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Novel natural coagulants for water clarification

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ABSTRACT

The aim of the present study was to develop new flocculating agents in the form of beads and to check their capability for water clarification. To this end mucilage was isolated from three different plant species i.e., *Portulaca oleracea* L. (aerial parts), *Hibiscus rosa-sinensis* L. (leaves), *Aloe barbadensis* Miller (leaves). Mucilage-sodium alginate beads were prepared by entrapment method. Jar test experiments were conducted separately with 6-60mg/L dose range of mucilage powder and 150-1500mg/L dose range of beads that contained 6-60mg/l of entrapped mucilage using water samples with turbidity 35 and 450 (NTU). Beads were used for 3 repeated cycles to check the effectiveness in turbidity removal. *Portulaca* and *Aloe* beads were found to be more effective in water clarification when compared with that of *Hibiscus rosa-sinensis*.

KEYWORDS

Water clarification;
Portulaca;
Hibiscus;
Aloe vera;
Mucilage-sodium alginate beads.

INTRODUCTION

Water clarification and filtration are important steps in the treatment of source water for drinking and industrial applications. To meet the requirements of potable and industrial water, the immediate need is water clarification. Reduction in turbidity of water is nothing but the clarity. Turbidity is a measure of cloudiness of water which is due to the presence of suspended or colloidal particles. Although turbidity itself is not a major health concern, high turbidity can interfere with disinfection process and provide a medium for microbial growth. This is the reason why turbidity is one of the main water quality parameter that will be measured during the water treatment and is a very useful tool to assess the drinking water quality, in general and in monitoring the public water delivery systems. World Health Organization (WHO) has set the standard turbidity value for drinking water as 5 Nephelometric Turbidity Units (NTU) and industrial water as 10 NTU[1].

Turbidity in raw water supplies is removed by various methods to make water suitable for domestic purposes and most industrial requirements. The particles with higher density than water should ultimately settle due to gravitational force. Small particles with density close to water such as bacteria and colloidal particles may not settle and remain suspended in water. Therefore, collecting tiny particles together into a mass of large size particle makes them settle more rapidly, and it is a necessary step for their removal by sedimentation[2] (HDR Engineering, Handbook of Public Water System, 2001). Turbidity removal can be done by coagulation, flocculation, sedimentation and filtration[3]. Coagulation is the process of destabilization by charge neutralization this can be accomplished through the addition of inorganic salts. On the other hand flocculation is the process of forming agglomerates of small particles to settle in the solution.

Clarification of water using coagulating agents has been practiced from ancient times. Aluminium sulphate (Alum), ferric chloride ($FeCl_{3)}$, Ferrous sulphate ($FeSo_4.7H_2o$) Polyaluminium chloride[4-6] and organic synthetic polymeric coagulant aids[7,8] are the most extensively used coagulants in water turbidity removal. Several studies reported that high grades of Aluminium in water are associated with the occurrence of the Alzheimer's disease[9,10]. The monomers of some synthetic polymers on the other hand are mostly non-biodegradable, highly expensive and may be toxic[11,12]. For this reason many researchers have explored the use of plant-based substances as flocculants, e.g. *Moringa oleifera*[13,14], *Opuntia* species[15], *Coccinia indica*[16], Okra, Fenugreek[17] and seeds of Red Sorrela[18], Nirmali[19] as alternatives to synthetic coagulants.

Mucilages are glutinous, transluscent amorphous substances and they are the polymers of monosaccharides or mixed monosaccharides and many of them are combined with uronic acids.

In the present study *Hibiscus rosa-sinensis Linn.* (Leaves), *Aloe barbadensis Miller.* (Leaves) and *Portulaca oleraceae Linn.* (Aerial parts) were selected for the isolation of mucilage and to evaluate their application in water turbidity removal. *Hibiscus* is a genus of flowering plants in the mallow family, Malvaceae. *Aloe vera* is also known as the *true aloe* or *medicinal aloe*, is a succulent plant, belongs to the family Liliaceae, *Portulaca oleracea* (Common Purslane, Pigweed, Little Hogweed, or Pusley), is an annual succulent in the family Portulacaceae. Mucilage of *Hibiscus Rosa-sinensis* contains L rhamnose, D galactose, D galactouronic acid, and D glucuronic acid. Mucilage of both *Hibiscus* and *Aloe* leaves were found for their applications as sustained releasing agents in drug formulations[20,21]. The major composition of aloe gel consists of a mucilage of polysaccharide substances. Most of these polysaccharides are glucomannans, mannans, or pectins having a range of molecular weights. The major carbohydrate fraction isolated from aloe gel, "acemannan", consists of a polydispersed â-1, 4-linked acetylated mannan interspersed with O-acetyl groups[22]. *Aloe* has found wide applications in traditional medicine[23-25] and cosmetic[26] system of India. *Portulaca oleracea* is an edible vegetable plant and its mucilage has a complex arabinogalactan structure similar to Gum Arabic and contains d-galactose:l-arabinose:d-galacturonicacid:d-xylose:l-rhamnose at ratio of 40:20:5:1:31 and it has been known for its mucoadhesive properties[27,28].

The objective of present study was to develop mucilage-alginate beads using sodium alginate (binding agent) and calcium chloride to enhance the stability and investigate the effectiveness of developed beads with that of un-entrapped mucilage in water clarification. Sodium alginate is the sodium salt of alginic acid derived from green algae. It does not dissolve in water, but it absorbs water. Its unique property is to form gels when combined with divalent ions like calcium. This reaction is widely used in most of the industries to stabilize many substances. The mucilage of three plants were particularly used due to their viscous nature, which will be the suitable property for adsorption of suspended particles in turbid water.

The coagulation test results obtained by beads were compared with the results that obtained by combination of widely used coagulants i.e., Alum and *Strychnos potatorum Linn.* (Nirmali) seeds[19].

MATERIALS AND METHODS

All the three plants were collected from in and around Hyderabad, India. Sodium alginate, calcium chloride and Kaolin clay were purchased from Sigma Aldrich Chemicals, India. Systronic Digital Nephelo-turbidity meter 132 was used for turbidity measurement.

Isolation of mucilage

Mucilage was isolated by the method described by[20].

The fresh leaves of plants were collected and washed with water. The leaves were crushed and soaked in water for 5–6 h, boiled for 30 min and left to stand for 1 h to allow complete release of the mucilage into the water. The mucilage was extracted using a multi layer muslin cloth bag to remove the marc from the solution. Acetone of three times the volume of filtrate was added to precipitate the mucilage. The mucilage was separated, dried in an oven at 35°C, collected, grounded, passed through a # 80 sieve and stored in a desiccator at 50 °C till use.

Immobilization of mucilage

Immobilization of mucilage into calcium alginate beads was carried out at 0.5, 1, 2, 3 and 4% concentrations according to the modified method[29,30].

Different concentrations of sodium alginate were used to optimize the concentration. 50ml of 0.5, 1.0, 2.0, 3.0 and 4.0 % solutions of sodium alginate were prepared by dissolving 0.25gm, 0.5gm, 1.0gm, 1.5gm, 2.0 gm of sodium alginate in 50ml of distilled water under stirring. 50ml of 0.5, 1.0, 2.0, 3.0 and 4.0% solutions of mucilage were also prepared by dissolving 0.25, 0.5gm, 1.0gm, 1.5gm, 2.0gm of mucilage powder in 50ml of distilled water under stirring. Prepared sodium alginate solutions were mixed with an equal volume of mucilage solution to get the final concentration of sodium alginate 0.25, 0.5, 1.0, 1.5, 2.0% respectively. Different concentrations of sodium alginate mixed with mucilage solution were added separately from a height of nearly 1-2cm into excess of $CaCl_2$ solution. For 10ml of sodium alginate-mucilage mixture, 100ml of $CaCl_2$ solution was required. The formed beads were left to harden for 2h in the same $CaCl_2$ solution. The beads were washed with Tris- buffer (7.5 P^H, 0.05M) to remove the loosely bound mucilage and allowed to dry the beads at room temperature.

Preparation of synthetic turbid water

Kaolin clay synthetic turbid water was used in jar test experiments.

Turbid water for coagulation tests was prepared by adding 10 gm of kaolin to 1L of tap water. The suspension was stirred for 1 h to achieve uniform dispersion of kaolin particles, and then it was allowed to remain for 24 hrs for complete hydration of the particles. This suspension was used as the stock solution. Turbid water of required NTU was prepared by dilution of stock suspension to the desired volume using tap water just before the coagulation test. 35 and 450 NTU solutions were prepared representing low and high ranges of turbidity.

Jar test experiments

Experiments were carried out with dried mucilage powders as well as immobilized beads at various concentrations according to the method[18]. 500 ml of aliquots of turbid water in 1L beakers were placed on a magnetic stirrer. Mixing was started at 100 RPM. After addition of requisite doses of coagulant, the mixing was continued for 1 min to ensure proper dispersion of coagulant in water. The stirring was continued for 9 min at a speed of 40 RPM. At the end of the mixing period, the beakers were removed and the contents were allowed to settle for 1 h. 30 ml of settled water sample was withdrawn for turbidity measurements.

Several batches were run with varying concentrations of coagulants. 6 mg/l, 12 mg/l, 20 mg/l concentrations of un-entrapped mucilage and 150 mg/l, 300 mg/l, 500 mg/l concentrations of dry as well as wet mucilage beads of above three plants were used for low water turbidity range (35 NTU). In high water turbidity (450 NTU) experiments 20 mg/l, 40 mg/l, 60 mg/l dosages of un-entrapped mucilage, 500 mg/l, 1000 mg/l, and 1500 mg/l dosages of mucilage beads were used. The optimum concentrations chosen based on highest observable efficiency. Experiments were also conducted with blank calcium alginate beads to ascertain their role in turbidity removal. All the studies were conducted in triplicate.

STATISTICS

Statistical data was analyzed using statistical parametric method (t-test). All the P-values were verified at 0.05 significant levels. Micro cal Origin 6.0 software was used for data analysis.

RESULTS AND DISCUSSION

Mucilage-alginate beads prepared from 0.25, 0.5, 1.0% mixture were more fragile and they lost their stability under stirring. Beads prepared from 1.5% mixture were partly stable and beads prepared from 2.0% mucilage-sodium alginate mixture were quite stable even under agitation condition.

In low water turbidity experiments, residual turbidities went on decreasing by increasing the coagulant dose initially up to 12mg/l beyond that concentration % decrease in turbidity levels was reduced in case of all the three plants (Figures 1 & 2). Treatment with a dose of 12 mg/l of dry mucilage powder reduced the turbidity to 5.4 NTU (84.5% turbidity removal) and 5.5 NTU (84.2% turbidity removal) with *Portulaca* and *Aloe* respectively, whereas it was 7.2 NTU (79.4% turbidity removal) with *Hibiscus* mucilage. The results obtained by 300 mg/l dose of dry and wet mucilage beads were also in the same range of NTU that was obtained by dry mucilage powder. The turbidity without using coagulant that is by simple gravitational force was found to be 12 NTU and the percentage removal of turbidity was 65.7%. This was far beyond the turbidity value obtained by *Hibiscus* mucilage, which was found to be least effective in turbidity removal among the three plants.

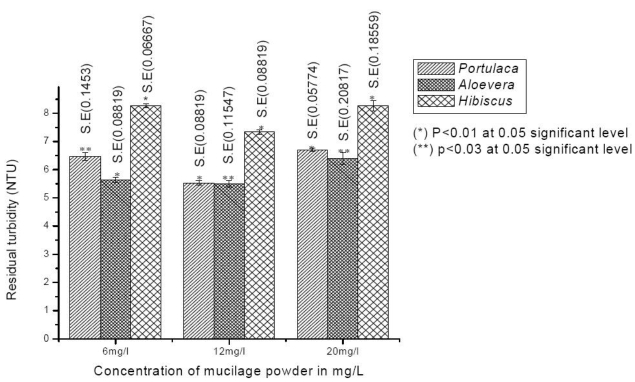

Figure 1 : Water clarification by mucilage powder of three plants in low turbid (35 NTU) Water in 1h.

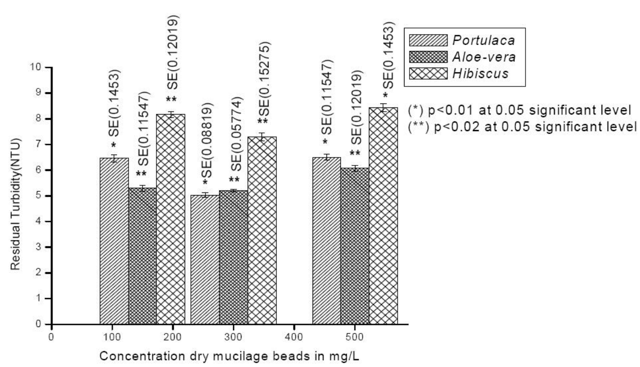

Figure 2 : Water clarification by mucilage-alginate beads in low turbid (35 NTU) water in 1h.

In high water turbidity experiments, residual turbidities went on decreasing by increasing the coagulant dose as in low water turbidity experiments. Turbidity removal was found to be effective when high turbid water was tested (Figures 3 & 4). Treatment with 40 mg/l of dry mucilage powder and 1g/l of dry mucilage beads reduced the turbidity to 13.5 NTU (97% turbidity removal) with *Portulaca* and *Aloe* respectively, whereas it was 22.5 NTU (95% turbidity removal) with *Hibiscus* mucilage. The turbidity was reduced to 42 NTU without using coagulant and the percentage removal of turbidity was 90.6%. This is also far beyond the turbidity value obtained by *Hibiscus* mucilage. Turbidity values obtained by using *Aloe* and *Portulaca* mucilage in different forms were found to be comparable with that of *Nirmali* seeds; *Hibiscus* was not as effective as the other two plants.

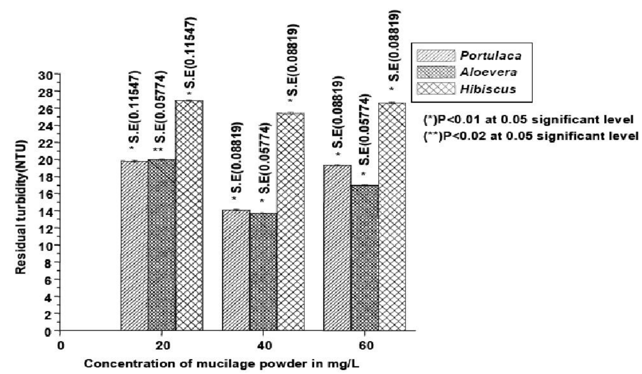

Figure 3 : Water clarification by mucilage powder in high turbid (450 NTU) water in 1 h.

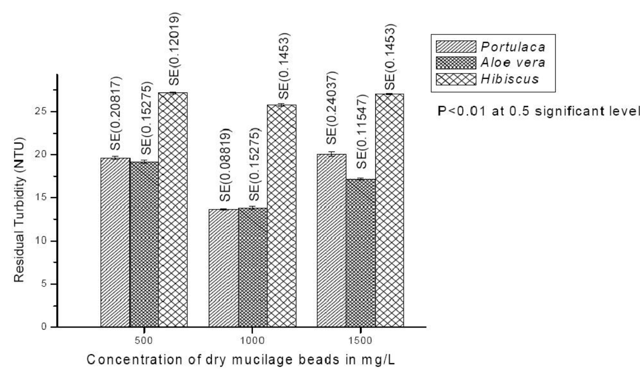

Figure 4 : Water clarification by mucilage-alginate beads in high turbid (450 NTU) water in 1 h.

During the coagulation, there could be an unavoidable interaction between the particles of different sizes, and form larger particles which fall faster than smaller ones and also tend to collide further and combine with other smaller particles till the coagulation process ends. Mucilage particles accumulate this process by the bridging mechanism. The mechanism of coagulation and flocculation could be explained by charge neutralization and bridging mechanism. Mucilage is a mixture of polysaccharides with high molecular weights. The increase in molecular weight of flocculant favours the bridging relative to charge neutralization mechanism. Bridging is may be due to mucilage hydrophilic character, several hydrogen bonds are formed between polyelectrolyte and water molecules[31]. This association tends to occupy larger surface area causing its very high viscosity, which in turn is the characteristic feature to form agglomerates.

Optimum dosage of coagulants

Wet beads of 2% and 3% solutions were used for trial batch but the turbidity removal was very low, due to their fragile nature, they broke down during the coagulation process and formed high sludge which could not be settled by coagulation process and further contributed to an increase in turbidity level. Experiments with 4% mucilage solution yielded satisfactory results.

The optimum dose of coagulants in the form of mucilage powder and mucilage beads required were found to be 12mg/l and 300 mg/l respectively for all the species studied to obtain maximum turbidity removal for low turbid water (35 NTU). Residual turbidity levels for water treated with *Hibiscus* were in the range of 7-8 NTU and the percentage removal was 77-80%. Whereas, in case of *Portulaca* and *Aloe* the residual turbid level after treatment were in the range of 4.5-5.5 NTU and the percentage removal was 84-87% respectively. Blank beads reduced the turbidity level to 9.5 NTU and the percentage removal of turbidity was 72.8%. These values were far below that of *Hibiscus* which showed minimum efficiency. Percentage removal of turbidity in high turbid waters (450 NTU) was 95% in the case of *Hibiscus*, 97% in the case of *Aloe* and *Portulaca* and the optimum dosage has been found to be 40mg/l (mucilage powder), 1000mg/l (in the form of beads). Filtration of so clarified water through Whatmann No.1 filter paper produced a very clear filtrate of less than one unit turbidity, which is in the range of WHO standard limits of drinking water. However, the treatment with blank beads showed 94.6% turbidity removal and the residual turbidity was found to be 24 NTU and the efficiency/added advantage of the mucilage of *Hibiscus* was dismal and in case of *Aloe* and *Portulaca* it was high.Mucilage–alginate beads used in the coagulation tests were based on equivalent concentration range of mucilage, that was entrapped into calcium alginate beads and results obtained were almost similar to that of un-entrapped mucilage. In general, any immobilized substance never shows 100% activity when compare to its actual form, but here using sodium alginate for entrapment was an added advantage because of its flocculating property. This is evident from the experiments conducted with blank beads. It is one of the reasons to show 100% efficiency. Similar observations were made by Singh et al[32]. When alginate-mucilage mixture is coordinated to sodium, it is a very flexible chain. When sodium is replaced by calcium however, each calcium ion coordinates to OH group of one alginate chain and one polysaccharide chain of mucilage and linking them together. The flexible chains become less flexible and form a huge network – a gel. All this happens within seconds after the alginate-mucilage mixture is dripped into the water bath with the calcium ions.

The optimal dose of each coagulant varies according to the initial turbidity of water. It was observed that by increasing the concentration of coagulant, turbidity removal also increased initially, but after attaining the optimum dosage level it shows no remarkable decrease in turbidity. Several studies reported that up to particular dosage flocculation increases and beyond that flocculation diminishes[33,34]. This could be explained by the fact that, the higher dose of flocculent would create the steric hindrance among the agglomerates to disturb the settling of particles.

Repeated use and capability

In order to check out the effectiveness in repetitive cycles the recovered mucilage–alginate beads were washed with distilled water and used for three repeated

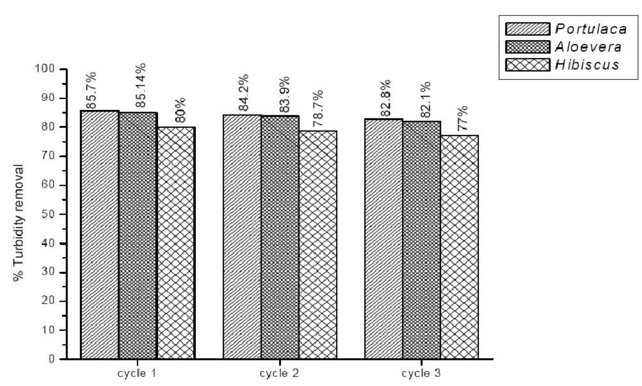

Figure 5 : Percentage turbidity removal in low turbid (35 NTU) water by recycled beads in 3 repeated cycles at their optimum dosage (12mg/l).

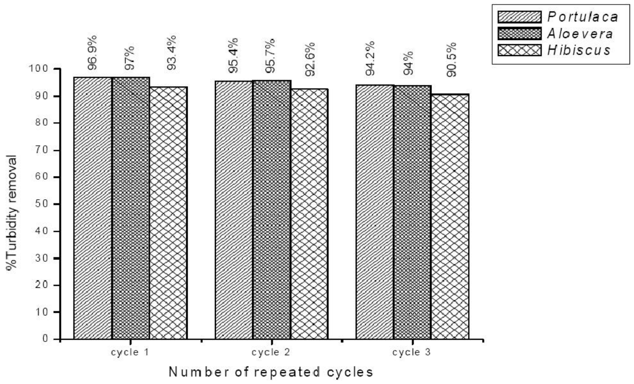

Figure 6 : Percentage turbidity removal in high turbid (450 NTU) water by recycled beads in 3 repeated cycles at their optimum dosage (20mg/l).

cycles. The measured activities were same for first two cycles. But the effectiveness decreased slightly in the 3rd cycle (figure 5 and 6). This may be due to that, some particles remain attached on to the beads which cause an accumulation of more suspended matter on to the beads. So that there will be no free space to bridge with colloidal particles that are suspended in water.

CONCLUSIONS

This study demonstrated the potential of encapsulated mucilage-alginate beads to act as water clarification agent's alternative to the most widely used synthetic coagulants. Overall study concludes that mucilage-alginate beads have essentially improved stability and compatibility of mucilage.

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