

Enhanced coagulation: a viable option to advance treatment technologies in the South African context

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Abstract Laboratory and pilot scale tests were conducted to compare the effectiveness of enhanced coagulation with the more advanced technologies of ozone and granular activated carbon in treating a range of clean, eutrophic and industrially polluted waters. Particular attention was paid to the removal of disinfectant by-product precursors, organics and micropollutants that could be achieved using the various types of treatment. Reductions of up to 50% trihalomethane formation potential and between 40 and 70% organic carbon and colour were obtained using enhanced coagulation, which compared favourably with the advanced treatment processes. The more sophisticated processes were especially effective in the removal of micropollutants, this generally being in excess of 70%, which was not achievable using enhanced coagulation. pH depression using acid addition allowed for increases in organics removal at lower coagulant doses and inorganic coagulants were found to be more effective than the polymeric coagulants for organic matter removal. It was shown that the advanced treatment processes became more cost effective for larger plants and as water quality deteriorates, but for smaller water works, enhanced coagulation is cheaper. **Keywords** Enhanced coagulation, ozone, advanced oxidation, granular activated carbon, natural organic matter (NOM).

Introduction

Although South Africa is one of the most developed countries in Africa, in the water treatment field it displays a mixture of First and Third world characteristics. The large cities with established industrial areas have high specific water consumption patterns typical of the First World and the water supply to these areas is treated to International Standards. However in many of the rural areas a Third World situation prevails where low financial resources and technical skills exist. There is a need to treat water to suitable standards at relatively low cost without sophisticated advanced treatment processes which have high capital requirements and require a high degree of expertise for sustained operation.

Umgeni Water is a treatment authority based in Kwazulu-Natal on the East coast of South Africa. Much of the population exists in the large urban areas of Durban and Pietermaritzburg. However Umgeni Water's area extends over 21 000 square km and also contains many poorer areas where severe cost constraints apply. In conjunction with the Water Research Commission a research project was undertaken to compare the efficacy of enhanced coagulation with more advanced technologies and establish whether suitable removal of organic matter could be achieved with relatively low costs and technology requirements.

Although many water authorities in South Africa supply water from impoundments where a degree of self purification takes place, pollution of raw water supplies frequently results in the presence of herbicides, pesticides and other harmful organics as well as eutrophication with subsequent taste and odour compounds produced by algal blooms. These organic compounds, together with naturally occurring organic matter (NOM) can pass through traditional treatment processes and when chlorinated result in the formation of harmful disinfection by-products (DBP). Sophisticated treatment options such as ozonation and granular activated carbon (GAC) are not always viable, especially at smaller water

treatment facilities where both the capital costs and the need for skilled personnel to operate such processes are limited. Enhanced coagulation may enable treatment facilities to significantly reduce NOM at relatively small additional cost and may even obviate the need for more costly and sophisticated treatment processes.

Conventional coagulation is generally defined by the conditions that lead to optimal turbidity removal, rather than optimal NOM removal as is the case with enhanced coagulation (Cheng *et al.*, 1995). The impending USEPA D/DBP Rule, in addition to setting Maximum Contaminant Levels for THMs and haloacetic acids (HAA), establishes best available technologies for the reduction of natural organic matter. Enhanced coagulation, which is defined in the proposed D/DBP Rule as the addition of excess coagulant for the improved removal of DBP precursors by conventional filtration treatment (Crozes *et al.*, 1995), has been introduced as a requirement in the D/DBP Rule. Only a very small fraction of chlorination by-products and the associated health risks have been identified to date and therefore improved precursor removal would reduce both known and unknown risks from the water. The D/DBP Rule has two steps (Crozes *et al.*, 1995); the first two steps TOC removal requirements based upon raw water TOC and alkalinity, obviously the higher the raw water TOC, the greater the percentage TOC removal achievable by enhanced coagulation. Coagulation and softening tests conducted on natural waters, showed that TOC (and therefore NOM) removal is strongly pH dependent (Quaism *et al.*, 1992) and Randtke (1998) found that the optimum pH range for NOM removal was 5.0 to 6.0. Therefore, the higher the alkalinity of the water, the lower the NOM removal achievable using enhanced coagulation.

The impact of enhanced coagulation when used under Southern African conditions has not been assessed. Other factors apart from TOC and alkalinity need to be taken into consideration, such as coagulant type and doses and the nature of the organic matter (Singer and Harrington, 1993). Using inorganic coagulants on their own, it is possible to achieve the optimal pH values for maximum TOC removal, although the addition of acid in conjunction with coagulant allows for a reduction in coagulant dose while still achieving the same TOC removals. Polymeric coagulants however, require the addition of acid in order to obtain pH depression. Careful control of acid addition is needed as poor floc formation and turbidity reduction can otherwise result.

Natural organic matter is usually divided into two major classes: hydrophobic and hydrophilic organic matter. The hydrophobic fraction is generally less soluble, of higher molecular size and contains greater aromaticity than the hydrophilic fraction (Singer and Harrington, 1993) and basically consists of humic and fulvic acids. The humic acid fraction is highly reactive and readily removable by coagulation, while the fulvic acid fraction is less reactive (Randtke, 1988). TOC is usually employed as a surrogate parameter for NOM, although THMFP and other DBP formation potential tests as well as UV absorbance can also be used.

Methods

Laboratory scale enhanced coagulation tests were conducted on three different types of water, namely:

- A eutrophic water containing cyanobacteria (predominantly *Microcystis* and *Anabaena*) in cell concentrations varying between 10 000 and 500 000 cells/ml. Eutrophic conditions were artificially created by spiking water with concentrated cultures of cyanobacteria (TOC approximately 4–8 mg/l).
- A clean water low in organic content (TOC approximately 3–5 mg/l).
- A water high in organic contaminants from an industrial source (TOC between 15 and 35 mg/l).

The treatment processes of coagulation, flocculation and settling were simulated in the laboratory using a jar test procedure. Treated water was then filtered through Whatman No. 1 equivalent filter paper to reproduce the effect of rapid gravity filtration. For turbidity removal, the optimum coagulant dose was considered to be the dose which produced a filtered turbidity of less than 0.5 NTU. The optimum enhanced coagulation conditions were considered to be the coagulant dose at which maximum NOM removal was obtained. A variety of coagulants, including aluminium sulphate, ferric chloride and a number of polymeric organic coagulants, was used over a wide range of concentrations, both with and without the addition of acid (hydrochloric acid) for pH depression.

The pilot plant enhanced coagulation tests were conducted on a water treatment unit providing facilities for coagulation, pulsator clarification and rapid gravity sand filtration. Enhanced coagulation tests using ferric chloride at concentrations varying between 6 and 30 mg/l (as FeCl_3) were undertaken in order to confirm the findings of the laboratory tests.

Laboratory scale ozonation was carried out in a glass contact column (1.57 m high, 10.6 l capacity). A Sorbios ozone generator (GSG 1.2, 1g ozone per hour capacity) was used to generate ozone from oxygen (>99.5% oxygen, <10mg/l moisture) at a pressure of 0.5 bar and a flow rate of 15l/h. The ozone was introduced into the column through a sintered glass diffuser (number 1 diffuser) positioned at the base of the column, while the water in the column was recirculated from the bottom to the top of the contact column to prevent concentration gradients from occurring. Gas exiting the column was fed through a potassium iodide trap before passing through a gas flow meter (Alexander Wright Model Number DM3 B). The apparatus was calibrated by filling the column with a solution of potassium iodide and passing a measured volume of ozone-containing gas through the column.

Pilot plant tests to assess conventional treatment followed by ozonation and GAC were carried out. Provision was made for dosing chemical coagulant and organic contaminants (e.g. atrazine and geosmin) prior to flocculation in a pipe flocculator and filtration through a dual media filter (anthracite and sand). The ozone plant consisted of a SORBIOS ozone generator (GSF 025.2, 25 g/h capacity) and two contact columns which allowed a 12 to 14 minute contact time with the ozone. The ozone was bubbled into the bottom of the contact columns and the off gases from the columns fed through a thermal destructor before being exhausted to atmosphere. Due to limitations in the pilot plant it was not possible to treat the water with pre-ozonation as well as intermediate ozonation. The GAC filter was designed with an empty bed contact time of 10 minutes, a carbon depth of 1.0 m and a filtration rate of 6 m/h and contained a standard grade coal-based GAC for water treatment.

The pilot plants were operated using only the clean water low in organic content as it was not possible to operate them using the eutrophic and polluted water. However, since laboratory tests carried out on the clean water were in close agreement with those of the pilot plants, it can be assumed that the laboratory tests on the other types of water investigated are representative.

Water samples taken before and after the various treatments were analysed for the following.

- Total and dissolved organic carbon (TOC and DOC) using a persulphate-ultraviolet oxidation method (Standard Methods, 1998) on an Aquadoc Total Organic Carbon Analyser. Samples were filtered through 0.45 μm membrane filters (Millex, Millipore) prior to DOC analysis.
- Biodegradable dissolved organic carbon (BDOC) and assailable organic carbon (AOC). BDOC was determined using the method described by Servais *et al.* (1989) and AOC using a method based on that proposed in Standard Methods (1998) with slight modifications introduced from the method proposed by van der Kooij *et al.* (1982).

- Trihalomethane formation potential (THMFP) using the test described in Standard Methods (1998) and measuring THM on a Varian 3600 gas chromatograph using direct aqueous injection with a suitable thermal programme and an internal 1,2-dibromomethane standard.
- UV absorbance at 254 nm. This was analysed after filtration through 0.45 μm membrane filters (Millex, Millipore) using a Pharmacia Ultraspec III spectrophotometer and Autofill III autosampler with a 10 mm quartz cell.
- Geosmin and 2-methylisoborneol (2-MIB). These were extracted from 1 litre of sample using three 50 ml portions of dichloromethane, which were concentrated to 1 ml and then measured on a Hewlett-Packard 5890/5970 gas chromatograph-mass selective detector according to an NLA accredited procedure.
- Atrazine. This was extracted by the same procedure as for geosmin and measured using a Hewlett-Packard 5890/5970 gas chromatograph-mass selective detector according to an Umgeni Water method.
- Turbidity, pH and alkalinity. Turbidity was measured on a Hach Ratio/XR model 43900 turbidity meter, pH was determined using a Radiometer PHM 95 pH/ion meter and alkalinity was measured using a Mettler DL25 Autotitrator using 0.02N HCl and titrating to pH 8.2 and 4.3 according to an NLA accredited method.
- Colour. Samples were filtered through a 0.45 μm membrane filter (Millex, Millipore) and colour was then measured relative to a chloroplatinate standard at 400 nm on a Shimadzu UV 2100 spectrophotometer according to an NLA accredited procedure.

Fractionation of the NOM in the polluted water was also carried out using an Amberlite XAD-16 resin (Rohm and Haas, Philadelphia, PA) for adsorption chromatography according to the procedure described in Thurman and Malcolm (1981).

Results and discussion

Enhanced coagulation

The term natural organic matter (NOM) is used to describe all organic compounds occurring in water, both in dissolved and particulate form and including anthropogenic compounds (Owen *et al.*, 1995). Since NOM impacts so significantly on all aspects of water treatment, it is important to monitor it properly after the different treatment processes, but NOM is very difficult to characterise and therefore surrogate parameters have to be used.

THMFP. In terms of THMFP removal, enhanced coagulation was often found to have little or no effect, but in higher DOC waters, such as eutrophic and industrially polluted waters, THMFP reduction of around 50% was possible using ferric chloride at concentrations of 1.5 to 5 times the optimum turbidity removal dose. This was confirmed in pilot scale studies in which THMFP removals of up to 40% were obtained when using ferric chloride for enhanced coagulation of a low TOC (3–5 mg/l) water at doses of between 6 and 30 mg/l (i.e. 2 to 5 times the optimum dose for turbidity removal). Removals of between 50 and 75% of DBP have been cited in the literature (Amy and Chadik, 1983; Babcock and Singer, 1979; Reckhow and Singer, 1984), although this depends on the nature of the organic matter present in the water.

TOC and DOC. Enhanced coagulation of eutrophic and low TOC waters yielded TOC and DOC removals of up to 50% when using the inorganic coagulants (ferric chloride and alum), see Figure 1. These removals were obtained at coagulant doses in the region of 1.5 to 5 times the dose required for optimal turbidity removal. The polymeric coagulants did not bring about more than approximately 10% TOC and DOC removal, even in highly

eutrophic and polluted waters using a coagulant dose up to 6 times that required for optimal turbidity removal. The TOC of these waters consisted predominately of DOC and as a result the effect of enhanced coagulation on the DOC content of the water mirrored that for the TOC content.

The pilot scale tests carried out on the low TOC water using ferric chloride confirmed the results of the laboratory tests. At ferric chloride doses of 8 to 18 mg/l, TOC and DOC of approximately 20 to 40% were obtained. Increasing the dose to 30 mg/l FeCl_3 , it was possible to achieve between 50 and 75% removal of TOC and DOC. The optimum dose for turbidity removal was 6 mg/l.

Better TOC and DOC removals were obtained for enhanced coagulation of an industrially polluted water. The TOC and DOC of this water was between 15 and 35 mg/l. It was possible to achieve TOC and DOC removals of 60% and more from these water samples when using ferric chloride and alum. The optimal coagulant dose for organic carbon removal was between 1.5 and 7 times the optimal dose for turbidity removal.

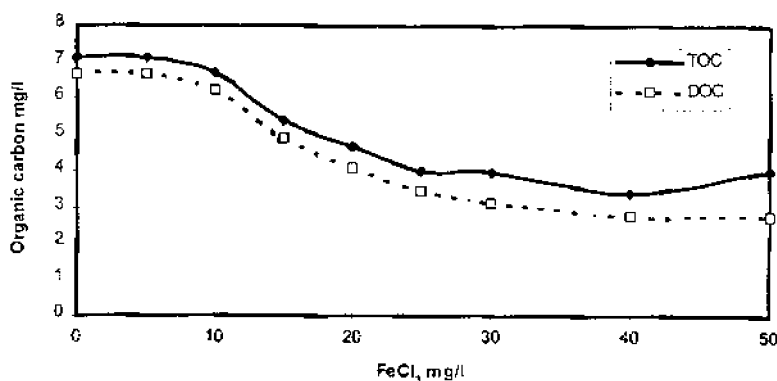


Figure 1 Effect of enhanced coagulation on the TOC/DOC of a eutrophic water (10 000 *Anabaena* cells/ml).

BDOC. Removal of BDOC was generally good (30 to 50%) at the inorganic coagulant doses for optimal turbidity removal, however it was possible to obtain between 70 and 90% removal by increasing to between 2 and 5 times the dose for optimal turbidity removal. Ferric chloride generally gave better BDOC removals than alum, while the organic polymers resulted in little or no reduction in BDOC. Biodegradable dissolved organic matter present in a water can be determined using either the BDOC or assimilable organic carbon (AOC) tests, although these two tests are essentially different, the BDOC being the portion of DOC than can be mineralised by heterotrophic micro-organisms, while AOC is the portion of DOC that can be converted to biomass (Huck, 1990). Although measured in quite different units, the results obtained from the two different test procedures correlated well.

UV absorbance. The UV absorbance followed the same trend as both the turbidity and the TOC/DOC. In low DOC waters (< 7 mg/l), almost complete UV extinction at 254 nm was obtained at the optimal inorganic coagulant doses for turbidity removal. The organic polymeric coagulants were not as effective in reducing UV absorption, but were able to reduce the UV by as much as 60 to 80%. However, polymeric coagulant doses higher than the optimum for turbidity removal, gave little or no additional UV removal. Reductions in UV absorbance in eutrophic waters and industrially polluted water was usually between 50 and 75% at the optimal alum and ferric chloride doses for turbidity removal. Using between 2 and 4 times these concentrations could increase UV removals to between 70 and 90%. The

results of the pilot plant investigation confirmed the laboratory results, indicating that UV removals in the region of 50 to 75% could be obtained at ferric chloride doses of between 8 and 18mg/l (i.e. 2 to 3 times the optimal turbidity removal dose). Reductions in UV absorbance of between 80 and 90% were obtained when the ferric chloride dose was increased to around 30mg/l (± 5 times the optimal dose for turbidity removal).

Micropollutants and other determinants. Enhanced coagulation using ferric chloride, alum or a polymeric coagulant did not give rise to any significant removal of geosmin, 2-MIB or atrazine, but colour removal of between 50% and 100% was possible when using the inorganic coagulants. These results have been confirmed in the pilot plant studies where the colour was reduced from as high as 11.5° Hazen to less than 1° Hazen at ferric chloride concentrations of between 6 and 30mg/l. Enhanced coagulation was also found to be effective for the removal of algal cells. This is to be expected as “sweep coagulation” which is similar to enhanced coagulation has been used for many years for algal cells and particle removal from water. Pilot plant studies have confirmed the laboratory tests with an excess of 90% and often complete removals of algal cells occurring at ferric chloride doses of between 10 and 30mg/l, see Figure 2.

Fractionation of the NOM was only carried on on the industrially polluted water. The NOM of this water was found to consist of almost equal quantities of hydrophilic and hydrophobic compounds. Recoveries of DOC after XAD-16 fractionation were typically around 80%. Enhanced coagulation was found to be more effective in removing the hydrophobic fraction, with removals of between 50 and 75%, although removal of the hydrophilic DOC compounds was also fairly good (35 to 70%). This is in agreement with other researchers who reported that hydrophobic organic compounds are more readily removed than the hydrophilic compounds during coagulation (Babcock and Singer, 1979; Jewel, 1985; Collins *et al.*, 1986). Ozonation increased the proportion of hydrophilic DOC relative to the hydrophobic by less than 10%. Furthermore ozonation was often found to have a detrimental effect on the removal of the hydrophobic compounds by enhanced coagulation, although removal of the hydrophilic compounds did not appear to be affected. This implies that the fraction of hydrophobic DOC that is most easily removed during coagulation is the same fraction that appears to be oxidised by ozone. It could also be due to ozone reducing the molecular weight and increasing the polarity of the hydrophobic DOC (Edwards *et al.*, 1994; Jekel, 1994; Reckhow and Singer, 1991), both of which would adversely affect its removal by coagulation (Edwards *et al.*, 1994; Jekel, 1994).

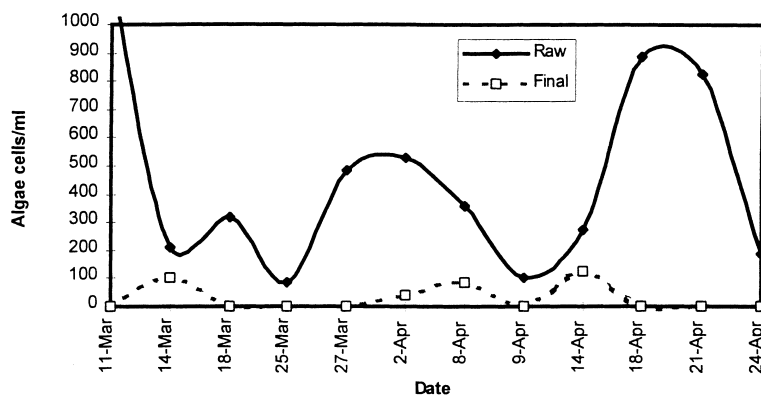


Figure 2 Pilot plant results depicting the effect of enhanced coagulation on the algal cell concentration.

Effect of pH on enhanced coagulation. Tests were carried out to assess the effect of pH on enhanced coagulation. It was found that the optimal pH for turbidity removal is higher than that for optimal organic carbon removal when using the inorganic coagulants. The optimal pH for turbidity removal was between 5.5 and 7 when using ferric chloride and between 6 and 7 when using alum, which is in agreement with the literature (Water Treatment Plant Design, 1978). Organic carbon removal was best at a pH of around 5 when using ferric chloride and at between 5 and 5.5 when using alum, which is in agreement with the findings of Randtke (1988). If the pH dropped below 4 solubilisation of iron and manganese occurred and removal of turbidity, UV and organic carbon concentrations deteriorated. Using pH depression the removal of DOC could also be increased by between 15 and 25% when using inorganic coagulants. These could be obtained by keeping the coagulant dose constant and reducing the pH of the water with acid (acid was added prior to coagulant to produce a pH after coagulant addition of approximately 5 for ferric chloride and between 5 and 5.5 for alum). For example an alum concentration of 10 mg/l when used to treat a water with a DOC concentration of 7.3 mg/l produced a final treated water with a pH of 7.3 and had no effect on the DOC. Adding acid to the water prior to coagulation to produce a final treated water with a pH of 5.6 resulted in a DOC of 5.7 mg/l, a decrease in DOC of 22%. Krasner and Amy (1995) observed a similar effect. These results imply that significant coagulant cost savings are possible if pH adjustment is used, although the cost of the acid and the extra complication of adding two chemicals to the system needs to be considered.

Comparison of enhanced coagulation and advanced treatment technologies

Laboratory and pilot plant scale tests were carried out using the same water types as those used for the enhanced coagulation investigation. Pilot plant tests in which water was first treated conventionally, followed by ozonation and GAC filtration could affect a THMFP reduction of 40%, TOC and DOC removals of 15 to 30% and reductions in BDOC in excess of 80%. The advanced treatment pilot plant tests also brought about good reductions in colour and algal cells, effecting complete removal in many cases. However, enhanced coagulation was as effective as, if not better than the advanced treatment processes in reducing all these contaminants. Enhanced coagulation was less effective in the removal of micropollutants, such as geosmin, 2-MIB and atrazine. Advanced treatment processes were able to reduce these by over 70%, while enhanced coagulation did not result in any significant removal of these contaminants.

Cost assessment

A cost assessment was carried out based on three typical situations in which ferric chloride was used. The first was for a clean water low in organic carbon content (TOC 3–5 mg/l), the second was for a eutrophic water containing approximately 10 000 *Microcystis* cells/ml (TOC approximately 8 mg/l and the third was for a water polluted with industrial effluents (TOC approximately 15 mg/l). Based on optimal doses as shown in Table 1, a ferric chloride cost of \$75.00 per tonne at 14.3% iron, a sludge production factor of 0.65 (Geldenhuis, 1992) and estimating lime requirements assuming 50% precipitation, the costs shown in Table 1 were obtained. Sludge treatment and disposal costs of \$6–70 per 7 m³ load at 25% solids are based on plant experience.

Cost assessments were also conducted for treatment works in which conventional treatment was followed by ozonation and GAC filtration. For a 350 MI/d plant at a dosage of 5 mg/l, capital costs were estimated at approximately five million dollars amortised over 15 years at 15%. Including operating costs yielded an overall figure of \$0–89/MI for ozonation. Costs for GAC filters for a 350 MI/d plant were based on a 20 year amortisation for

Table 1 Cost assessment comparing advanced treatment processes and enhanced coagulation for 350 and 175 ml/d plants.

Water	Advanced Treatment Processes					Enhanced Coagulation	
	Coagulant Dose mg/l	Coag cost \$/MI	Ozone \$/MI	GAC \$/MI	Total cost \$/MI	Coagulant dose mg/l	Total cost \$/MI
350 MI/d Plant							
Clean	4	1–14	0–89	19–33	21–36	12	3–40
Eutrophic	25	7–08	0–89	19–33	27–30	60	16–99
Polluted	60	16–99	0–89	19–33	37–21	106	45–46
175 MI/d Plant							
Clean	4	1–14	1–39	29–00	31–53	12	3–40
Eutrophic	25	7–08	1–39	29–00	37–47	60	16–99
Polluted	60	16–99	1–39	29–00	47–38	160	45–46

civils and 15 years for mechanicals and electricals. The first fill of GAC was capitalised. Using an interest rate of 15%, an overall GAC cost of \$19–33 was calculated. Costs for a 175 MI/d plant based on the same criteria used above, yielded ozone costs of \$1–39 and GAC costs of \$29–00.

This implies that advanced treatment processes become economically attractive at large works when treating relatively polluted waters. In all other cases enhanced coagulation is more attractive.

Conclusions

The following conclusions were drawn.

- Enhanced coagulation removes up to 40% THMFP, 60% TOC and DOC, 70–90 BDOC and UV absorbance and up to 100% colour and algal cells. This is at least as good as the advanced treatment processes.
- Enhanced coagulation is not effective for the removal of micropollutants, in which case more advanced treatment technology, such as ozonation and GAC filtration is required.
- Inorganic coagulants are better than organic polymeric coagulants for enhanced coagulation.
- Using enhanced coagulation, optimal NOM removal is achieved at coagulant concentrations of between 1.5 and 7 times the optimal dose for turbidity removal, depending on the NOM type and concentration.
- Enhanced coagulation is cost-effective at smaller works but advanced treatment becomes competitive on large works (± 175 MI/d and larger) and as water quality deteriorates.

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