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Improving Filtrate Quality Using Agrobased Materials as Coagulant Aid

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In the present study, an evaluation of agrobased materials (ABM) as a coagulant aid in conjunction with alum has been conducted to determine their efficacy in water treatment. The agrobased materials evaluated are Surjana seed (*Moringa oleifera*), Nirmali seed (*Strychnos potatorum*) and maize (*Zeemays*). Experiments have been conducted simulating a conventional water treatment train consisting of coagulation-flocculation-settling and granular media filtration. Emphasis has been given to the filtration aspect of the treatment train using synthetic turbid water. The filter performance was defined by water quality and head loss development across the filter bed. When Nirmali seed or maize was used as a coagulant aid, the alum dose required was 25 and 15 mg/L, respectively, and the filtrate turbidity achieved was less than 0.2 NTU, whereas alum alone with a dose of 45 mg/L achieved filtrate turbidity levels higher than 1 NTU. Thus, the use of ABM improved the filtrate quality. Head loss in filter with Surjana seed and Maize as coagulant aids was comparable to that of alum alone, whereas it was higher when Nirmali seed was used as a coagulant aid.

Key words: **agrobased materials, coagulant aid, filtration, water quality**

Introduction

The removal of particulate matter in water treatment has gained greater importance due to a tightening of the turbidity standard, and the recognition that naturally suspended particles represent transport vehicles for undesirable organic and inorganic contaminants, taste-, odour-, and colour-causing compounds, and pathogenic microbial contaminants. Conventional water treatment processes involve coagulation, flocculation, settling and filtration. In water treatment, filtration is an essential step in the production of high-quality water, which is achieved by deposition of suspended solids within or on the filter media. 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 drinking water increases the aluminum concentration in finished water (Barnett et al. 1969; Miller et al. 1984; Pitchai et al. 1992; Selvapathy and Vijayaraghavan 1994). A higher finished water turbidity may result in

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a decrease in the effectiveness of disinfection due to enmeshment of microorganisms by alum floc, thus protecting them from the disinfectant (Hoff 1977). The high concentration of aluminum is also of concern because of potential adverse health effects. Aluminum intake into the body has been linked with several possible neuropathological diseases including presenile dementia and Alzheimer's disease (Crapper et al. 1973; Alfrey et al. 1976; Martyn et al. 1989). An approach to reduce the concentration of residual aluminum in finished water, is to use a synthetic polymer or a naturally occurring polyelectrolyte as a coagulant aid with primary coagulant alum.

A polyelectrolyte in conjunction with a metal coagulant improves coagulation by accelerating the process of coagulation. The coagulant aid reduces the requirement of alum and improves the physical characteristic of flocs, which results in better quality of treated water. Although synthetic polymers have significant potential for water treatment, their use has been restricted due to high cost and uncertainties regarding chemical impurities associated with polymer synthesis. Toxicity of any polyelectrolyte has to be checked before it can be used as coagulant or coagulant aid (CPHEEO 1991). Because most of the naturally occurring polyelectrolytes are of plant origin and are generally nontoxic in nature, these are given more importance by several workers (Bulusu et al. 1968; Rao and Sastry 1974; Tripathy et al. 1976; Bhole 1990, 1993, 1995). Studies on such material have been restricted to only finding the optimum dose required for clarification after settling and comparing the results to that of alum. The objective of coagulation is, however, to form flocs which are not only settlable but should be non-filterable by the sand bed. Therefore, it was necessary that an evaluation of agrobased materials (ABM) for the whole treatment train of coagulation-flocculation-settling and filtration should be conducted to evaluate their efficacy.

Performance of agrobased materials, Surjana seed (*M. oleifera*), Nirmali seed (*S. potatorum*) and maize (*Zeemays*) as coagulant aids with alum as a coagulant was tested through the process train of coagulation-flocculation-settling and filtration. Filtrate turbidity and head loss development as filtration proceeds were considered as the basis for the evaluation.

Materials and Methods

Surjana seed, Nirmali seed, maize and commercial-grade alum used in this study were procured from the local market. The characteristics of the alum used are shown in Table 1.

Processing of Agrobased Materials

The maize and Surjana seeds were dried in an oven at 40°C for 24 h. Dried maize and Surjana seeds were powdered in a grinder and sieved through a nylon cloth (size of opening: 212 μm) to obtain very fine pow-

Table 1. Specification of alum used

Characteristic	Value
Insoluble matter, % mass, max.	0.5
Soluble iron (as Fe), % mass, max.	0.7
Water soluble aluminum compounds (as Al_2O_3), % mass, min.	15.0
pH (5% aqueous solution)	2.7
Lead (as Pb), ppm, max.	30
Arsenic (as As), ppm, max.	6.0

der. One gram of powder was soaked in distilled water, blended and the volume was made up to 100 mL. As a preservative, 0.5 mL HCl was added per 100 mL of solution (Bhole 1993; Tripathi et al. 1976). Nirmali seeds, due to their hard structure, could not be powdered in a grinder. The seeds were kept immersed in 50 mL water containing 2 mL conc. HCl. After a week, the mixture was mashed to a soup-like solution, which was washed through a nylon cloth and the material retained on the cloth was oven dried for 24 h at 103 to 105°C and weighed. By calculating the actual weight of the seeds dissolved, the strength of the stock solution was determined.

Preparation of Turbid Water

Natural clay was used to prepare turbidity by soaking the clay overnight 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 preparation of turbidity. A portion of the stock suspension was diluted with tap water and after 30 min of settling in a tray of 10-cm depth. The supernatant was carefully decanted. A desired turbidity of 100 NTU was obtained by diluting it. The suspension obtained was subjected to sedimentation analysis and the particle size distribution (PSD) showed that 71% of particles were finer than 8.6 µm, 28% of particles were finer than 1.4 µm, 21% of particles were finer than 1.25 µm, 10.2% of particles were finer than 0.72 µm, and 10% of particles were finer than 0.62 µm. Chemical characteristics of the tap water are shown in Table 2. The chemical analyses were conducted as per Standard Methods (APHA 1985). Turbidity was measured by Systronic Nepheloturbidity meter type 131, Systronics, India. The procedure conformed to that described in Standard Methods for the Examination of Water and Wastewater (APHA 1985).

Coagulation and Flocculation

A jar test protocol was developed to simulate coagulation-flocculation and settling at water treatment plants. The jar test apparatus could

Table 2. Quality parameters of the tap water

Parameter	Tap water
pH	7.8–8.1
Turbidity	0.3 NTU
Alkalinity (as $CaCO_3$)	370 mg/L
Calcium hardness (as $CaCO_3$)	18 mg/L
Magnesium hardness (as $CaCO_3$)	28 mg/L
Potassium (K^+)	3 mg/L
Sodium (Na^+)	25 mg/L
Chlorides (Cl^-)	108 mg/L

accommodate six 1-litre jars at a time. The sequence of addition was coagulant aid followed by alum. Flash mixing was done for 2 min at 100 RPM followed by slow mixing for 8 min at 25 RPM. The jar test mixer was then turned off and the flocs were allowed to settle for 30 min. Settled water samples were collected at 5 cm below the water surface to determine settled water turbidity. The jar test experiments were conducted with an objective to select the combination of coagulant aid and alum that would achieve settled water turbidity of less than 10 NTU. The experiments were conducted with a fixed dose of agrobased material (coagulant aid) and varied alum dose. The initial synthetic water turbidity was 100 NTU and the water temperature varied between 25 and 27.5°C. The experimental conditions employed are summarized in Table 3.

Laboratory Filtration Studies

A schematic diagram of the filtration setup used is presented in Fig. 1. A plastic transparent column having an internal diameter of 25 mm was used to prepare the filtration assembly. The depth of sand was kept at 200 mm supported by 100 mm gravel. The sand conformed to the specifica-

Table 3. Experimental conditions employed for coagulation-flocculation and settling

Dose range of alum	Coagulant aid and its dose range	pH after settling
5–45 mg/L	None	7.2–8.2
10–25 mg/L	Surjana seed: 10–30 mg/L	7.5–8.2
10–25 mg/L	Nirmali seed: 3–5 mg/L	7.6–7.9
10–25 mg/L	Maize: 10–30 mg/L	7.2–7.6

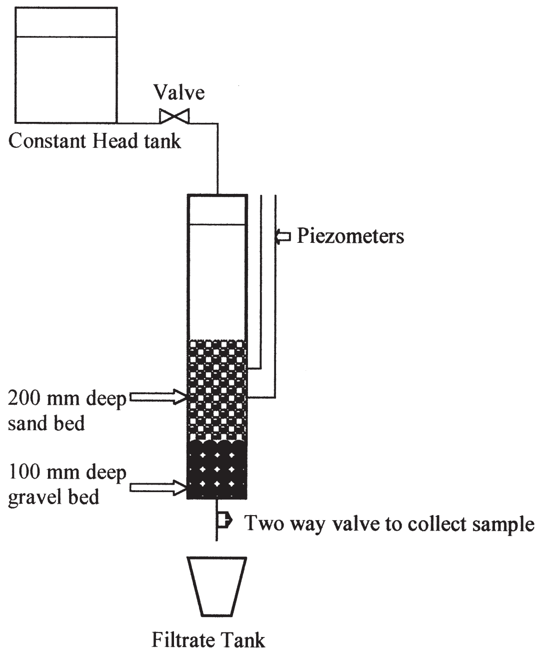

Fig. 1. Schematic of bench-scale filtration setup.

tions of sand for rapid sand filter in water treatment plants and had the following characteristics: effective size (d_{10}): 0.45 mm; d_{60} size: 0.61 mm; uniformity coefficient (U.C.): 1.355; specific gravity: 2.65; and porosity of bed (f): 0.447. A six-mm diameter flexible transparent tube was used as piezometric tube and inserted at depths of 50 mm and 100 mm from the top of the sand bed. The influent to the filter was the supernatant obtained from the jar test experiment. Flow was controlled with a valve and the flow rate was maintained at 5.0 m/h which is the standard rapid sand filtration rate (CPHEEO 1991). To ensure a proper compaction and cleanliness of the filter bed, clean bed head loss was determined by the Carman-Kozeny equation modified by Fair and Hatch (Fair et al. 1971) and the computed value (1.84 cm per 5 cm depth of sand) was verified with actual clean filter bed head loss before the commencement of the filter run. To monitor the head loss development, head losses at 5-cm and 10-cm filter bed depth were recorded at regular intervals and the filtered water quality was monitored. A summary of experimental conditions employed is presented in Table 4.

Results and Discussion

Coagulation-Flocculation

Table 5 presents the jar test results. The dose of alum required was 45 mg/L to achieve a settled water turbidity of less than 10 NTU, where-

Table 4. Experimental conditions employed for filtration study

Coagulant aid	Dose of alum (mg/L)	Dose of coagulant aid (mg/L)	Temperature (°C)	pH	Turbidity of settled water used in filtration (NTU)
Surjana seed	20	10	27.5	7.9	7.2
Maize	15	10	26.8	7.9	9.8
Nirmali	25	3	27.0	7.8	7.2
None	45	not applicable	27.5	8.0	8.0

as the alum requirement decreased when the agrobased materials were used as a coagulant aid. The minimum dose combination of ABM and alum that gave a supernatant turbidity of less than 10 NTU was used in filtration experiments.

Table 5. Selection of dose for coagulant and coagulant aid

Coagulant aid	Dose of coagulant aid (mg/L)	Dose of alum (mg/L)	Turbidity remaining after 30 min of settling (NTU)
Surjana seed	10	10	11.5
	10	15	12.6
	10	20	7.2
	10	25	4.4
Maize	10	10	12
	10	15	9.8
	10	20	6.6
	10	25	4.8
Nirmali seed	3	10	13.6
	3	15	14.6
	3	20	12
	3	25	7.2
None		10	22.5
		15	15.4
		20	12.2
		25	11
		35	10.5
		45	8

The volume of settled sludge (after 30 min of settling) produced was: alum + Surjana seed: 1.75 mL/L; alum + Nirmali seed: 1.6 mL/L; and alum alone: 2.8 mL/L, which indicated less sludge production in the case of ABM as a coagulant aid compared to alum alone. The observation was in agreement with the reported literature, that when synthetic polyelectrolytes are used as coagulant aid or alum sludge conditioning agents, they produced quite less sludge (Kawamura 2000; Hudson 1981).

Filtration

Figure 2 presents filtrate turbidity and head loss with only alum as a coagulant. Ripening of the filter was achieved within 60 min which was indicated by the filtrate turbidity. The filtrate turbidity at the beginning of the filter run was 2 NTU and it decreased to 1.1 NTU after 60 min and remained the same up to 150 min. After 150 min, the filtrate turbidity started to increase and attained a value of 2.3 NTU at 240 min. It can be observed from Fig. 2 that head loss profiles are almost parallel up to 120 min. This showed that most of the flocs/particles were retained in the top 5-cm layer with no or very little penetration of flocs in the lower layers. The profiles started to diverge after 150 min indicating deposition/penetration of flocs below a 5-cm depth, which was also indicated by turbidity breakthrough.

Figure 3 presents the filtrate quality and head loss profiles with Surjana seed as a coagulant aid. Ripening of the filter was achieved within 30 min of filter run. Filtrate turbidity was not stable and varied between 0.6 NTU and 0.9 NTU up to 240 min. Head loss profiles were parallel during the filter run indicating no or very little penetration of flocs below a 5-cm depth. Jahn (1981) reported that Surjana seed has a cationic polymer

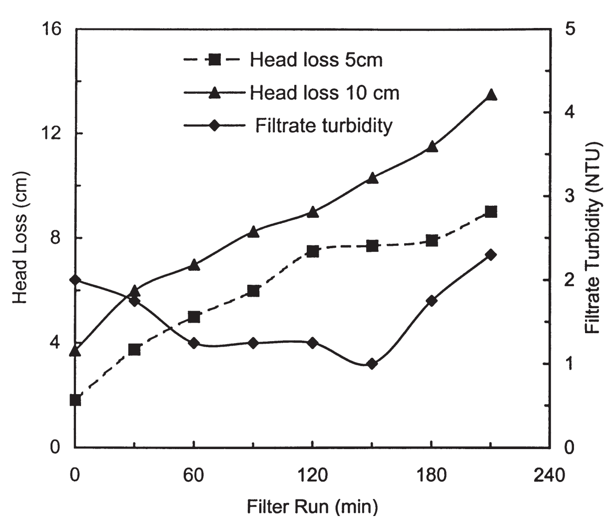

Fig. 2. Filtrate turbidity and head loss with alum.

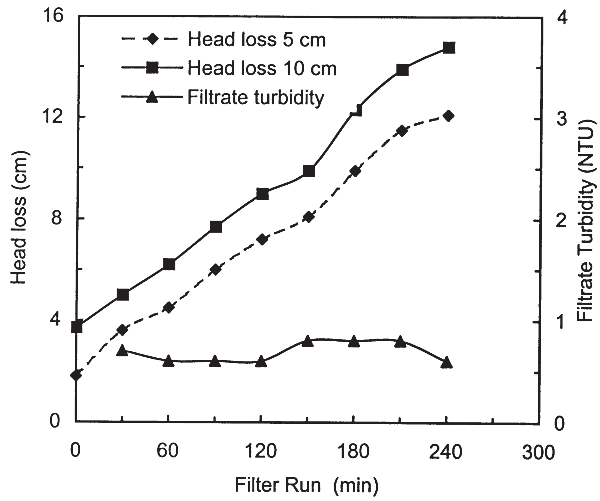

Fig. 3. Filtrate turbidity and head loss with Surjana seed as coagulant aid.

of water-soluble basic polypeptides with a molecular weight of 6000 to 7000 Daltons. Ndabigengesere et al. (1995) reported that the active ingredients in an aqueous *Moringa oleifera* (Surjana seed) extract are dimeric proteins with a molecular weight of 13,000 Daltons. Therefore Surjana seed can be considered to contain low molecular weight cationic polymer. It is known that a low molecular weight polymer is less efficient in turbidity removal compared to a high molecular weight polymer (Stump and Novak 1979) and also that low molecular weight cationic polymer develops less head loss (Zhu et al. 1996).

Figure 4 presents the filtrate turbidity and head loss profiles with Nirmali seed as a coagulant aid. Filtrate turbidity attained a quite low value of 0.2 NTU from the beginning, indicating that ripening was quite rapid. The head loss profiles were parallel indicating deposition in the top 5-cm of filter bed. Nirmali seed extract is an anionic polyelectrolyte which contains carboxyl (COO^-) and hydroxyl (OH^-) as main active groups (Tripathi et al. 1976). The presence of divalent cations in water can greatly increase the ability of anionic polyelectrolyte to aggregate negative colloids (Black et al. 1965). The influent water for filtration in the present study, had in addition to divalent cations, trivalent aluminum, which might have enhanced turbidity removal during filtration.

Figure 5 presents the filtrate quality and head loss profiles with maize as a coagulant aid. Filtrate turbidity at the beginning of the filter run was 0.2 NTU indicating rapid ripening of the filter. Filtrate turbidity decreased to 0.1 NTU as filtration progressed. Head loss profiles were parallel during the filter run. The active ingredient in maize is starch, which is a non-ionic polymer.

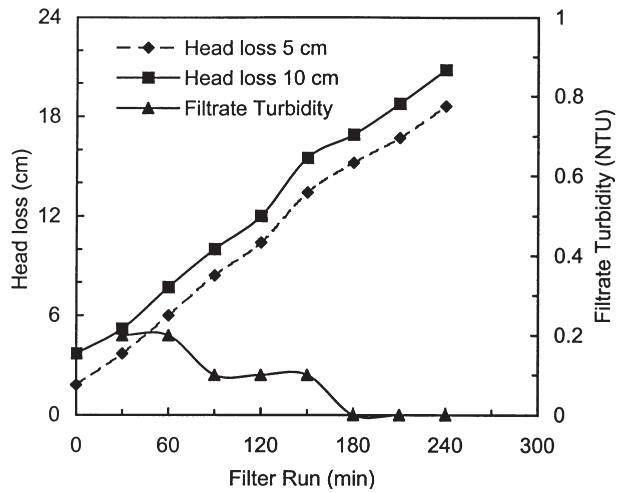

Fig. 4. Filtrate turbidity and head loss with Nirmali seed as coagulant aid.

Filtrate Quality

It is evident from Fig. 2 to 5 that filtrate turbidity was quite less when ABM was used as a coagulant aid in comparison to alum alone. Turbidity breakthrough occurred when alum alone is used as coagulant with no coagulant aid (Fig. 2). It may be that the particulate matter captured with-

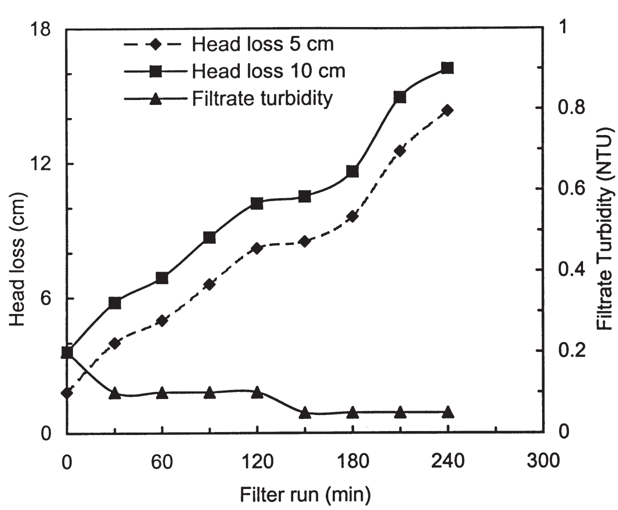

Fig. 5. Filtrate turbidity and head loss with maize as coagulant aid.

in or on the filter bed re-entrained in the water due to shearing of the deposited material. Alum flocs are too weak to withstand high shear forces (Kawamura 2000). McCormic and King (1982) also reported that alum coagulant filter runs exhibited a high rate of turbidity breakthrough with 43% of alum coagulant trials being terminated because of early turbidity breakthrough. Polyelectrolytes in ABM function as a coagulant aid and can neutralize/bridge particles together into flocs. These flocs tend to be more resistant to hydraulic shear forces than alum floc alone and are easier to be captured either on the surface of filter media or within the media, thereby performance of filtration improves (Kawamura 2000).

A lower turbidity of finished water indicates better removal of organic and inorganic contaminants. Many contaminants, such as viruses, heavy metals or some pesticides, may be associated with particulate, and thus efficient removal of particulates can improve the overall water quality (Kavanaugh et al. 1978). Earlier studies have emphasized that at such low finished water turbidity (0–0.1 NTU) the bacterial removal efficiency would also be high. Srinivasan et al. (1998) observed that residual aluminum in treated water consisted mostly of particulate and organic forms of aluminum, implying that lower turbidity of finished water would lead to a reduction of residual aluminum concentration. Driscoll and Letterman (1988) found that high concentrations of aluminum in drinking water were related both with high raw water aluminum concentration and high treated water turbidity. Jekel (1991) reported a correlation between residual aluminum and effluent turbidity. Specifically, residual aluminum concentration was less than 0.1 mg/L when the effluent turbidity was less than 0.1 NTU. As the filtrate turbidity obtained with ABM as coagulant aid is lower than with alum alone, it is probable that finished water aluminum concentrations are reduced when the ABM is used as a coagulant aid.

Head Loss

Head loss development in a filter is just as important as filtrate turbidity to evaluate the filter performance. Based on volume of filtered water produced it was observed that the head loss development in the case of Surjana seed and maize as coagulant aids were comparable with suspension using alum alone, whereas, it was more in the case of Nirmali seeds. The difference observed in head loss development by different coagulant aids may be due to the difference in the sizes of flocs formed. The active ingredient in Surjana seed is a low molecular weight polymer and in maize it is starch (nonionic polymer). It seems that the flocs present in the influent for filtration treated by maize and Surjana seeds were larger, as for the same mass deposition in the filter bed, larger flocs/particles cause less head loss in comparison to smaller ones (Tobiason et al. 1993). Some researchers also mentioned that cationic polymer could be preferred to facilitate the formation of pin flocs, thereby reducing the development of head loss. Zhu et al. (1996) also reported that the use of low molecular weight polymer resulted in less/moderate head loss development. From Fig. 3 to 5 it is obvious that when ABM is used as a coagulant aid, there is less penetration of flocs in comparison to when alum alone is used. The head loss profiles indicated that most of the head loss occurred in the top 5-cm layer of bed and also there was almost no increase in head loss in comparison to clean bed head loss which was observed in lower sections of the filter bed. When polymer is used, the larger and stronger flocs are captured in the top portion of the filter, resulting in most of the head loss occurring near the top of the filter (Zhu et al. 1996).

Conclusions

The following conclusions can be drawn from the study:

1) All the three ABM tested are much more effective for turbidity removal in comparison to alum alone. The filtered water turbidity obtained when Nirmali seed and maize were used as a coagulant aid was approaching zero NTU.
2) Head loss development in filter bed with time was comparable to alum use, in the case of maize and Surjana seeds, whereas, it was more than that of alum for Nirmali seeds.
3) The addition of ABM as a coagulant aid reduces the alum dose significantly.
4) The sludge produced in treatment with Nirmali seeds and Surjana seeds as a coagulant aid with alum was approximately forty percent less than that produced by alum alone.

References

Alfrey AC, Legendre GR, Kaehny WD. 1976. The dialysis encephalopathy syndrome—a possible aluminum intoxification. J. Med. **294:**184–188.

APHA, AWWA, WPCF. 1985. Standard methods for the examination of water and wastewater 16th edition, Washington D.C.

Barnett PR, Skorrgstad MW, Miller KJ. 1969. Chemical characterization of a public water supply. J. Am. Water Works Assoc. **61:**61–67.

Bhole AG. 1990. Performance studies of a few natural coagulants. J. Indian Water Works Assoc. 81–84.

Bhole AG. 1993. Relative evaluation of a few natural coagulants, p. 146–156. *In* Proceedings of the International Workshop on Alternative Methods of Water Treatment, Nagpur.

Bhole AG. 1995. Relative evaluation of a few natural coagulants. J. Water Supply Res. Technol. – Aqua. **44:**184–190.

Black AP, Birkner FB, Morgan JJ. 1965. Destabilization of dilute clay suspension with labeled polymers. J. Am. Water Works Assoc. **57:**1547–1560.

Bulusu KR, Thergaonkar VP, Kulkarni DN, Pathak BN. 1968. Natural polyelectrolytes as coagulant aid. Indian J. Environ. Health **10:**239–264.

CPHEEO. 1991. Manual on water supply and treatment. Ministry of Urban Affairs and Employment, New Delhi.

Crapper DR, Krishnan SS, Dalton AJ. 1973. Brain aluminum in Alzheimer's disease experiment at neurofibrillary degeneration. Science **180:**511–573.

Driscoll CT, Letterman RD. 1988. Chemistry and fate of Al(III) in treated drinking water. J. Environ. Eng. Div. ASCE **114:**21–37.

Fair GM, Geyer JC, Okun DA. 1971. Elements of water supply and wastewater disposal. John Wiley & Sons. New York.

Hoff JC. 1977. The relationship of turbidity to disinfection of potable water. *In* Conf. on Evaluation of Microbial Standards for Drinking Water. U.S. Environmental Protection Agency, Office of Water Supply. Washington D.C.

Hudson HE. 1981. Water clarification processes–practical design and evaluation. Van Nostrant Reinhold. New York.

Jahn SAA. 1981. Traditional water purification in tropical developing countries–existing methods and potential applications. German Agency for Technical Cooperation (GTZ), Federal Republic of Germany.

Jekel MR. 1991. Aluminum in water: how it can be removed? Use aluminum salts in treatment, p. 25–31. *In* Proceedings of Conference of the International Water Supply Assoc., Copenhagen.

Kavanaugh MC, Toregas G, Chung M, Pearson EA. 1978. Particulate and trace pollutant removal by depth filtration. Prog. Water. Technol. **10:**197–215.

Kawamura S. 2000. Integrated design and operation of water treatment facilities. John Wiley & Sons. New York.

Martyn CN, Barker DPH, Osmond C, Harris EC, Edwardson JL, Lacey RF. 1989. Geographical relation between Alzheimer's disease and aluminum in drinking water. Lancet **1:**59–62.

McCormic RF, King PH. 1982. Factors that effect use of direct filtration in treating surface waters. J. Am. Water Works Assoc. **74:**234–261.

Miller GR, Kopfler FC, Kelty KC, Stober JA, Ulmer NS. 1984. The occurrence of aluminum in drinking water. J. Am. Water Works Assoc. **76:**84–91.

Ndabigengesere A, Narasiah KS, Talbot BG. 1995. Active agents and mechanism of coagulation of turbid waters using *Moringa oleifera*. Water Res. **29:**703–710.

Pitchai R, Subramanian R, Selvapathy P, Elangovan R. 1992. Aluminum content of drinking water in Madras City, p. 81–84. *In* Proc. of International Workshop on Aluminum in Drinking Water. Hong Kong.

Rao MN, Sastry CA. 1974. Studies on Nirmali seed extract as a coagulant and coagulant aid. Indian Eng. **18:**17.

Selvapathy P, Vijayaraghavan K. 1994. Turbidity removal using alum and polyaluminum silicate sulphate. Indian J. Environ. Prot. **14:**161–166.

Srinivasan PT, Viraraghavan T, Kardash B, Bergman J. 1998. Aluminum speciation during drinking water treatment. Water Qual. Res. J. Canada **33:**377–388.

Stump VL, Novak JT. 1979. Polyelectrolyte selection for direct filtration. J. Am. Water Works Assoc. **71:**338–342.

Tobiason JE, Jahnson GS, Westerhoff PK, Vigneswaran B. 1993. Particles size and chemical effects and contact filtration performance. J. Environ. Eng. Div. ASCE **19:**520–539.

Tripathi PN, Chaudhuri M, Bokil SD. 1976. Nirmali seed – a naturally occurring coagulant. Indian J. Environ. Health **18:**272–280.

Zhu H, Smith DW, Zhou H, Stanely SJ. 1996. Improving removal of turbidity causing materials by using polymers as a filter aid. Water Res. **30:**103–114.