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Natural Polymers as Alternatives to Conventional Coagulation Agents – Lab-Scale Research with Moringa Oleifera Seeds and Chitosan

T Kucera[1], L Hofmanova[1] and R Biela[1]

[1]Brno University of Technology, Faculty of Civil Engineering, Institute of Municipal Water Management, Žižkova 17, Brno, 602 00, Czech Republic.

Abstract. This article presents the results of a laboratory-scale research focused on the use of alternative coagulants in water treatment. Attention was particularly paid to natural cation coagulants derived from Moringa oleifera seeds and chitin. These two coagulant agents were used in laboratory research for treating real raw water. Several series of laboratory jar tests were performed to identify the effectiveness of the selected coagulants in removing turbidity from water. The results of the experiments show that the efficiency of the process using natural polymers is not as high as that of conventional chemical reagents. A turbidity reduction efficiency of 80-82% was achieved, which is not as impressive as aluminum sulphate. However, it is a certain alternative in cases of secondary product formation (residual Fe and Al) and associated problems. Here it is possible to use a combination of natural and conventional reagents.

1. Introduction

The introduction of a coagulation process is often required for treating water from surface sources to decrease turbidity and color, as well as the volume of pathogenic organisms. The process may also be optimized for the removal of natural organic matter (NOM) and heavy metals. Various coagulant agents are used for the process and the selection of a suitable coagulant depends upon the type of substance removed, as well as the overall water treatment process [1]. Coagulation may be combined with other physical, chemical or biological processes.

Coagulants are primarily divided into the organic and inorganic. Inorganic coagulants are metal salts, or possibly in a hydrolyzed polymer form. Organic coagulants are available in many types of substances and create long chains. Coagulants may further be divided based on their charge into cation and anion and, according to their origin, divided into synthetic or natural.

Salts of aluminum and iron appear to be most commercially successful. However, in recent years, polymer coagulants are also used, most often as auxiliary agents used to increase the efficiency of water treatment on the one hand and decrease costs on the other.

2. Natural polymers

Over time, there has been an effort to replace classic coagulants with natural non-toxic and environmentally-friendly substances. The reason may be the high cost or limited availability of commonly used coagulants (aluminum sulfate, iron(III) sulfate, synthetic polymers etc.), the requirement of highly effective doses of these agents, production of an notable volume of sludge [2] and the undesirable toxicity of the residual coagulants that remain in the treated water.

The use of seeds of certain plants (for example *Lens esculenta*, *Tamarindus indica*, *Cyamoppsis psoraloides)* presents a cost-effective, practical and suitable solution for water treatment in developing countries and beyond. A certain preparation is necessary in order to use these seeds, including drying, grinding and dilution with water.

2.1. Moringa oleifera tree seeds

Moringa oleifera is an oil-producing tree originally from Northern India (Himalayas), currently spread across tropical regions worldwide. The plant is notable for its high resistance to drought. *Moringa* seeds are composed of approximately 31% protein, 18% saccharides and 37% fats. Powder from ground seeds contains soluble proteins capable of flocculating turbidity contained in water. The solubility of these proteins is increased with the content of salts in water. Sodium chloride (NaCl) is thus sometimes used during laboratory preparations of the coagulation agent. The coagulation effect of the extract prepared with the use of salt shows greater effectiveness than agents prepared using only tap or distilled water. Studies prove that, in the case of *Moringa oleifera,* adsorption, charge neutralization and formation of polymer bridges between particles are the predominant mechanisms for removing turbidity (Lea 2014; Yarahmadi 2009).

The processes of preparing a coagulation agent from *Moringa oleifera* seeds may differ. Nonetheless, the polyelectrolyte used for water treatment is in most cases obtained by grinding dried seeds from the tree and mixing them with water. The proteins with predominantly positive charge on their surface are thus transferred into the water solution. A typical dose for preparation ranges from 10 to 50 grams of ground seeds per liter of water. The suspension of seeds with water is shaken intensively to allow the development of molecules, followed by filtration. The dosage of coagulation agent ranges from 75-200 $mg \cdot l^{-1}$ depending on the properties (particularly turbidity) of treated water (Ndabigengesere and Narasiah, 1998).

2.2. Chitosan

Chitosan is a substance insoluble in most organic solvents. However, it dissolves in both organic and inorganic acids (such as acetic acid, formic acid, etc.). This polysaccharide is produced through an alkaline deacetylation of chitin, which is found in the outer shells of crustaceans, insects and gastropods, as well as within the cellular walls of yeast and certain fungi. Chitin and chitosan may be produced from the waste products of processing marine animals (Younes and Rinaudo 2015). Chitosan produced for the purposes of water treatment is typically prepared using acetic acid where it is a subject of hydrolysis. Chitosan is a cation polyelectrolyte used for the reduction of water turbidity (Pontius 2016), sorption of metal irons, as well as removal of organic pollution. Destabilization primarily takes place through the mechanism of charge neutralization. Chitosan is a biodegradable substance and its use in water treatment is therefore more environmentally friendly than commonly used coagulants. The frequent use of chitosan is increasing in recent years. This substance is used for example in Norway, where it was selected as a coagulant in several water treatment facilities, used either exclusively or in a combination with metal-based coagulants (Pontius 2016).

Chitosan has many advantages in comparison with traditional coagulants (flocculants). Among them are effectiveness in low doses, success in the reduction of chemical oxygen demand (COD), the ability to remove metal ions, the formation of large (voluminous) floccules with higher sedimentation rates, biological degradability, major effect in the removal of algae and suspended particles, anti-bacterial effects and zero formation of secondary pollution (Bhalkaran 2016). Chitosan is capable of binding both fats and metals, including arsenic, molybdenum, cadmium, chromium, lead and cobalt (Li et al. 2012).

Many studies have been conducted regarding the use of chitosan as a single coagulant or in combination with other agents in removing turbidity (Bina et al. 2009; Pontius 2016).

Chitosan, as a positively charged polymer with long chains, is able to coagulate negatively charged and finely dispersed colloidal particles through adsorption, charge neutralization, hydrophobic effect or through the formation of polymer bridges. Considerable savings in terms of the volume of agents used may be achieved by using such natural coagulants in combination with the more classic forms, resulting in additional cost reductions for sludge removal, as it is produced in smaller quantities (Bhalkaran 2016).

3. Results

Our research focused on the comparison of effectiveness in reducing turbidity after applying three coagulants used in water treatment. The selected coagulants were aluminum sulfate commonly used as a conventional coagulant, as well as chitosan and *Moringa oleifera* as alternatives. An optimization jar test was performed with the tested aspect being turbidity. The research was carried out in the laboratory format as a series of jar tests.

3.1. Moringa oleifera

The coagulant obtained from Moringa oleifera seeds by the method described above was used to treat water of identical quality as above and also within a laboratory-scale test. The same parameters were set for the coagulation test. The formation of small light-colored floccules in the treated water was recorded as early as the slow blending stage.

The amount of these small observed clusters grew with increased amounts of the coagulation agent. However, the floccules remained suspended in the volume of the vessels throughout the entire phase and were only partly sedimented. Only a portion of the floccules settled on the bottom, along with sediment. Despite this, lower levels of turbidity were measured than prior to water treatment. Results are shown in Figure 1.

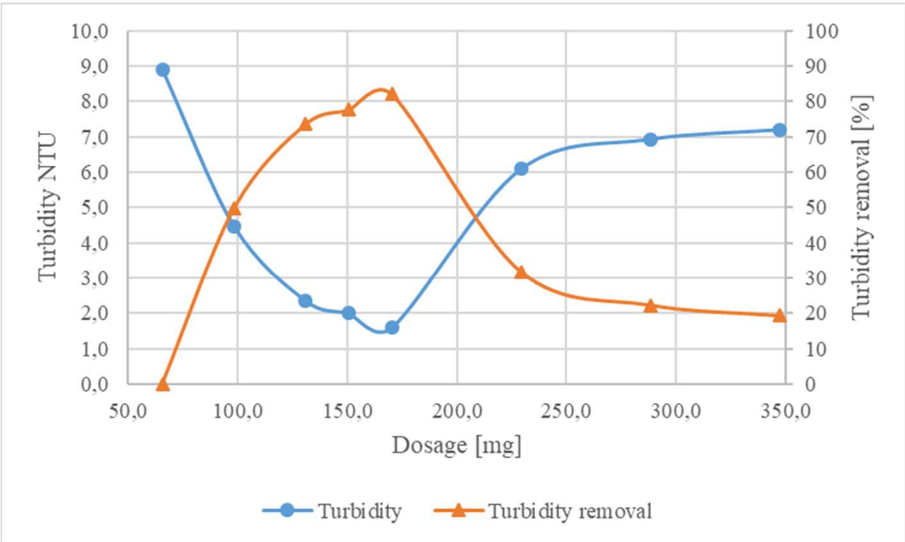

Figure 1. Turbidity removal – Moringa oleifera.

As is apparent from the above results, the most effective dose of Moringa oleifera for turbidity removal was 164.6 mg. The initial turbidity value was reduced to 1.60 NTU, representing the removal of 82.04% of turbidity. The lowest value of turbidity removal was measured at the dose of 20.6 mg of Moringa oleifera. Here, the turbidity value only decreased by 0.01 NTU.

When using Moringa oleifera, the floccules formed during coagulation were of pale color. They were visible to the naked eye only in the second phase of blending. The sedimentation of the aggregates took place very slowly. Even after one hour of sedimentation, there were no apparent changes. The optimum dose was 164.6 $mg \cdot l^{-1}$, which is the largest in comparison with both aluminum sulfate and chitosan. Better results of turbidity removal could apparently be achieved by prolonging the sedimentation period or using filtration.

3.2. Chitosan

The next coagulation tests were using chitosan as an agent – also under the same conditions. The formation of floccules in raw water was visible from approximately the middle of the slow blending phase. The small aggregates had a light brown color and settled more quickly than the aggregates using Moringa oleifera. Although most floccules settled after 60 minutes of sedimentation, a portion of the floccules remained suspended. As regards turbidity removal, the resulting values are shown in Figure 2.

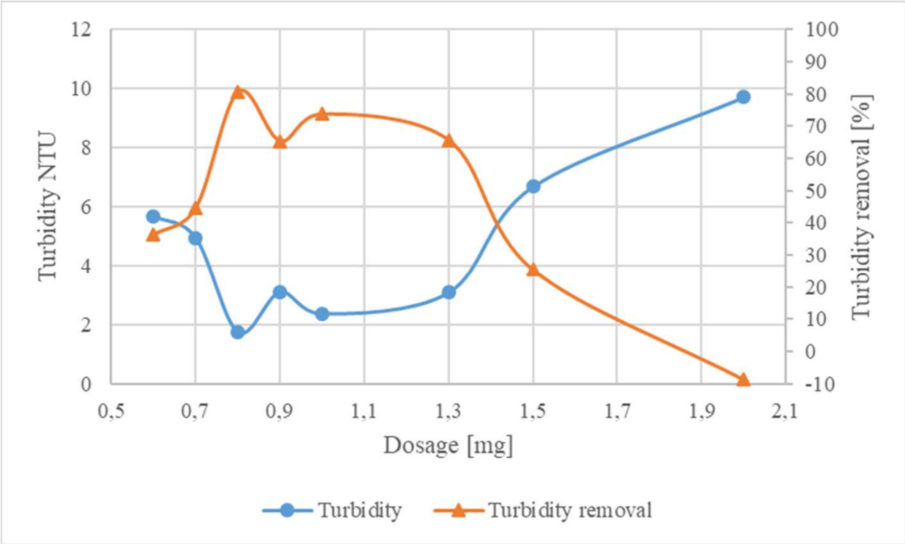

Figure 2. Turbidity removal – chitosan.

In the case of chitosan, the optimal dose was low – the mentioned 0.8 mg·l^{-1}. At this dose, the remaining turbidity measured had a value of 1.73 ZF, representing 80.66% less than measured in raw water. In the jar test, the effect of chitosan was only visible during the second half of slow blending. Very tiny light brown floccules were formed in the vessels and their amount was in direct proportion to the coagulant dose. The aggregates showed much better sedimentation properties than in the case of Moringa oleifera.

4. Discussion and Conclusions

The laboratory experiments described evaluated the removal of turbidity from a water environment using a coagulation process with three different coagulation agents. As is apparent from Figures 1-2, the course of coagulation and flocculation during the optimization jar test, as well as the dependency of the agent dosage on turbidity removal, were different for each agent.

The most effective coagulant in terms of turbidity removal under the given conditions is without doubt aluminum sulfate (from previous research). But in case of chitosan, the much lower optimum dosage compared to the other two agents is noteworthy (Table 1).

Table 1. Comparison of the results upon using three different coagulants.

Coagulant	Optimal Dosage	Turbidity	Turbidity removal
		@ 60 min	
	[mg]	[NTU]	[%]
Aluminum sulfate	150.7	0.46	94.84
Moringa oleifera	164.6	1.60	82.04
Chitosan	0.8	1.73	80.66

The best results in removing turbidity from raw water were achieved by aluminum sulfate. However, the use of aluminum sulfate in water treatments from secondary products had related health risks. The results of the experiment using Moringa oleifera are less impressive, although removing turbidity by 82% indicates a very good overall effectiveness of this agent. Better results could be obtained through different preparation of the coagulant (the use of NaCl, distilled water). In the case of chitosan, a slightly better result was expected. However, the reduction of turbidity by 80.66% indicates very good properties of this polyelectrolyte in removing turbidity from raw water. Chitosan efficiency may be supported by using it in combination with traditional coagulants.

Studies researching this version prove that the application of certain amounts of substances with natural origins reduces the necessary optimal amounts of commonly used coagulants. Water treatment using a combination of both substances is more environmentally friendly. Lesser amounts of harmful residual products are formed during the course of such process (residual Fe and Al) and the amounts of necessary agent are reduced, resulting in savings as the volume of the formed sludge is reduced.

The authors of this study are aware that the results in case of Moringa oleifera seeds and chitosan might even be better, as the jar test conditions were optimized for aluminum sulfate. In the case of a possible replacement of an existing coagulant (aluminum sulfate) by an alternative in a real-application situation, it would be necessary to also adjust the parameters of the coagulation process, such as blending rates and the period of placement in blending and separation basins.

An interesting criterion for discussions of possible replacements of conventional coagulants for natural polymers may also be their cost. The prices of all three coagulants are subject to fluctuations depending on the location of use. Our research did not establish the price of the optimum dosage, as the natural polymers used for the purpose of our study were obtained under price-inefficient conditions at low quantities.

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