at 4˚C. The outer layer was removed with the help of a knife, and subsequently inner pads were sliced into small pieces to facilitate drying. The sliced cactus was then dried in oven for 8 h at 80˚C. The dried cactus was ground into a fine powder using a domestic mixer, and subsequently sieved to sizes 53–106 μm [15,39].

2.5. Experimental work

Sedimentation jar tests were carried out to determine the coagulation properties of the natural coagulants. One beaker was used as control and in other beakers coagulants of varying dosages were were added. Jar tests were conducted on 500-ml synthetic turbid water samples. Following the addition of the coagulants dosages (chitosan, *M. oleifera*, and cactus) the samples were subjected to rapid mixing at 100 rpm for 1 min and a slow mixing step at 30 rpm for 30 min. The stirrer was then switched off and the floc was allowed to settle undisturbed [40] for 30 min. Fig. 1 show the jar test apparatus used for the experimentation. The samples for the residual turbidity measurement were withdrawn using a pipette from a height of 5 cm below the surface of each beaker, and residual turbidity was measured. Fig. 2 shows jars showing the removal of turbidity after flocculation for various dosages of coagulants. Turbidity measurement was carried out by Systronics Turbidity meter type 131, Systronics, India. The pH value of the suspension was measured using an Elico digital pH meter model 121. The chemical analyses were conducted as per standard methods. The procedure conformed to that described in Standard Methods for the Examination of Water and Wastewater [41].

Fig. 1. Jar test apparatus used in the experimentation.

Fig. 2. Jars showing removal of turbidity for various dosages of coagulants.

3. Result and discussion

3.1. FT-IR spectrum studies

IR spectra graphs for chitosan, *M. oleifera*, and cactus were taken between the wave numbers 4,000 and 1,000 per cm and are presented in Figs. 3–5.

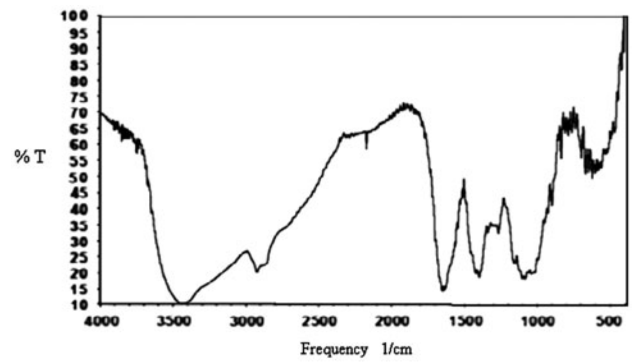

Fig. 3. FT-IR for chitosan powder.

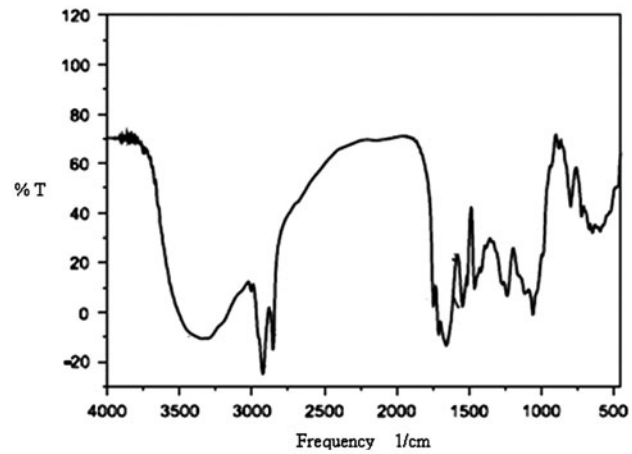

Fig. 4. FT-IR for *M. oleifera* seeds powder.

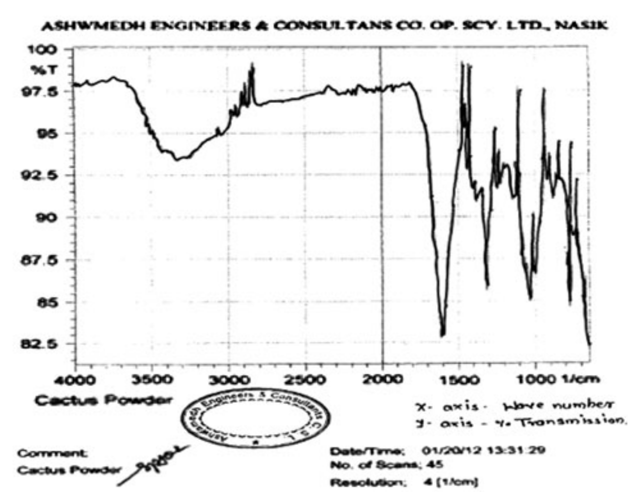

Fig. 5. FT-IR for Cactus Opuntia powder.

3.1.1. IR spectra for chitosan powder

FT-IR spectra for the chitosan powder were analyzed, and the results showed that both the $CONH_2$ and NH_2 groups of chitosan are slightly cross-linked with a sodium polyphosphate molecule (Fig. 3) [42]. As can be seen from the IR spectrum of chitosan, a peak indicating P=O stretching at 1,217 per cm appears [43].

The amine groups on chitosan bind metal cations at pH close to neutral. At low pH, chitosan is more protonated, and therefore it is able to bind anions by electrostatic attraction [44].

3.1.2. IR spectra for M. oleifera seed powder

The functional groups present in *M. oleifera* were characterized by Fourier transform infrared (FT-IR) spectrometer. The spectral range varied between 4,000 and 500 per cm with 28 scans in a resolution of 4 per cm.

The broadband centered at 3,420 per cm in the IR spectra in Fig. 4 can be attributed to O–H stretching of the connection in this protein, fatty acids, carbohydrates, and the lignin units. The peaks that appear in 2,923 and 2,852 per cm correspond, respectively, to the asymmetric and symmetric stretching of the connection of the C–H in CH_2 group. Due to the high intensity of these bands, it is possible to assign them to the predominantly lipid component of the seed that is present in high proportion similar to the proportion of the protein. In the region of 1,800–1,500 per cm, there are a number of overlapping bands between 1,750 and 1,630 per cm. This set can be recognized to the C=O connection stretching. Owing to the heterogeneous type of the seed, the carbonyl group may be related to different neighborhoods as part of the fatty acid portion of the lipid and protein portion of the amides. The carbonyl that appears due to the lipid component in 1,740 and 1,715 per cm, which can be seen in the spectrum as a small peak at the shoulder part of the main band at 1,658 per cm, was assigned to the carbonyl amides in the protein portion [45].

3.1.3. IR spectra of Cactus Opuntia powder

Fig. 5 represents the IR Spectra for Cactus Opuntia powder. The weak and broad vibrations at wave numbers 1,700 per cm and 1,600 per cm in the cactus spectrum indicate the presence of carboxylic acid groups and are negatively charged. The high-coagulation capability of cactus is most likely attributed to the presence of mucilage which is a viscous and complex carbohydrate. Previous studies have established that mucilage in Cactus Opuntia contains carbohydrates, such as L-arbinose, D-galactose, L-rhamnose, D-xylose, and galaturonic acid [46,47]. Miller and coresearchers

[48] recently reported that galacturonic acid is possibly the active ingredient that affords the coagulation capability of Opuntia. Opuntia operates predominantly through a bridging coagulation mechanism where solution particulates do not directly contact one another, but are bound to a polymer like material that originates from the cactus species.

Though not extensively reported in open literatures, it is highly possible that galaturonic acid [as a major constituent of pectin in plants] exists predominantly in polymeric form (polygalaturonic acid) that provides a "bridge" for particles to adsorb on. Relevant dominant molecular interactions associated with adsorption and bridging in coagulation are shown in Fig. 5. The acid structure evidently indicates that it is nonionic. The polygalaturonic acid structure evidently indicates that it is anionic due to partial deprotonation of carboxylic functional group in aqueous solution. The existence of such functional groups along the chain of charged particles and polygalaturonic acid implies that chemisorption between charged particles and $COOH^-$ may occur. The presence of OH^- groups along its polymeric chain also infers possible intermolecular interactions that may distort the linearity of the chain.

As per the study of many researchers and the results of this research state, the presence of OH^-, $COOH^-$, galaturonic acid, the connection of the C–H in CH_2 group, and the $CONH_2$ and NH_2 groups at different wave numbers are the main causes for the effective coagulating capacity of the natural coagulants studied.

3.2. Assessment of the effect of chitosan dosing on turbidity removal

Results on optimization of chitosan dosages for turbid water of 314.4 NTU turbidity are presented in Fig. 6(a) and (b). The behavior of turbidity after coagulation by chitosan was investigated at pH adjusted to 7.0–7.5. According to previous investigations [49,50], the results of this study show that chitosan was found to be effective in this pH range for turbidity removal. The jar test experiments were run with chitosan as a coagulant in dose ranging from 0.5 to 2.5 mg/l. The optimum dose of chitosan was found to be 1.5 mg/l, above which the suspensions showed a tendency to restabilize.

The percentage of turbidity removal was increased with increase in chitosan dose, and the maximum removal takes place at the dose of 1.5 mg/l. At optimum dose condition, turbidity removal efficiency of chitosan was 95.3% for the initial turbidity of 314.4 NTU. The result of this study agrees with previous studies. Fig. 6(a) and (b) illustrate the variation in residual turbidity with increase in dosages.

3.3. Assessment of the effect of *M. oleifera* dosing on turbidity removal

The results of *M. oleifera* as a coagulant using jar test are presented in Fig. 7(a) and (b). The results indicated that at the optimum dosage of 80 mg/l, *M. oleifera* dose reduced the turbidity from 314.4 to 16.3 NTU with removal percentage of 94.8%. The pH values revealed that *M. oleifera* did not significantly affect the pH of the treated water, which remained almost constant at 7.2–7.5 for all dosages applied [22,51].

3.4. Assessment of the effect of Cactus Opuntia dosing on turbidity removal

Pads of Cactus Opuntia. were evaluated for turbidity removal from synthetic turbid water of 314.4-NTU turbidity. Results for the same are presented in Fig. 8

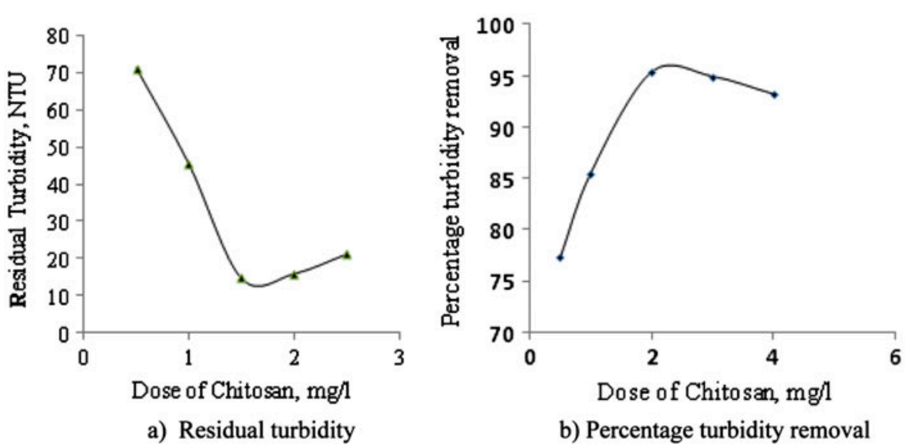

Fig. 6. Performance of chitosan at varying dosages.

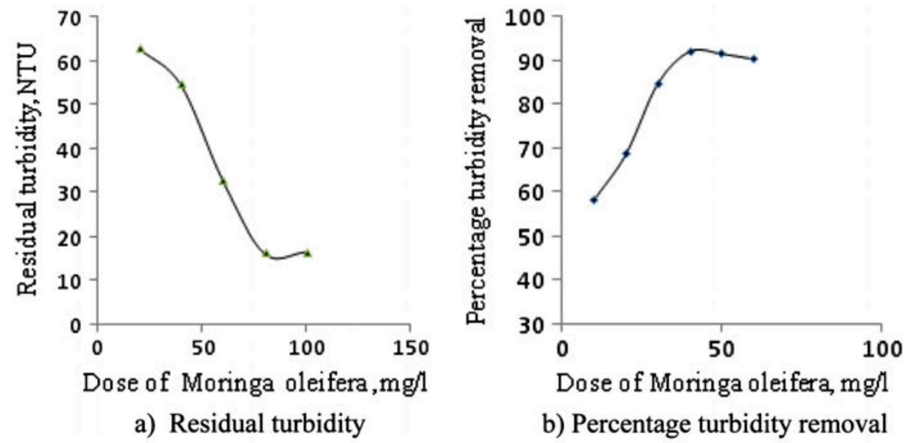

Fig. 7. Performance of *M. oleifera* at varying dosages.

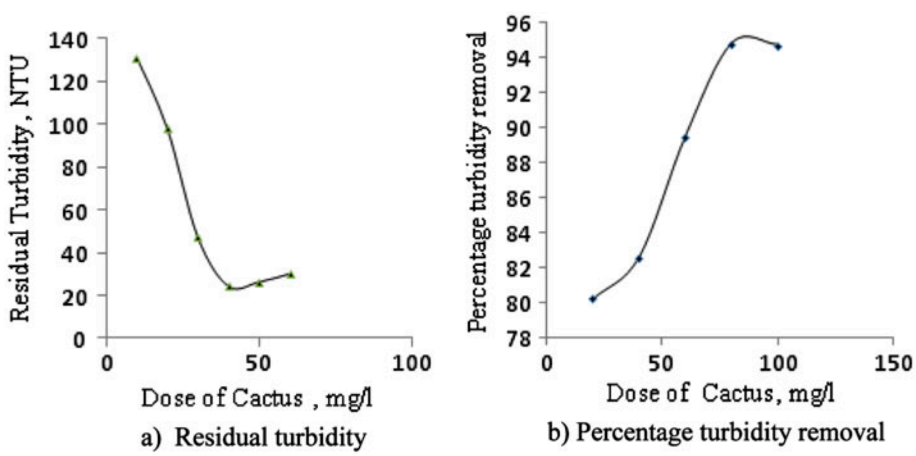

Fig. 8. Performance of cactus mucilage at varying dosages.

(a) and (b). The residual turbidity achieved was 24.5 NTU for optimum dose of 40 mg/l. The maximum removal efficiency achieved was 92.2%. The high coagulation capability of Opuntia is most likely attributed to the presence of mucilage, which is a viscous and complex carbohydrate stored in the inner and outer pads of the cactus that has great water retention capacity [46]. Miller et al. recently reported that galacturonic acid is possibly the active ingredient that affords the coagulation capability of Opuntia sap. They suggested that Opuntia Sap. operates predominantly through a bridging coagulation mechanism. The presence of galacturonic acid was also reported by Japanese researchers. All these studies point to the importance of galacturonic acid which possibly acts as one of the major active coagulating agents in plants [48].

3.5. Modeling of coagulation process

The physical system turbidity coagulant may be ruled by adsorption-like relationships. Taking into consideration the need for characterizing how this coagulant works on turbidity removal, a model must be proposed to determine the best way of scaling-up this new treatment agent. To this end, one must define a target variable that should be referred to the removal efficiency for the specific contaminant [52]. Consequently, it must link the removal percentage and the amount of coagulant. Coagulation and flocculation processes are rather difficult to model mathematically, due to two main reasons: (1) the complex nature of the phenomenon, which implies physico-chemical interaction molecule–molecule (van der Waals and hydrogen bridges forces) [53] and (2) the fact that the intrinsic composition of the organic material that forms the flocculant active principle is not completely known. The parameter extensively used in adsorption processes is the adsorption capacity, q. Our working hypothesis was that contaminant removal by coagulation and flocculation occurs in two stages. First, there is destabilization of colloids which may be governed by chemical interactions between molecules of the coagulant (cationic, positively charged) and of the contaminant (anionic, negatively charged). Then, once the coagulant–contaminant complex is formed, flocs begin to grow by sorption mechanisms. This should be the controlling stage, so that the entire process can be simulated as an adsorption phenomenon. Previous studies have found the coagulation capacity q to be a suitable evaluation parameter [52]. Although other hypotheses are feasible [54], the main adsorption (and coagulation) model is the one that Langmuir presented in the early years of the twentieth century [55]. The fact that it theoretically deduced makes it still feasible and appropriate. Probability of adsorption rate is proportional to the number of active sites, whereas probability of desorption is proportional to the number of already occupied sites. Those probabilities are related to the strength of the interaction between the adsorbent surface and the adsorbate.

In this section of study, the turbidity removal (high turbidity, 314.4 NTU) for various dosages of coagulants after jar test was reckoned for model analysis. To validate the results of coagulation/flocculation achieved, modeling was carried out using Langmuir and Freundlich isotherms.

3.5.1. *Langmuir and Freundlich modeling for high turbid water with chitosan as a coagulant*

Fig. 9(a) and (b) shows the Langmuir and Freundlich models which were fitted for the results obtained when chitosan was used as a coagulant for high turbid water. The R^2 value obtained was 0.799 as per Langmuir model and 0.929 as per Freundlich model. This clearly indicates that the data fitted well to both the adsorption models as the value of R^2 is almost equal to and greater 0.8.

3.5.2. *Langmuir and Freundlich modeling for high turbid water with M. oleifera as a coagulant*

Fig. 10(a) and (b) represents the Langmuir and Freundlich models which were fitted for the results obtained when *M. oleifera* was used as a coagulant. The R^2 value obtained was 0.869 as per Langmuir model and 0.9369 as per Freundlich model. This clearly indicates that the data fitted well to both the adsorption models since the value of R^2 is almost nearer to 1.

3.5.3. *Langmuir and Freundlich modeling for high turbid waters with Cactus Opuntia as a coagulant*

Fig. 11(a) and (b) demonstrates the Langmuir and Freundlich models which were fitted for the results obtained when Cactus Opuntia was used as a coagulant. The R^2 value obtained was 0.9016 as per Langmuir model and 0.9724 for Freundlich model. This clearly indicates that the data fitted very well to Freundlich model when compared to Langmuir

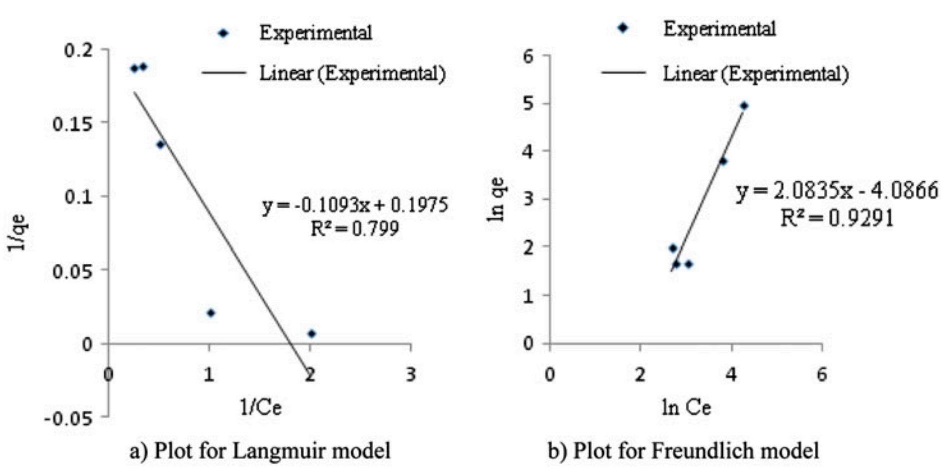

Fig. 9. Langmuir and Freundlich model for coagulation with chitosan as the coagulant.

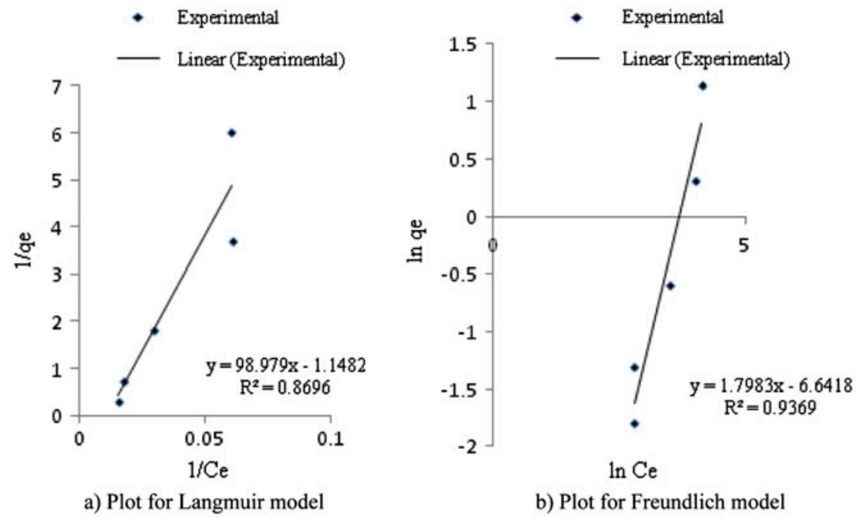

Fig. 10. Langmuir and Freundlich models for coagulation with *M. oleifera*as coagulant.

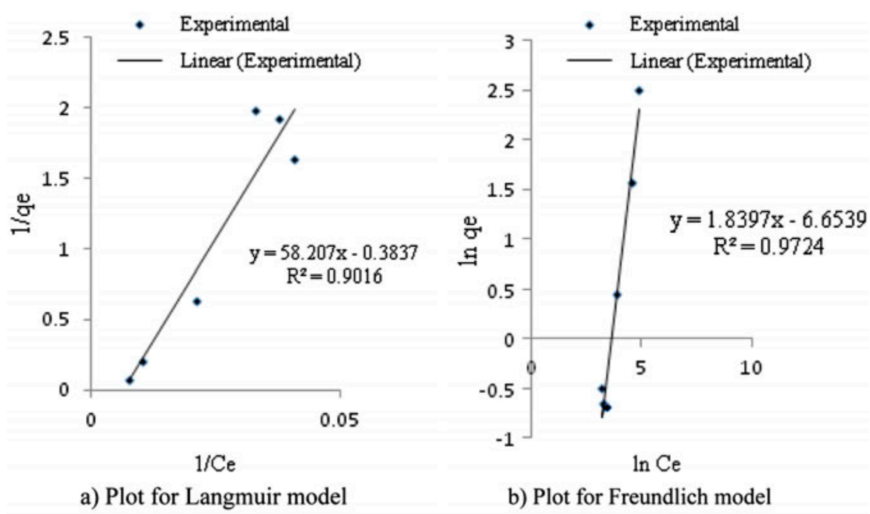

Fig. 11. Langmuir and Freundlich models for coagulation with Cactus Opuntia as the coagulant.

model, since the R^2 value is almost nearer to unity in the case of Freundlich model.

The results of this study shows that the predominant coagulation mechanism for cactus is adsorption and bridging, whereby clay particles do not directly contact one another but are bound to a polymer-like material. Adsorption may occur through hydrogen bonding or dipole interactions. It is likely that natural electrolytes from within the cactus pads are particularly divalent cations, which are known to be important for coagulation with anionic polymers facilitating adsorption [48].

Table 1
Modeling of coagulation process for chitosan, *M. oleifera*, and Cactus Opuntia through the Langmuir and Freundlich models for turbidity removal

Name of the coagulant	R^2 value as per Langmuir model	R^2 value as per Freundlich Model	Best fitted model
Chitosan	0.799	0.929	Freundlich models
Moringa oleifera	0.869	0.969	Langmuir and Freundlich models
Cactus Opuntia	0.901	0.972	Langmuir and Freundlich model

Table 1 gives the summary of the results. From the above results, we can observe that chitosan could be best modeled by Freundlich model (R^2 value is 0.929) for high turbid water. *M. oleifera* can be modeled by both the Langmuir and Freundlich models, with R^2 values of 0.869 as per Langmuir model and 0.969 for Freundlich models. Cactus Opuntia could also be best modeled by the Langmuir and Freundlich models, as the R^2 value obtained by the Langmuir model is 0.901 and with the Freundlich model it is 0.972, which are almost nearer to 1. The results are in line with the results of Md. Ridwan Fahmi et al. [56] and Mataka et al. [57], they found R^2 value of 0.94 for Langmuir model and 0.97 for Freundlich model when *M. oleifera* was used as a coagulant/adsorbent.

4. Conclusions

The purpose of this study was to investigate the capability of natural coagulants, chitosan, *M. oleifera*, and Cactus Opuntia, for turbid water clarification. The experiments conducted confirm the positive coagulation properties of the investigated natural coagulants. Among them, chitosan and M. oleifera were the most efficient, expressing the highest coagulation activities, about 95.35 and 94.8%, respectively, for high turbid water (314.4 NTU).

(1) From the plant extract testimony and laboratory analysis, natural coagulants derived from chitosan, *M. oleifera*, and cactus have been identified to be suitable for treating turbid water.
(2) A large reduction in turbidity is achieved at optimal dosage conditions. All the coagulants have performed better for high turbid water.
(3) With the addition of natural coagulant dosage, there has been minor change in the pH of treated water, but there is no necessity for pH correction. Quality of sludge with natural coagulants was observed to be thick and settles more rapidly than sludge with conventional coagulants.
(4) Only a small increase in the organic matter content in the water after coagulation with these crude extracts was found and it was below the maximum allowable concentration according to WHO directive and that amount, possibly, will also be removed by filtration.
(5) The interaction system between the coagulant and turbidity removal can be modeled according to Langmuir as well as Freundlich model and best-fit results were found for the cactus coagulant followed by M. oleifera and chitosan. The R-squared values for the Langmuir model are 0.901, 0.869, and 0.901, while for the Freundlich model they are 0.972, 0.969, and 0.929, respectively, for cactus, *M. oleifera*, and chitosan.

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