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Effects of polydiallyldimethyl ammonium chloride coagulant on formation of chlorinated by products in drinking water

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ABSTRACT

The objectives of this research work was to evaluate the reduction of THM precursors by cationic p-DADMAC and determine the correlations between the chlorine demand and trihalomethane formation in the presence of electrolyte solutions and ambient light. The chlorine demand was found to be significantly reduced provided that the H₂SO₄ electrolyte was fed to the sample solutions. The amount of CHCl₃ formation was also decreased when the Na₂SO₄ electrolyte was introduced in spite of the levels of light intensity. The p-DADMAC can not only effectively remove the turbidity but also reduce the formation of CHCl₃. The optimum dosage of p-DADMAC for reducing the turbidity, TOC and CHCl₃ in the humic acid and source water samples was determined and depended upon the nature of organics. ©1999 Elsevier Science Ltd. All rights reserved

Key words : Polyelectrolyte, Polydiallyldimethyl ammonium chloride (p-DADMAC), cationic polymer, Trihalomethane (THM), TOC, Chlorine demand.

INTRODUCTION

The polyelectrolyte coagulants (polymer) used in water treatment are high molecular-weight, synthetic organic compounds that have a strong tendency to be adsorbed on the surfaces of the particles in an aqueous suspension. Cationic polyelectrolytes can be classified as primary coagulants, and nonionic and anionic polyelectrolytes which also can be referred to either as coagulant aids or flocculants [1]. Stringent filtered-water quality requirements resulting from the 1986 amendments to the State Drinking Act may lead to the increased use of polyelectrolytes for water treatment in the United States. Enhanced coagulation technique has been successful in removing natural organic matters (NOMs) at the water treatment plants [2~7].

Polyelectrolyte formulations contain contaminants from the manufacturing process such as residual monomers, other reactants, and reaction by-products that could potentially affect human health [1]. Velta reported the cationic polyelectrolyte in the finished water appeared as the polymer and monomers, which could be reacted with the added chemicals (e.g., chlorine) and/or the organic compounds in raw water to form new substances [8]. It was also noted that degradation of these polyelectrolytes in drinking water reservoirs may occur due to interaction with chemicals such as chlorine [9-11]. Because of concern about certain hazardous contaminants in the treated water, Switzerland and Japan do not permit the use of polyelectrolytes in the drinking water treatment, West Germany and France have established stringent limits on application rates of polyelectrolytes in water treatment plants [1].

However, in some cases, this risk is not associated with the polyelectrolyte itself but unreacted monomer residues. For example, Mallevialle (1984) found that chlorination of polyacrylamide and acrylamide monomers showed low reactivity, and generated a small amount of total organic halides (TOX) and trihalomethane which had an insignificant effect on the mutagenicity of the polymer solution [12]. Polymers can not effectively remove dissolved organic matters and, however, would react with chlorine resulted in having negative effects on coagulation. Polymers decreased their molecular weight and formed organic halides as by-product of chlorination. Furthermore, acrylamide monomer which was an impurity of polymer formed chloroform and 2,3-dichloropropionic acid by chlorination [13].

Polydiallyldimethylammonium chloride (p-DADMAC), as a quaternary ammonium salt, is usually described as a chlorine-resistant polymer. Only limited information on the degradation and removal of p-DADMAC constituents was reported in the literature. According to the AWWA standard for p-DADMAC, the DADMAC monomer (m-DADMAC) contained in the p-DADMAC product should be less than 0.05 mg m-DADMAC per milligram of polymer. The maximum allowable dosage of polymer in the source water at the water treatment plant is 10 mg [14]. Hason et al. reported that the maximum intake of DADMAC from drinking water was estimated about 700 mg per person per year [15]. Kasier and Lawrence chlorinated the non-ionic polyacrylamide under the thermal and UV light- activation conditions. The chlorine dose was fed at 10 mg/L level with the UV light activation, the chloroform yielded about 10.9 $\mu\text{g/L}$. When the thermal activation was performed for 4 hrs, chloroform was not-detectable [9].

With the rapid development of industrialization and urbanization in Taiwan, the source water quality is deteriorating and can not be easily treated to meet the national drinking water standards by the conventional method. If the polymers prove to be safe under appropriately regulated conditions, Taiwan waterworks will be authorized to use them in water treatment plants. In general, there is limited information on the chemical compositions and impurities of commercial polyelectrolytes formulations and their removal by the treatment processes. Fortunately, the USEPA promulgated the ANSI/NSF standard 60 and proposed minimum requirements of water treatment chemicals to prevent the adverse effects on human health [16]. However,

further studies on cost-effective analyses have to assess their benefits and potential health related issues caused by the polymers reacted with NOMs and chlorine. Thus the objectives of this research work were intended to evaluate the reduction of THM precursors by the cationic p-DADMAC as a primary coagulant in the presence of higher turbidity water. Specific studies were focused on the chlorine demand and trihalomethane formation between the p-DADMAC and chlorine in the presence of electrolyte solutions and ambient light.

EXPERIMENTAL METHODS

1. Preliminary tests on chlorination of p- and m-DADMAC

The chlorine demand and CHCl_3 formation of p-DADMAC (10 and 50 mg/L) and m-DADMAC (50 mg/L) were measured under the specific chlorine dose (2.5 and 6.0 mg/L) and contact time. Further studies were performed to evaluate the environmental factors, i.e., ionic strength and light intensity affecting the chlorine demand and CHCl_3 formation during the chlorination process. To understand the effects of ionic strength on chlorine demand and CHCl_3 formation of 10 mg/L p-DADMAC, two typical electrolytes, i.e., H_2SO_4 and Na_2SO_4 (0.001 to 0.005 mol/L) were introduced in the samples, respectively. The ambient light intensity (50 to 500 mW/cm²) was determined by UVX Radiometer (UVP, Inc.) in the course of coagulation process (Run #1, #2 and #3). The wavelength in the ambient light intensity were referred to 254, 310 and 360 nm.

2. Coagulation tests on chlorination of p- and m-DADMAC

In the prechlorination procedure, both the humic acid (Aldrich co.) and source water samples from the Pan-Hsing water treatment plant were first added the bentonite to increase the turbidity, and then chlorinated by sodium hypochlorite at a dosage about 12 mg/L as Cl_2 concentration. The prepared samples were mixed with p-DADMAC (Aldrich co.) at 180 and 40 rpm for 3 and 15 minutes, respectively, during the coagulation tests. After 2 hours, the supernatant from the reactor was collected and analyzed. For the postchlorination procedure, both the humic acid and source water samples were chlorinated after the slow mixing procedure.

3. Evaluations of Chlorine Demand [17]

An amount of appropriate chlorine solution was first added to the 200 mL sample and then mixed. The residual free chlorine was measured after a specific time interval. If free chlorine residual was lower than 1.0 mg/L, an additional amount, typically, 3 to 20 mg Cl_2 /L, was added to establish a definite residual (Standard Methods, 2350B). All samples were subjected to a chlorine demand analysis at room temperature.

For each run, a blank test was performed at the same pH as the chlorinated water. The chlorine demand was subtracted from the chlorine consumption observed for the water samples. Total residual chlorine and free residual chlorine were analyzed by the DPD titration method (Standard Methods, 4500-C1 F). The chlorine dosing solution was prepared from a 12.5 percent reagent grade stock solution of sodium hypochlorite. The chlorine concentration required in the samples in order to minimize dilution error.

4. Determinations of Trihalomethane (THMs) Formation

The appropriate chlorine dose of water sample was determined from 3.5 mg Cl₂/L to 0.5 mg Cl₂/L based on Standard methods, 5710B. The target free chlorine residual was controlled in the range of 1.1 to 3.5 mg/L after 7 days of incubation at 20 ± 2°C. For the THMs formation experiments, samples were taken at 0.5, 1, 2, 4, 8, 24, 48, 72, 96, 120, 144 and 168 hours after chlorine addition. The pH of the raw samples was controlled with 5 mL phosphate buffer solution and temperature was held at 20 ± 2°C throughout these chlorination experiments. These treated samples were immediately sealed with a TFE-lined screw cap and then stored in the dark room. THMs was analyzed by HP 6890 GC/ED (Standard methods, 6230D).

5. Measurements of TOC and UV₂₅₄

Water samples for TOC and UV₂₅₄ analyses were first filtered through a pre-washed glass fiber filter and then determined by the TOC instrument (O. I. Corp. model 700) and UV spectroscopy (Hitachi U-2000). All analyses, unless otherwise noted, were performed according to the 19th edition of Standard methods [18].

RESULTS AND DISCUSSIONS

1. Effects of p-DADMAC on chlorine demand and CHCl₃ formation

Figure 1 presents the chlorine demand of p- and m-DADMAC measured under the specific chlorine dose and contact time. In general, the chlorine demand of monomer exhibits the stable phase after the chlorination for 100 hrs. It was evident that the chlorine-resistance occurred in the presence of Cl₂ demand less than 0.1 mg/L which indicates the unsaturated double bond of the dilute m-DADMAC did not react with chlorine to form vicinal halohydrins. The reactions between the chlorine and the fed concentration of p-DADMAC were shown in Figure 1 which clearly demonstrates that the stoichiometric relationship between them was held. It implies the dose response curve between the chlorine demand and concentration of p-DADMAC was occurred. This was not inconsistent with the findings suggested by Kaiser et. al. in which cationic polyelectrolytes were degraded by chlorine. Chlorine can react with polymers by the addition and substitution mechanisms to generate the disinfection by-products (DBPs). For the DBPs concerns, there were only two THMs species, i.e., chloroform and bromodichloromethane which were detected by GC/ECD

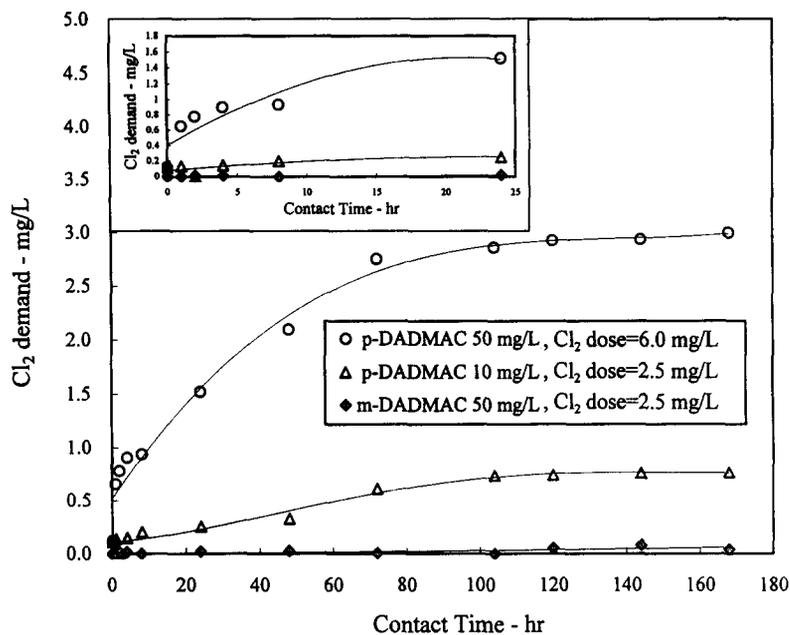


Figure 1. Chlorine demand of DADMAC measured under the specific chlorine dose and contact time

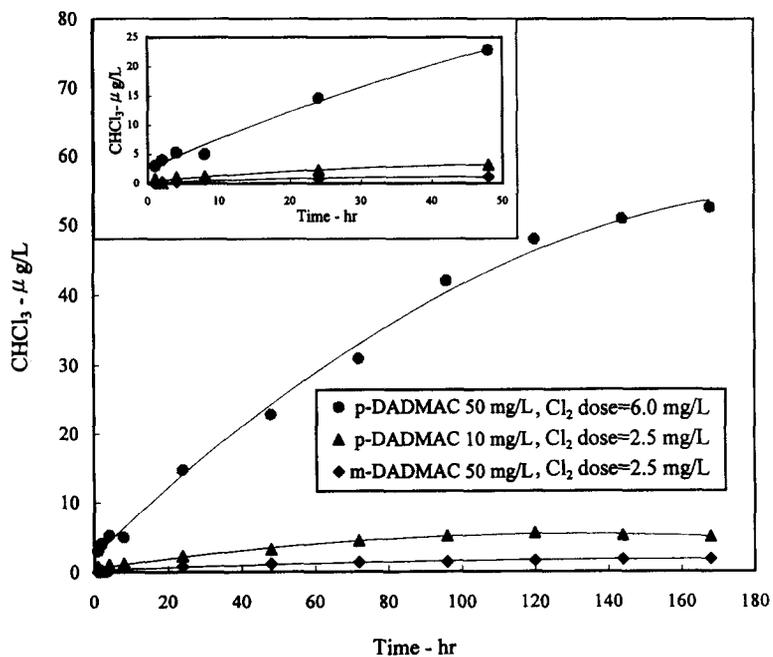


Figure 2. Effect of chlorine contact time on chlorform formation at various levels of p-DADMAC and chlorine dose

and GC/ELCD analyses. The rest DBPs such as haloacetonitrile (HANs) and chlorinated volatile organic carbons (VOCs) were not found. The relatively lower measured bromodichloromethane concentration, i.e., less than 2 $\mu\text{g/L}$ (MDL), could be ascribed to the fact which a trace amount of impurity of bromide were observed in this monomer.

The effects of chlorine contact time on chloroform formation for the DADMAC are shown in Figure 2 which indicates that the very lower formation of CHCl_3 is occurred between the reaction of m-DADMAC and chlorine. In comparison of Figures 1 and 2, it was also observed that the rate of CHCl_3 formation was lower than that of chlorine demand. Especially, when the higher dose of p-DADMAC was introduced at 50 mg/L for 7 days, where the CHCl_3 formation approached the steady state under which a certain amount of CHCl_3 could be generated throughout the chain-reaction.

However, it was noted that in spite of the concentration of DADMAC, there was a linear correlation (Type I) between Cl_2 demand ($< 2.8 \text{ mg/L}$) and CHCl_3 formation ($< 30 \mu\text{g/L}$) shown in Figure 3, until the chlorine demand reached to 3.0 mg/L (Type II). Type I reaction indicates the stoichiometric ratio is held between the two major reactants, i.e., chlorine and p-DADMAC; while Type II reaction shows the higher CHCl_3 formation potency is occurred due to the successive reactions among the reactants, substituted and/or decomposed products.

2. Effects of ionic strength and light intensity on chlorine demand and CHCl_3 formation for p-DADMAC

The effects of ionic strength on chlorine demand of 10 mg/L p-DADMAC were presented in Figure 4 which indicates the chlorine demand decreases as the ionic-strength increases. The chlorine demand was also found to be significantly reduced if the H_2SO_4 electrolyte was fed to the sample solutions. Similar observations regarding the CHCl_3 formation were also shown in Figure 4 which confirms the general principle, i.e., the more acidic condition, the less THMs formation. Since these reactions were performed under the excessive Cl_2 -dose conditions, these phenomenon could be interpreted by the evidence that the polymer become more stable and resulted in reducing the chlorine demand in the electrolyte solutions. These observations were consistent with the findings reported by Butler et. al. who suggested the fluoride ion could be utilized for accelerating the polymerization rate in the entire chemical reaction [19].

Figure 5 gives the changes of ambient light intensity at various wavelength in which the Run #2 possesses the higher light intensity than the Run #1 and Run #3. The higher light intensity measured in the Run #2 reflected the larger amount of Cl_2 -demand and CHCl_3 -formation presented in Figure 6. At various levels of ambient light intensity (Figure 6), the chlorine demand and CHCl_3 formation increases as the light intensity increases within 4 hrs reaction time. In general, the sequence of the formation of CHCl_3 and the chlorine demand in the various experiments are strictly dependent upon the level of ambient light intensity,

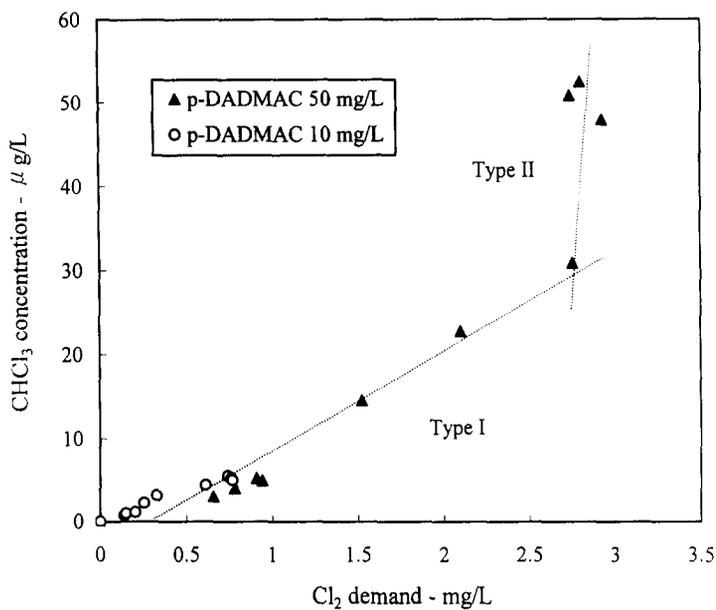


Figure 3. Relationship between chlorine demand and chloroform formation of p-DADMAC at various contact time.

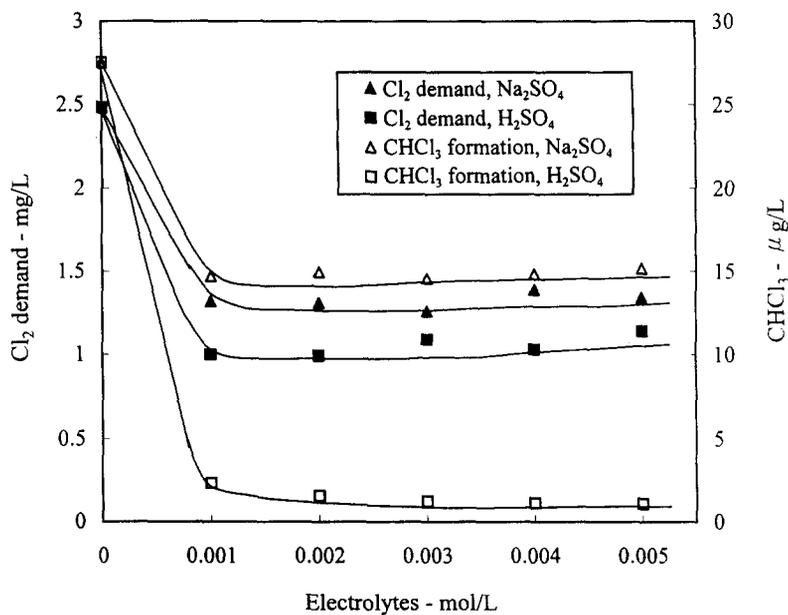


Figure 4. Effects of ionic strength on Cl₂-demand and CHCl₃ formation of p-DADMAC without introducing light intensity

i.e., Run #2 > Run #3 > Run #1. In comparison with the Cl_2 -demand and/or CHCl_3 formation among the 254, 310, and 360 nm wavelength, the 254 nm (UV light) might react with chlorine to form the free chlorine radicals resulted in producing a relatively higher amount of CHCl_3 formation. From the above discussions, it was evident that the effect of light intensity on Cl_2 -demand was greater than that of the Na_2SO_4 electrolyte but lower than that of the H_2SO_4 electrolyte at the higher dose.

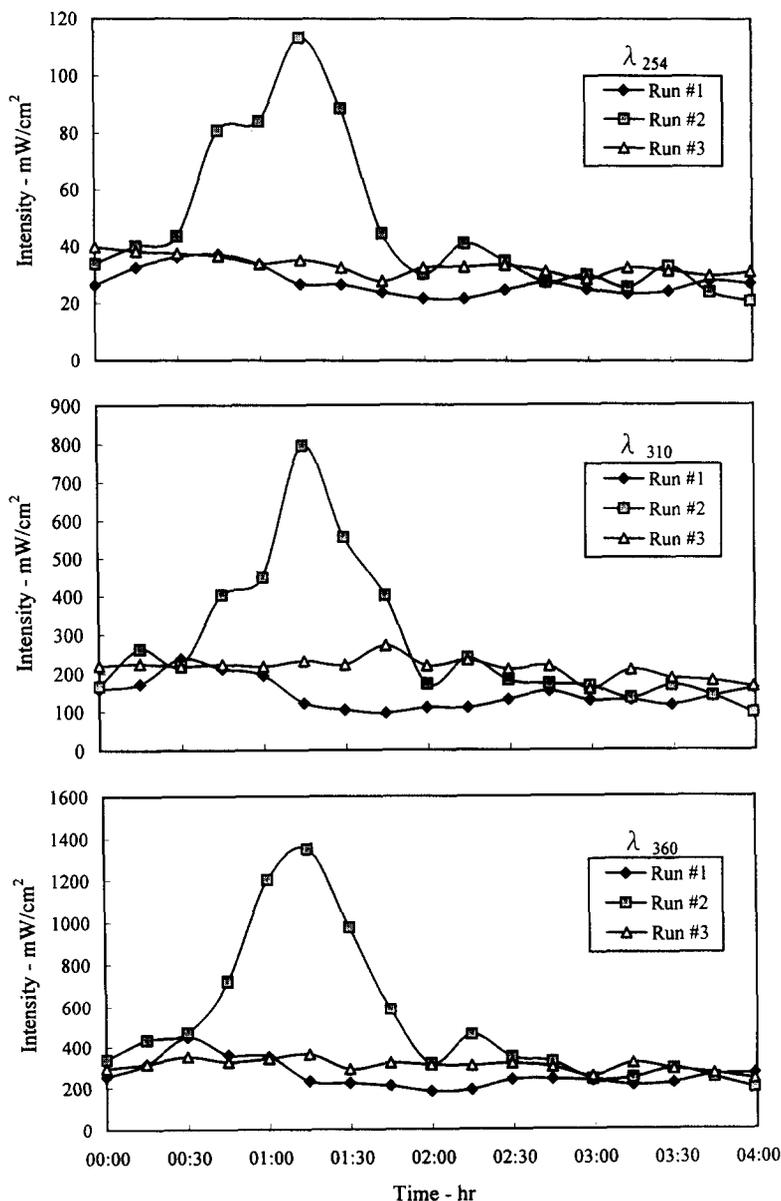


Figure 5. Changes of ambient light intensity in the course of coagulation at 10 mg p-DADMAC/L

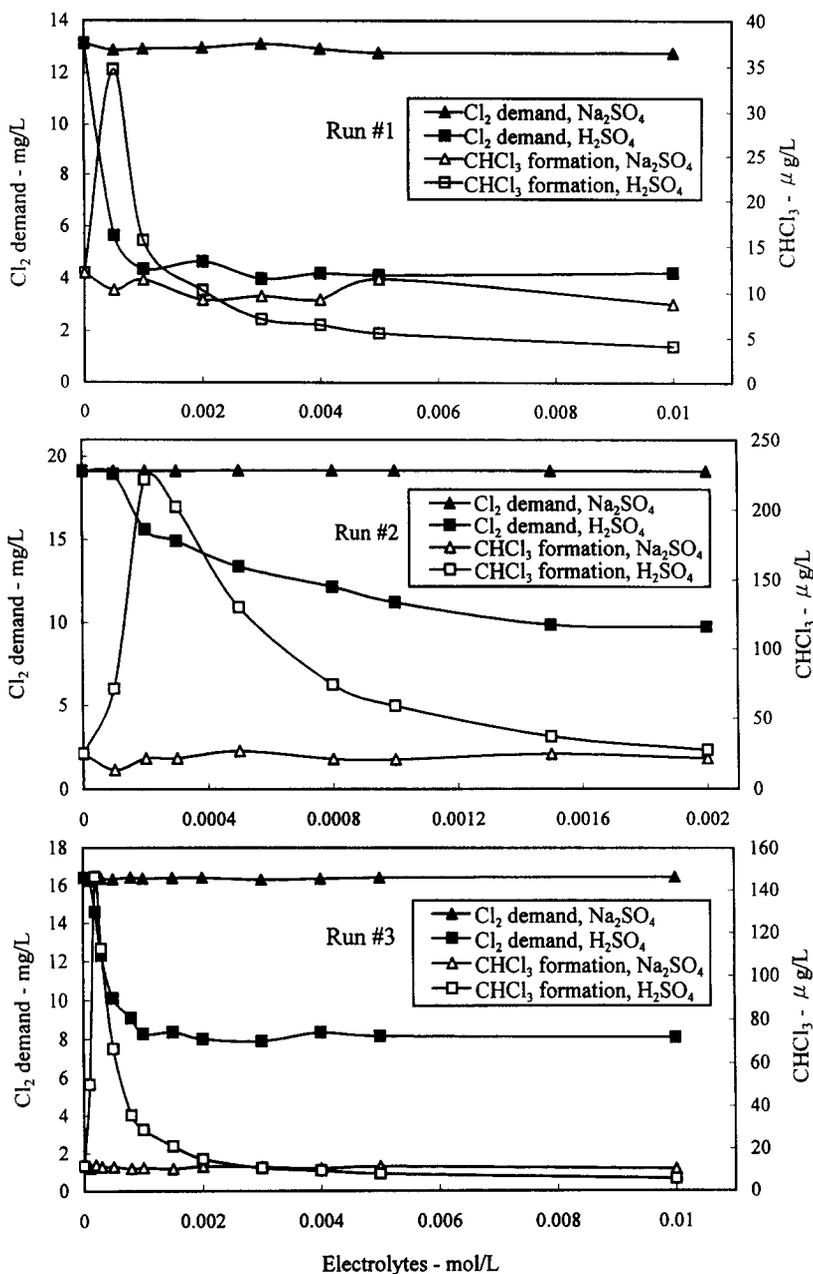


Figure 6. Effects of ionic strength on Cl₂-demand and CHCl₃ formation of p-DADMAC at various levels of ambient light intensity

Figure 7 presents the correlations of Cl₂-demand and CHCl₃ formation for p-DADMAC in Na₂SO₄ and H₂SO₄ electrolytes. The effects of H₂SO₄ electrolyte on CHCl₃ formation (or Cl₂-demand) were in a broad and wide range shown in Figure 7 which indicates the more complicated reaction mechanisms in the

presence of H_2SO_4 electrolyte. It was also observed that the amount of CHCl_3 formation was significantly reduced if the experiments were performed under the Na_2SO_4 electrolyte conditions regardless of the levels of light intensity (Figure 7).

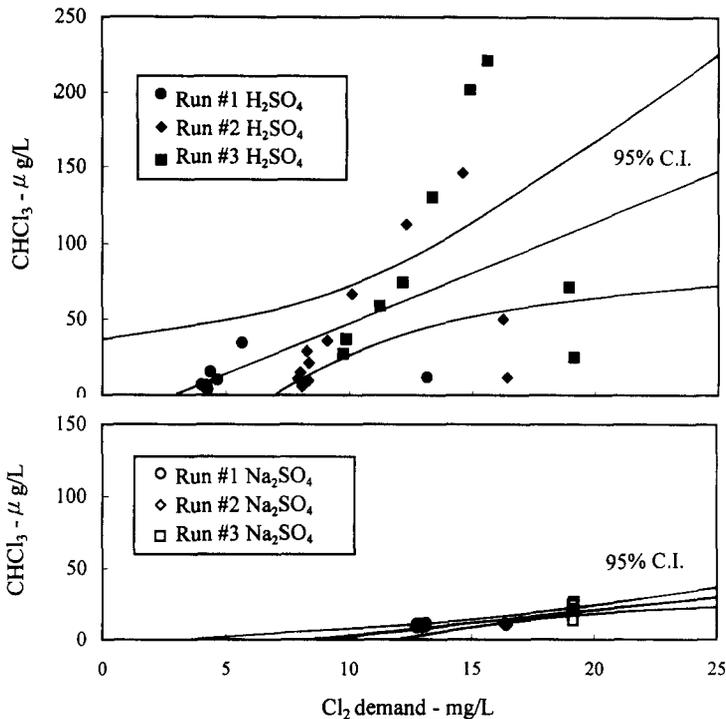


Figure 7. Correlations of Cl_2 -demand and CHCl_3 formation for p-DADMAC in Na_2SO_4 and H_2SO_4 electrolytes

3. Effects of p-DADMAC on reductions of turbidity, TOC and CHCl_3 for the humic acid and source water samples

Table 1 and 2 present the effects of p-DADMAC on reductions of turbidity, TOC, and CHCl_3 for the humic acid and source water samples respectively, in the course of prechlorination and post-chlorination procedures. Without introducing any polymers in the coagulation tests, the turbidity and TOC can be reduced to 25 and 18% respectively, for the humic acid solutions at the end of the prechlorination procedure (Table 1). However, at the lower dosage of polymer added in the humic acid solutions, the TOC and CHCl_3 formation were increased. It was noted that the significant reduction of TOC and CHCl_3 with lower residual turbidity (< 1 NTU) were observed, if the cationic polymer was fed at 10 mg/L level in these chlorinated samples. For the post-chlorinated samples, the reduction ratio of CHCl_3 formation could be reached to 82% at 10 mg p-DADMAC/L. This evidence was confirmed with the previous findings in the literature which suggests the p-DADMAC should be regarded as the primary coagulant which could enhance the coagulation

performance via the mechanisms of surface adsorption and charge neutralization. It was thus concluded that the enhanced coagulation technique performed at the higher level of p-DADMAC dose which can not only effectively remove the turbidity but also reduce the precursors and THMs. This observation was very consistent with the recommendation reported by the Surface Water Treatment Rule [20]. The unfavorable effects of p-DADMAC on reduction of TOC (humic acid) and CHCl_3 would be occurred in the cases of lower polymer dose and longer contact time with chlorine. In other words, the optimum dose of p-DADMAC for reducing TOC in the humic acid solution should be introduced at the higher level, i.e. 10 mg p-DADMAC/L.

Table 1. Effects of p-DADMAC on reductions of turbidity, TOC and CHCl_3 for the humic samples in the course of prechlorination and post-chlorination procedures

Coagulation Condition		Quality of treated water					
Run #	p-DADMAC (mg/L)	Residual turbidity (NTU)		TOC (mg/L)		CHCl_3 ($\mu\text{g/L}$)	
		Pre- Cl_2	Post- Cl_2	Pre- Cl_2	Post- Cl_2	Pre- Cl_2	Post- Cl_2
1	0	54.4 \pm 3.1	53.1 \pm 1.4	2.82 \pm 0.05	2.87 \pm 0.06	87.1 \pm 12.1	104.7 \pm 15.8
2	2	41.9 \pm 3.9	41.0 \pm 1.5	3.36 \pm 0.40	3.62 \pm 0.16	104.7 \pm 26.6	104.1 \pm 22.4
3	5	18.7 \pm 1.4	26.6 \pm 1.5	3.42 \pm 0.23	3.77 \pm 0.19	103.3 \pm 22.7	108.0 \pm 9.5
4	10	0.34 \pm 0.04	0.36 \pm 0.07	0.71 \pm 0.22	0.52 \pm 0.07	81.2 \pm 21.1	18.1 \pm 3.1

1. Sample preparations: TOC = 3.00 \pm 0.06 mg/L; Turbidity = 102.0 \pm 1.5 NTU; Cl_2 dose = 11.6 mg/L

2. Data presentation: mean \pm S.D. (replicated experiments n= 5)

Table 2 exhibits the effects of p-DADMAC on the reduction of turbidity, TOC and CHCl_3 for the source water from the Pan-Hsing water treatment plant. The optimum dose of p-DADMAC for reducing turbidity was observed to be at 2.0 mg/L and resulted in having the lowest concentration of TOC and CHCl_3 in the treated water. This observation could be explained that the restabilization phenomena was occurred to produce the higher residual turbidity at the expense of larger amount of polymer dose (5 and 10 mg/L). Similar results regarding the occurrence of the higher concentrations of TOC and CHCl_3 in the treated water were also observed in Table 2.

Table 2. Effects of p-DADMAC on reductions of turbidity, TOC and CHCl_3 for the source water samples in the course of prechlorination and post-chlorination procedures

Coagulation Condition		Quality of treated water					
Run #	p-DADMAC (mg/L)	Residual turbidity (NTU)		TOC (mg/L)		CHCl_3 ($\mu\text{g/L}$)	
		Pre- Cl_2	Post- Cl_2	Pre- Cl_2	Post- Cl_2	Pre- Cl_2	Post- Cl_2
1	0	46.8 \pm 1.7	40.0 \pm 1.6	2.97 \pm 0.25	3.48 \pm 0.74	101.6 \pm 11.5	92.6 \pm 9.8
2	2	1.60 \pm 0.20	0.78 \pm 0.18	1.83 \pm 0.23	1.48 \pm 0.06	85.3 \pm 5.5	26.3 \pm 5.4
3	5	9.70 \pm 0.37	9.6 \pm 0.6	2.56 \pm 0.06	2.47 \pm 0.09	94.3 \pm 2.1	65.5 \pm 0.7
4	10	20.8 \pm 0.1	20.1 \pm 0.0	5.03 \pm 0.71	5.06 \pm 0.35	136.6 \pm 1.0	95.2 \pm 2.7

1. Sample preparations: TOC = 3.39 \pm 0.06 mg/L; Turbidity = 131.0 \pm 2.8 NTU; Cl_2 dose = 11.6 mg/L

2. Data presentation: mean \pm S.D. (replicated experiments n= 3)

Figures 8, 9 and 10 show the effects of p-DADMAC doses on residual turbidity, TOC, and CHCl_3 , respectively, for the humic acid and source water samples in the course of prechlorination and postchlorination. It was clearly demonstrated that there were no significant difference in turbidity and TOC removal for humic acid and source water samples in the course of prechlorination and postchlorination. By observations of Figures 8, 9 and 10, it had a great difference in CHCl_3 reduction in comparison with the prechlorination and postchlorination procedures. For example, the TOC concentration in the source water can be reduced to 10-20% during the prechlorination and postchlorination processes. The relatively higher concentration of CHCl_3 was observed in the prechlorination procedure due to its longer contact time as compared with the postchlorination procedure. As a result, the concentrations of CHCl_3 were found to be 26 $\mu\text{g/L}$ and 85 $\mu\text{g/L}$ in the course of post-chlorination and prechlorination procedures, respectively, when the p-DADMAC was fed at 2 mg/L.

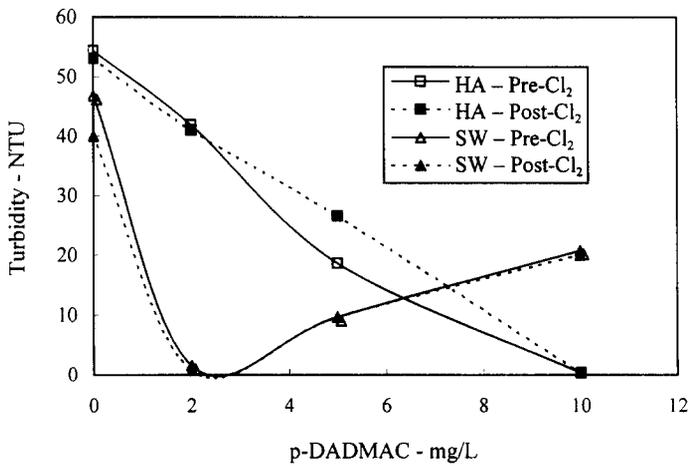


Figure 8. Effects of p-DADMAC dose on residual turbidity for humic acid (HA) and source water (SW) samples in the course of prechlorination and postchlorination

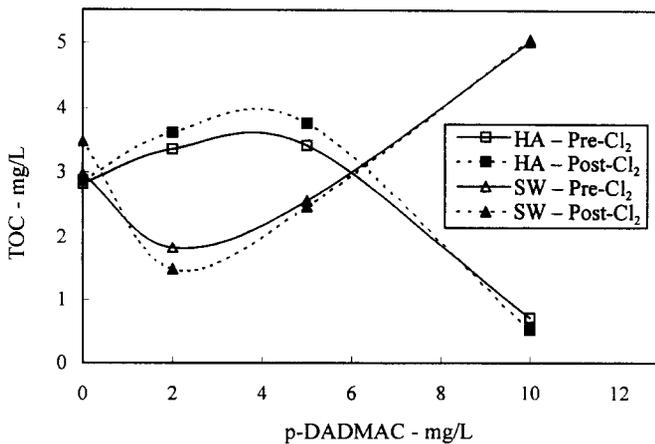


Figure 9. Effects of p-DADMAC dose on residual TOC for humic acid (HA) and source water (SW) samples in the course of prechlorination and postchlorination

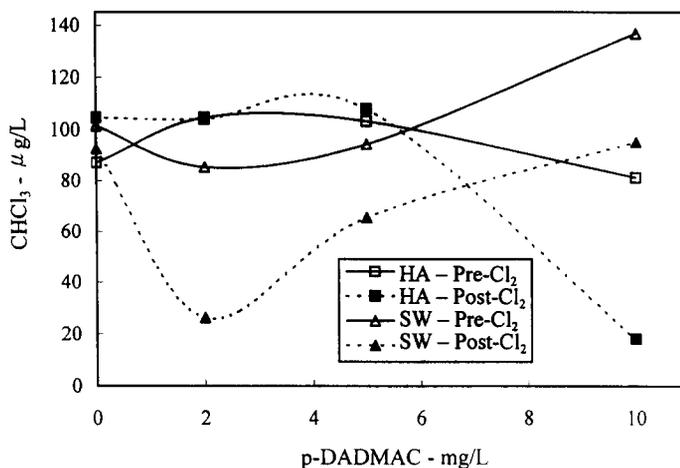


Figure 10. Effects of p-DADMAC dose on CHCl_3 formation for humic acid (HA) and source water (SW) samples in the course of prechlorination and postchlorination

CONCLUSIONS

In this investigation, it was observed that there was a linear correlation (Type I) between Cl_2 demand (<2.8 mg/L) and CHCl_3 formation (<30 $\mu\text{g/L}$) till the chlorine demand approached 3.0 mg/L (Type II). Type I reaction indicates the stoichiometric ratio is held constant between the chlorine and p-DADMAC; while Type II reaction shows the higher CHCl_3 formation potency because the successive reactions among the react two THMs species, i.e., chloroform and bromodichloromethane occurred after the chlorination of p-DADMAC. The chlorine demand was found to be significantly reduced if the H_2SO_4 electrolyte was fed to the sample solutions. This evident was confirmed by the general principle, i.e., the more acidic solution, the less THMs formation. The amount of CHCl_3 formation was also reduced when the Na_2SO_4 electrolyte was introduced in spite of the levels of light intensity. In general, the effect of light intensity on Cl_2 -demand was greater than that of the Na_2SO_4 electrolyte, but lower than that of the H_2SO_4 electrolyte at the higher dose.

The cationic p-DADMAC could be considered as an enhanced coagulant which could not only effectively remove the turbidity but also reduce the formation of CHCl_3 . The removal of turbidity was very consistent with reductions of TOC and THMs in these selected samples. The higher reductions of TOC and CHCl_3 with lower residual turbidity (<1 NTU) in the treated water were achieved by introducing the p-DADMAC in the coagulation process. These observations indicate the potency for reduction of turbidity in the source water could be regarded as a performance evaluation indicator in the coagulation process. In other words, this accomplishment could provide an alternative to the plant manager for selecting the p-DADMAC as an appropriate chemical coagulant to meet the requirement suggested by the enhanced coagulation technique. The optimum dosage of p-DADMAC for reducing the turbidity, TOC and CHCl_3 in the humic acid and source water samples was determined and dependent upon the matrix or molecular weight of organics. In general, the

larger molecular weight of organics (humic acid), the higher the amount of p-DADMAC. There were no significant difference in turbidity and TOC removal for humic acid and source samples, with the exception of CHCl_3 reduction, in the course of prechlorination and postchlorination processes.

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