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INVESTIGATION OF COAGULATION ACTIVITY OF NATURAL COAGULANTS FROM SEEDS OF DIFFERENT LEGUMINOSE SPECIES

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The ability of seeds of plants: Phaseolus vulgaris, Robinia pseudoacacia, Ceratonia siliqua and Amorpha fruticosa, to act as natural coagulants was tested using synthetic turbid water. This water was prepared by adding kaolin into tap water, just before the test. Active components were extracted from ground seeds with distilled water. The coagulation ability of this extract was assessed by the use of standard jar test measurements in water with various initial turbidity. Investigation of these natural coagulants was confirmed their positive coagulation activity. Of all plants that have been examined, the seed extract from Ceratonia siliqua appeared to be one of the most effective coagulants for water treatment. A dose of 20 mg/l of this coagulant resulted in 100% coagulation activity for clarification of water with 17.5 NTU initial turbidity.

KEYWORDS: Water; turbidity; clarification; natural coagulants

INTRODUCTION

Many coagulants are widely used in conventional water treatment processes for tap water production. These coagulants can be classified into inorganic coagulants (e.g. aluminum sulfate, polyaluminum chloride, ferric chloride), synthetic organic polymers (e.g. polyacrylamide derivatives and polyethylene imine) or naturally occurring coagulants (e.g. chitosan, plant extracts). These coagulants are used for various purposes, depending on their chemical characteristics.

Aluminium salts are the most widely used coagulants in water and wastewater treatment all over the world. However, the studies by several workers have raised doubts about introducing aluminum into environment (1, 2, 3). Ferric salts and synthetic polymers have

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been used as an alternative but with a limited success due to the fact that their influence on living organisms is not fully understood. However, some studies have reported that aluminum that remains in the water after coagulation, may induce Alzheimer's disease (4, 5). It was also reported that monomers of some synthetic organic polymers such as acrylamide have neurotoxicity and strong carcinogenic properties (1). Besides, many developing countries can hardly afford the costs of imported chemicals for water and wastewater treatment.

On the other hand, naturally occurring coagulants are biodegradable and are presumed safe for human health. Some studies on natural coagulants have been carried out and various natural coagulants were produced or extracted from microorganisms, animals, or plants.

The use of natural materials of plant origin to clarify turbid raw waters is not a new idea. Natural coagulants have been used for domestic household for centuries in traditional water treatment in rural areas. In these regions people have used original plants like seeds of the Nirmali tree *Strychnos potatorum*, then roasted grains of *Zea mays* or sap from the 'tuna' cactus *Opuntia fiscus indica* (6). In time there has been more interest in the subject of natural coagulants, especially to reduce the problems of water and wastewater treatment in developing countries and to avoid some health risks. Plants that were recently tested are mesquite bean (*Prosopis juliflora*) and *Cactus latifaria* in Venezuela (7), in Egypt and North Sudan various bean (*Phaseolus*), peas (*Pisum*), peanuts (*Arachis*) and lupines (*Lupines*) (8), *Cassia angustifolia* seeds (9), seeds from *Moringa oleifera* (8, 10, 11, 12), etc.

The aim of this study was to investigate the applicability of natural coagulants extracted from seeds of some leguminoses for water clarification. These natural coagulants should be substitute for commercial coagulation aids and thus avoid their negative efects. Plants like bean – common bean *Phaseolus vulgaris,* black locust *Robinia pseudoacacia,* carob *Ceratonia siliqua* and indigobush *Amorpha fruticosa* have been investigated. These plants are indigenous in Serbia and Montenegro and they are common in that area.

EXPERIMENTAL

Preparation of seed extracts

The seeds of *Phaseolus vulgaris, Robinia pseudoacacia, Ceratonia siliqua* and *Amorpha fruticosa* were obtained in pods and only seeds from dry pods were used. The seeds were removed from the pods and were ground to fine powder using a laboratory mill just before extraction. Ten grams of seed powder was suspended in 1000 ml of distilled water and the suspension was stirred using a magnetic stirrer for 10 min to extract the coagulation active component. The suspension was then filtered through a rugged filter paper. The filtered solutions were kept in refrigerator.

The seed extracts were added to turbid water as coagulant agents. The extracts were added in a quantity of 0.5, 1, 2 and 3 mililitres to 1 l of turbid water. These volumes of extract in fact contain active coagulation agents extracted (originated) from 5, 10, 20 and 30 mg of seeds and these quantities are named coagulant doses.

Preparation of synthetic turbid water

Synthetic turbid water for coagulation tests was prepared by adding kaolin into tap water. Ten grams of kaolin was added to one liter 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 h for complete hydration of the particles. This suspension was used as the stock solution. Immediately before the coagulation experiments, suspensions with initial turbidity of 17.5, 35 and 70 NTU were prepared by adding 2.5, 5 or 10 ml of stock suspension to 1000 ml of tap water.

Coagulation test

Jar test was used for evaluation of coagulation processes. Coagulation activity of each seed extract was verified by the jar test. The synthetic turbid water of different initial turbidiy was filled in four 500 ml bakers. Then, the suspensions were mixed at 200 rpm. Different doses of each coagulant were added into beakers and mixed for 2 minutes. The mixing speed was then reduced to 80 rpm and was kept for further 30 minutes. Then the suspensions were left for sedimentation. After sedimentation for 1 hour, an aliquot of 100 ml of clarified sample was collected and residual turbidity was measured – RT_S. The same coagulation test was conducted with no coagulant as a control. The residual turbidity in the control was defined as RT_B. Coagulation activity was calculated as:

$$\text{Coagulation activity} = (RT_B - RT_S)/RT_B$$

Analythical methods

Turbidity was measured using a nephelometer (VOS 4000) and it was expressed in nephelometric turbidity units (NTU) (13).

Organic matter concentration in water after coagulation was determined as $KMnO_4$ demand by Kübel-Tiemann (13).

pH was determined using a pH meter (HI 9321).

RESULTS AND DISCUSSION

Effect of pH on coagulation activity

Solubility of organic mater in aqueous medium depends on pH. The particles usually have some negative charge and they are stable at the isoelectric point. At that pH, the solubility of organic matter is the lowest. Because of that, we have to investigate the influence of pH on coagulation activity and to determine optimum pH value for further coagulation tests. The jar test experiments with the extract of common bean as coagulant, using synthetic water with turbidity of 35 NTU were run. The initial pH in the synthetic water was varied from pH 4 to pH 11. The results obtained from jar test experiments with 30 mg/l of coagulant are shown in Fig. 1.

As can be seen from Fig. 1, the extract of common bean showed high coagulation activity at pH 10 or more, and no coagulation activity at pH about 7. At pH about 7 the

low efficiency also was observed when purified extract of *Moringa oliefera* (14), and Al^{3+} or Al^{3+} + tannin (15) were used as coagulants. In this work, pH 10 was chosen for further experiments.

Coagulation activities of all prepared seed extracts were evaluated using synthetic water with initial turbidity of 17.5, 35 and 70 NTU, and these results are presented in Fig. 2, Fig. 3 and Fig. 4, respectively.

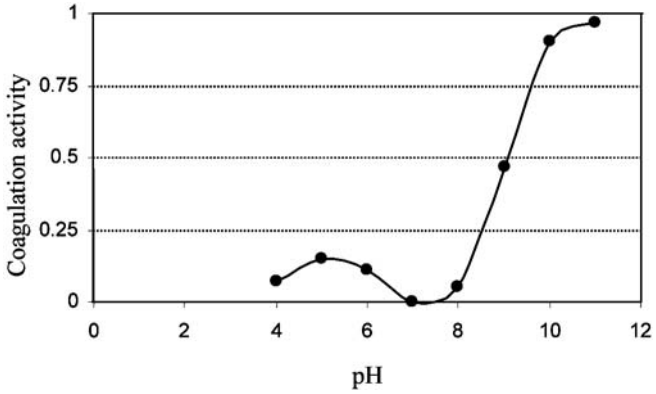

Fig.1. Effect of pH on coagulation activity of common bean extract (coagulant dosage 30 mg/l, initial turbidity 35 NTU)

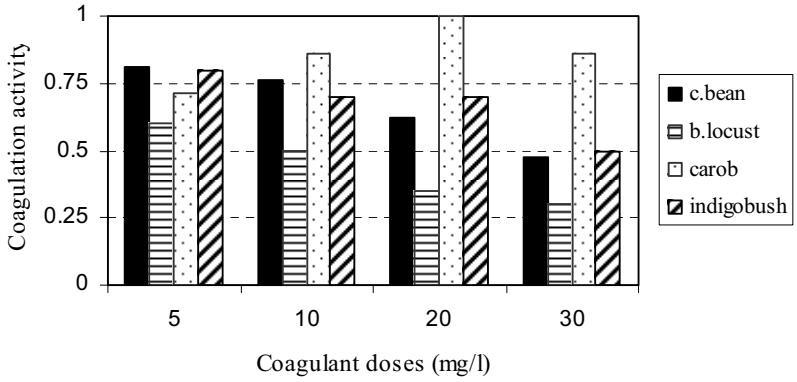

Fig. 2. Effects of coagulant doses on coagulant activities of different natural coagulants in water with 17.5 NTU initial turbidity

In water with the initial turbidity of 17.5 NTU, the coagulation activities mainly considerable decreased when the coagulant doses increased, except for the carob extract. Coagulation activities increased when the doses of carob extract were increased. The coagulation was the most effective at a dose of 20 mg/l, when the coagulation activity of carob extract was 100% (Fig. 2). In water with 17.5 NTU the extracts of common bean and indigobush at 5 mg/l dose were the most effective.

Extracts of common bean and indigobush showed similar coagulation activity at all doses applied, and the extract of black locust had the least coagulation activity, either in water with 17.5 or 35 NTU. In the water with these turbidities the extract of black locust seeds was the worst.

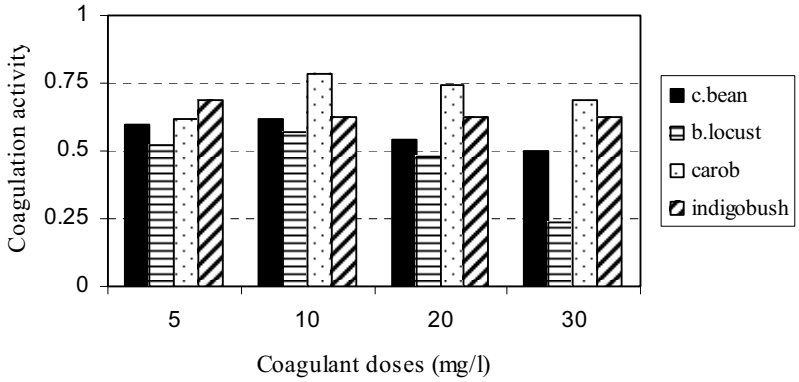

Fig. 3. Effects of coagulant doses on coagulant activities of different natural coagulants in water with 35 NTU initial turbidity

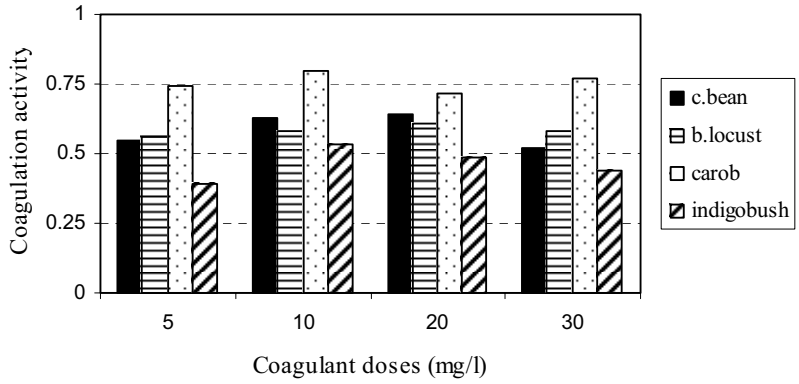

Fig. 4. Effects of coagulant doses on coagulant activities of different natural coagulants in water with 70 NTU initial turbidity

In the water with turbidity of 70 NTU the extract of carob was the best and the extract of indigobush the worst coagulant. Extract of black locust showed coagulation activity which was similar to that of common bean extract. In the water with higher turbidity the coagulation activities were almost independent of the coagulation dose. The coagulation effects of extracts of *Prosopis juliflora* and *Cactus latifaria* (7) and extract of *Moringa oleifera* (12) are also independent of their doses in water with the initial turbidity of 100 NTU. Coagulation process is usually dependent on a multitude of factors: initial turbidity, pH-value, composition of water, temperature, intensity and duration of stirring du-

ring mixing and nature and dose of the coagulant. Besides, the coagulation process depends on the extraction mode (10). Because of that, selection of the best coagulants and their sufficient dose for coagulation is rather difficult. On the basis of the presented results, the extract of carob seeds is proper for treatment of water with different initial turbidity, in a dose of 10 mg/l or higher. Also, the extracts of common bean and indigobush seeds are suitable for treatment of water with lower turbidity, in a dose of 5 mg/l.

CONCLUSION

The purpose of this study was to investigate the possibility of some natural coagulants - seed extracts of different species of leguminoses, to clarify turbid water. These investigations confirmed positive coagulation activities of the investigated natural coagulants. The results show that the extract of carob was the most effective coagulant in all doses, and in water with different initial turbidity. Dose of 20 mg/l of this coagulant has shown 100% coagulation activity for clarification of water with a turbidity of 17.5 NTU. The extracts of seeds of common bean and indigobush, in a dose of 5 mg/l, are the best for clarification of water with lower turbidity. The extract of seeds of black locust showed good coagulation activity in water with the turbidity of 70 NTU in a dose of 30 mg/l and for lower turbidity - lower doses are better. Thus, the seed extracts of some leguminoses can be used as natural coagulants with previous determination of their optimum dose for successful clarification.

REFERENCES

1. Mallevialle, J., A. Brichet and F. Fiessinger: How safe are organic polymers in water treatment. J. Am. Wat. Wks. Assoc. **76** (1984) 87-93.
2. Miller, R. G., F. C. Kopfler. K. C. Kelty, J. A. Stober and N. S. Ulmer: The occurrence of aluminium in drinking water. J. Am. Wat. Wks. Assoc. **76**, 1 (1984) 94 – 101.
3. Letterman, R.D. and C.T. Driscoll: Survey of residual aluminium in filtered water. J. Am. Wat. Wks. Assoc. **80** (1988) 154-158.
4. Martyn, C. N., D. J. P. Barker, C. Osmond, E. C. Harris, J. A. Edwardson and R. F. Lacey: Geographical relation between Alzheimer's disease and aluminium in drinking water. The Lancet **1** (1989) 59 – 62.
5. Letterman, R.D. and R.W. Pero: Contaminants in polyelectrolytes used in water treatment. J. Am. Wat. Wks. Assoc. **82** (1990) 87-97.
6. Sutherland, J.: *Moringa oleifera* pages (in Environmental engineering pages), http// www.le.ac.uk/engineering/staff/Sutherland/moringa/moringa.htm (2005).
7. Diaz, A., N. Rincon, A. Escorihuela, N. Fernandez, E. Chacin and C. F. Forster: A preliminary evaluation of turbidity removal by natural coagulants indigenous to Venezuela. Process Biochem. **35** (1999) 391 – 395.
8. Gassenschmidt, U., D. K. Jany, B. Tauscher and H. Niebergall: Isolation and characterization of a flocculating protein from *Moringa oleifera* Lam. Biochem. Biophys. Acta **1243** (1995) 477 – 481.

9. Sanghi, R., B. Bhatttacharya and V. Singh: *Cassia angustifolia* seed gum as an effective natural coagulant for decolourisation of dye solutions**.** Green Chem. **4**, 3 (2002) 252 – 254.
10. Okuda, T., A. U. Baes, W. Nishijima and M. Okada: Improvement of extraction method of coagulation active components from *Moringa oleifera* seed. Wat. Res. **33**, 15 (1999) 3373 – 3378.
11. Muyibi, S. A. and M. L. Evison: Optimizing physical parameters affecting coagulation of turbid water with *Moringa oleifera* seeds. Wat. Res. **29**, 12 (1995) 2689 – 2695.
12. Ndabigengesere, A., K. S. Narasiah and G. B. Talbot: Active agents and mechanism of coagulation of turbid waters using *Moringa oleifera*. Wat. Res. **29**, 2 (1995) 703 – 710.
13. Feliks, R. and S. Škunca-Milovanović (Ed.): Voda za piće – Standardne metode za ispitivanje higijenske ispravnosti, Savezni zavod za zdravstvenu zaštitu i NIP Privredni pregled, Beograd (1990) pp. 118-119, 134-136.
14. Okuda, T., A. U. Baes, W. Nishijima and M. Okada: Isolation and characterization of coagulant extracted from *Moringa oleifera* seed by salt solution. Wat. Res. **35**, 2 (2001) 405 – 410.
15. Özacar, M. and A. Şengil: Evaluation of tannin biopolymer as a coagulant aid for coagulation of colloidal particles. Colloids and Surfaces A: Physicochem. Eng. Aspects **229** (2003) 85 – 96.

ИСПИТИВАЊЕ КОАГУЛАЦИОНЕ АКТИВНОСТИ ПРИРОДНИХ КОАГУЛАНАТА ИЗ СЕМЕНА НЕКИХ ВРСТА ЛЕГУМИНОЗА

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Могућност да се екстракти семена пасуља (*Phaseolus vulgaris),* багрема *(Robinia pseudoacacia),* рогача (*Ceratonia siliqua)* и багремца *(Amorpha fruticosa)* примене као природни коагуланти испитивана је на синтетички мутној води. Та вода је припремана додатком каолина у водоводску воду, непосредно пре извођења огледа. Активне компоненте су екстраховане дестилованом водом из самлевеног семена. Ефикасност коагулације овако добијеним екстрактима семена је испитивана стандардним џар тестом у води различите почетне мутноће. Испитивање ових природних коагуланата је потврдило њихову позитивну коагулациону активност. Од свих испитиваних семена, екстракт семена *Ceratonia siliqua* се показао као најефикаснији коагулант за обраду воде. Доза овог коагуланта од 20 mg/l показала је коагулациону активност од 100% за бистрење воде почетне мутноће 17,5 NTU.

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