

POTABLE WATER TREATMENT BY POLYACRYLAMIDE BASE FLOCCULANTS, COUPLED WITH AN INORGANIC COAGULANT

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Abstract : For this study, we polymerized polyacrylamide base flocculants (PAA) and tested their properties and settling efficiency as a treatment for potable water. The most common chemicals for potable water treatment in Korea are alum or PAC. However, due to various reasons (such as rainy season or algae), inorganic flocculants cannot be solely depended on to solve all the problems caused by the poor quality of inflow water. When PAA coupled with coagulants in a potable water purification process is used, the turbidity removal efficiency increases by a factor of three on a single chemical system using PAC (Raw water: 5.21 NTU; Treated PAA+PAC: 0.34 NTU; and, Treated PAC: 1.04 NTU). It is possible to offset the toxic effect of residual monomers in treated water using PAA, because the concentrations of residual acrylamide are less than 400 mg/L in the polymer itself and less than 0.04 µg/L in the treated water base at a dosage of 0.1 mg/L. Therefore, PAAs may be a workable, and dependable, potable water treatment process for the high pollutant level of resource water.

Key Words : Coagulant, Polyacrylamide flocculant, Potable water treatment, Turbidity

INTRODUCTION

For water treatment plants, consistent quality is at a premium, which underscores the importance of timely responses to fluctuations in raw water quality. This is the reason there are many treatment methods for potable water treatment in Korea.

Primary treatments, such as screening, grit removal, and degreasing, can only be justified when surface waters are highly polluted. This type of process is designed to remove large

matter that is likely to hinder subsequent treatment processes. Coagulation generates floc formation from colloidal particles and is an essential element in the success of the solid-liquid separation process.

Solid-liquid separation by filtration, either through beds of sand, or by using activated carbon as an adsorbent, is used after coagulation. Other treatments consist of oxidation to remove excess organic matter and pathogenic germs, disinfection treatment, and removal of metal ions. Coagulation can be adjusted chemically and is a process that is very important for unit operation.

In most cases, inorganic flocculants have been sufficient to maintain the water quality. However,

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inorganic flocculants cannot solve all the problems caused by the poor quality of inflow water.¹⁾

For example, river water as a potable water resource has a high algal activity during each summer season, wherein the pH value increases from 7.5 to 9.0.

Inorganic coagulants are inefficient at high pH, because the ionic characteristics of metal ions are dependent on a pH balance.

Commonly, metal salts as inorganic coagulants, and polyamine and poly diallyl dimethyl ammonium chloride (polyDADMAC) as organic coagulants, have a higher charged density and a lower molecular weight than does polyacrylamide as a flocculant.

Coagulants are of two types: Inorganic, such as aluminum sulfate, PAC, and PACS; and, synthesized organic coagulants, such as polyamine and polyDADMAC. Both types are widely used for treatment of wastewater and drinking water.

Therefore, in many countries polyamine and polyDADMAC, as organic coagulants, are used in conventional coagulation/sedimentation processes instead of metal salts as primary coagulants and destabilizing agents via a charge neutralization and precipitation mechanism, because organic coagulants have little affect on the pH of raw water.²⁾ One of the major uses of organic coagulants in water treatment is as an aid to increase floc size after metal salt has been used as the primary coagulant. After coagulation by a metal salt, as inorganic coagulant, the particles produced may have an overall slightly positive or slightly negative surface charge depending on the dose of metal salt and the type of raw water.³⁾

Only polyamine, as a sub-primary coagulant, is adaptable to potable water treatment in Korea. However, many countries use polyDADMAC for drinking water treatment. According to the AWWA standard for polyDADMAC, it is more useful for potable water treatment when the DADMAC monomer contained in the poly DADMAC product is less than 0.05 mg per milligram. The maximum allowable dosage of polymer in the source water of a water treat-

ment plant is 10 mg.⁴⁾

Hanson et al. report that the maximum intake of DADMAC from drinking water is estimated to be about 700 mg per person per year.⁵⁾

Polyacrylamide base flocculant (PAA), which is used in the flocculation process, is a high molecular weight compound that is soluble in water and has major advantages such as being flexible and effective in treating wastewater, paper, sedimentation, and flotation with a wide range of concentrations due to rapid reaction and low dosage. PAA is also adaptable for drinking water treatment in many countries.

Chemical clarification methods have been used to improve the quality of drinking water supplies since the late 1880s. PAA belongs to a family of synthetic organic polyelectrolytes (also called polymers of flocculants) used in water treatment to improve operation unit performance in the treatment process, most often by increasing the extent or rate of liquid-solid separation.⁶⁾

PAA is superior in performance compared to both inorganic and organic coagulants, which cannot solve the quality problems caused by high concentration of green algae and overflow of activated carbon. And PAA as a polymeric flocculant produces both a lower dosage and a lower volume of sludge than conventional inorganic and organic coagulation, because the pH level of water only slightly influences PAAs effectiveness.

Generally, using PAA in the drinking water treatment process increases the settling rate, reduces costs, improves the finished water quality, and provides better dewatering characteristics for sludge with reduced sludge volume.⁷⁻¹⁰⁾

But this process is not permitted in Korea because of the residual monomer.

Polymer formulations contain contaminants from the manufacturing process, such as residual monomers, other reactants, and reaction by-products that could potentially negatively impact human health. Due to concern over human health, Japan and Switzerland have adopted regulations prohibiting the use of synthetic

organic polymers in drinking water treatment. France, Germany and the United States also have established stringent limits on organic polymer applications.

For this study, PAA, as a polymeric flocculant, were polymerized and their properties were found to be adaptable to the potable water process. And to confirm the application possibility using polyacrylamide flocculants as a drinking water treatment agent, the removal efficiency of turbidity and toxicity as residual acrylamide were estimated in various conditions. The polyacrylamide flocculants, coupled with coagulants, also were studied in the drinking water purification process.

Finally, we estimated the realities of potable water treatment in Korea using PAA on the Nak-dong River comparing the toxicity of residual acrylamide with exposure by various sources.

MATERIALS AND METHODS

Materials

PAC as an inorganic coagulant, polyamine as an organic coagulant, and PAA were used in the experiments. Coagulants were collected from different drinking water treatment plants in Korea. PAC (Poly Aluminum Chloride [$Al_2(OH)_nCl_{6-n}]_m$, 17w% Al_2O_3) used in this study was produced by Gyung-gi Chemicals, Korea. Polyamine (FL-17 PWG) and polyDADMAC (FL-45C), as cationic polymer coagulants, were produced by SNF, France. Polyacrylamide-type polymers were produced by Eyang chemical Co., Ltd., Korea, and were polymerized in the laboratory as an anionic polymer (Aseries; copolymer acrylamide /Acrylic acid, APAA), a cationic polymer (Cseries; copolymer acrylamide/ trimethyl ammonium ethyl acrylate chloride, CPAA), and a nonionic polymer (Nseries; acrylamide homopolymer, NPAA).

The polymerization was carried out in a 1 L reactor equipped with a nitrogen inlet/outlet, digital thermometer.

The monomer and catalyst were prepared and

the initiator injected. Polymerization times varied between 20 and 200 min, and the aging time was 1~6 hr. Molecular weight variations were produced by altering the concentrations of monomer and initiator.

To simulate an oxygen-free reaction environment, a nitrogen stream was bubbled into the monomer solution for 15 min and then a nitrogen blanket was passed over the solution for the duration of the polymerization.

Experimental

All tests were performed with a six cube jar test on a 1 L beaker. The rapid mixing time for coagulant was 5 min at a paddle speed of 200 rpm. The slow mixing for flocculation was 5 min at 50 rpm, and the sedimentation was 5 min. After settling, 50 mL of supernatant was taken on and turbidity and pH were measured immediately.

The turbidity during coagulation and flocculation was followed with the 6-channel turbidimeter which gives the spectrum for the turbidity evolution. The 6-channel turbidimeter, which is supplied by SNF, continuously measures the turbidity.

The dosing solution concentration using deionized water was PAA (0.01w%), PAC (1w%), polyamine, and polyDADMAC (0.1w%).

Measurement

Solution viscosity was measured at $25 \pm 2^\circ C$ in a constant temperature water bath using a Brookfield Viscometer, LVTDV-II. The sample was titrated to analyze ionicity of the polymer with anionic potassium polyvinyl sulfate (PPS) in the presence of blue toluidine as a cationic dye. The end point was determined as point at which the color changed from blue to purple.

The effective turbidity was measured at the end of each test with the DR 2000 Turbidimeter. The concentration of total organic carbon was measured by a TOC analyzer TOC-5000.

To estimate the toxicity for the residual acrylamide, a high-performance liquid chromatography (HPLC) was used with a Waters 2487

dual λ absorbance detector and an auto chro data module. The analysis conditions are as follows:

- Column: Econosil C18 10u (ID.:4.6 mm, length: 250 mm)
- Flow: 1.000/1.000 mL
- Pressure: 740~760 psi
- Collection ratio & time: 10 Hz & 10 minute
- Effluent: 6% Methanol solution using DDW

RESULTS AND DISCUSSION

The Effect of PAC, Polyamine and Poly DADMAC as Coagulants

The objective of water treatment chemicals is to improve the settling rate of the floc in the sedimentation tank to improve the final settling turbidity. To estimate the effect of PAC as a coagulant, using the Nak-dong River as a raw water resource, we first established the dosage for coagulants in various conditions.¹¹⁾

Figure 1 shows the effect of the dosage for PAC on turbidity and TOC removal.

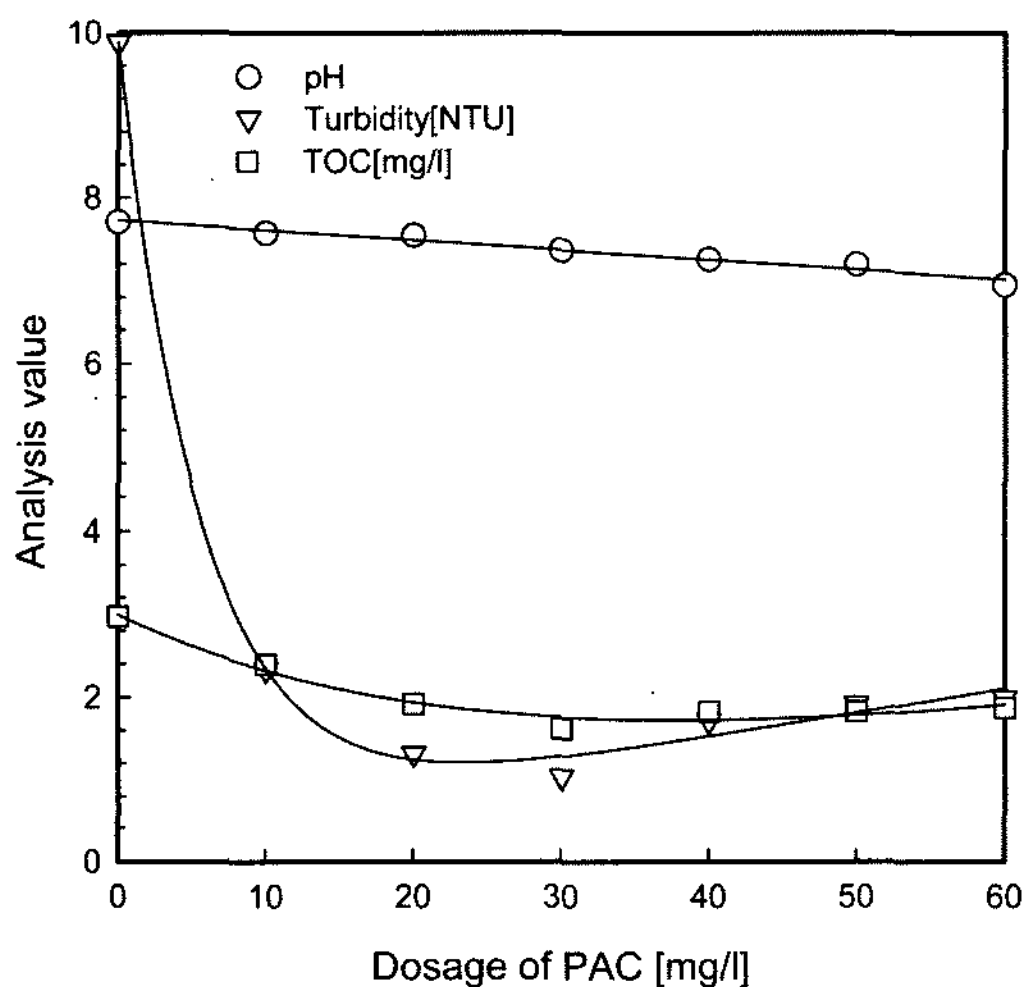


Figure 1. The effect of dosage on PAC on turbidity and TOC removal efficiency.

The optimal dosage of PAC is 30 mg/L in under these conditions. This means that the micro-floc is generated by the optimized attraction and charge neutralization between the negative charged suspended particles and the positive charged PAC particles as a cationic coagulant. This is the general dosage (5~50 mg/L) of PAC for potable water treatment in Korea.

In cases where the dosage of PAC is more than 30 mg/L, the turbidity and TOC removal efficiency is decreased by a reverse effect of the net charge; the surface charge of micro-floc is formatted as a positive or a neutral charge in the optimal dosage, but that is repulsed with free PAC particles because of the same surface charge in an over dosage.

The optimal dosage of both polyamine and polyDADMAC are approximately 3.0 mg/L under these conditions.

The relationship between turbidity and algae growth is very important for the improvement of drinking water quality. The pH of raw water is increased from a pH of 7.4 to a pH of 9.3 by algae growth. Table 1 shows the turbidity removal efficiencies for each coagulant in raw water after algae growth without flocculant. PolyDADMAC has a removal efficiency that is superior to other coagulants on algae growth. FL-45 has a low molecular weight and is the most suitable coagulant. When the molecular weight of polyDADMAC is high, the floc size is large, but the turbidity removal efficiency is poor.

The Effect of pH on Raw Water Coagulants

It is especially difficult to remove suspended solids and dissolved pollutants using an inorganic coagulant during the rainy season. During the rainy season, raw water has high green and blue algal properties with the attendant high pH value. Therefore, at this time of year it is very

Table 1. The turbidity removal efficiencies on various coagulants after algae growth

Items	Raw water	Alum	PAC	FL-17	FL45	FL45VHM
Dosage (mg/L)	-	60	30	3.0	3.0	3.0
Turbidity (NTU)	8.71	3.48	3.76	2.10	1.50	1.65

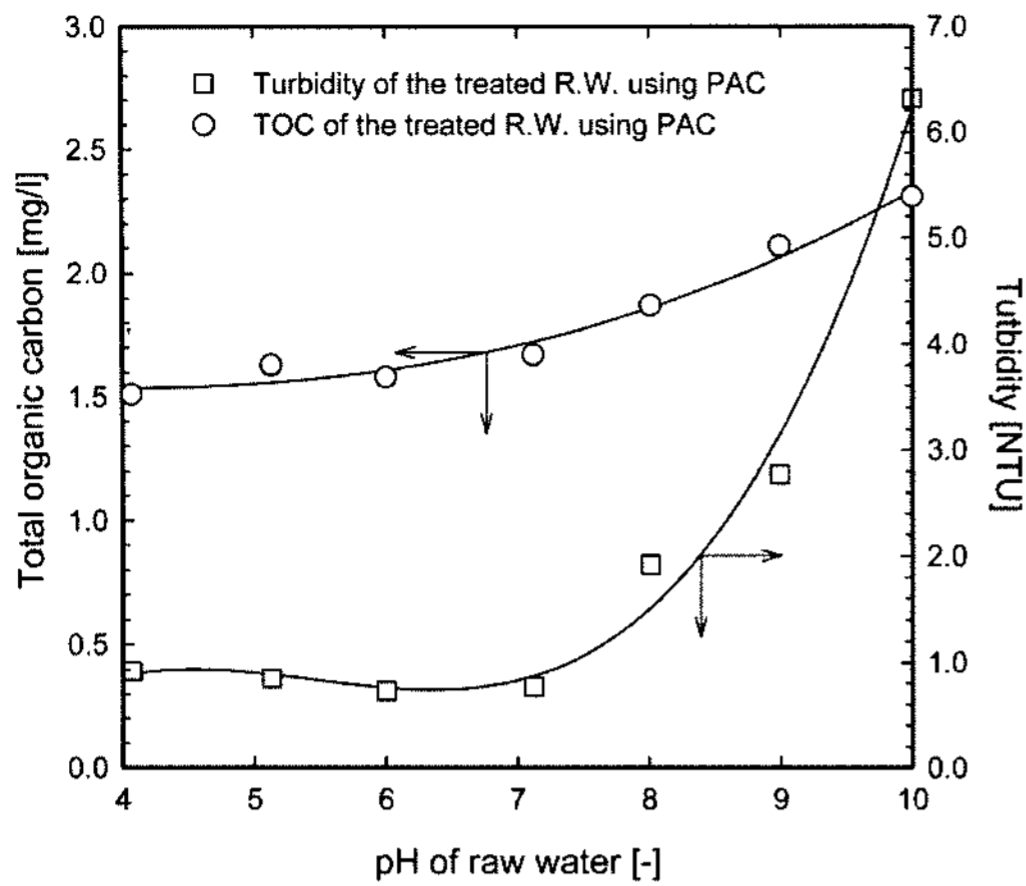


Figure 2. The effect of pH on PAC for raw water on turbidity and TOC removal efficiency.

important that water treatment facilities use a coagulant that works efficiently during abrupt changes in the pH of raw water.

Figure 2 shows the effect that the pH of raw water has on the efficiency of PAC turbidity and TOC removal. We changed the pH of raw water using 0.1 N NaOH and 0.1 N H₂SO₄ at a PAC dosage of 30 mg/L. The efficiency of PAC for turbidity removal varies greatly by pH change. The optimal range is from pH 5 to pH 7 with a removal ratio for TOC and turbidity at 10~40% and 70~90%, respectively.

In the case of PAC, its performance is inversely related to the pH of raw water – when the pH is high, the performance is poor. The removal efficiency of both TOC and turbidity is almost identical.

This means that the charged state and ionic interaction between aluminum ions and pollutants are dependent on pH and that the states influence efficiency. But for organic coagulants polyamine and polyDADMAC, both the state and the efficiency remains the same. Polyamine and polyDADMAC have no metal ions but do possess a functional group that is a cationic source. The performance efficiency is unchanged, because pH change has little effect on the charge of organic material.

The Effect of a Combination of Inorganic and Organic Coagulants

Polyamine and polyDADMAC are used as primary coagulant alternatives in wastewater treatment. They have similar high ionic characteristics compared to PAC. But polyamine and polyDADMAC, with a molecular weight in the range of from 100,000 to 1,000,000 Dalton, both have higher molecular weights than PAC. The coagulation mechanism between coagulants and pollutants is the same.

Therefore, the balance of inorganic to organic coagulant is important. Figure 3 shows the interrelationship between PAC and polyamine on turbidity and TOC removal efficiency. The optimal dosage of polyamine for each condition is 1.5~2.0 mg/L (PAC 10 mg/L), 0.6~0.7 mg/L (PAC 20 mg/L), and 1.0~2.0 mg/L (PAC 30 mg/L). The optimal condition is PAC 20 mg/L and polyamine 0.7 mg/L, but that is critical.

Generally, the properties of raw water resources vary. Therefore, a reasonable condition is PAC 30 mg/L and polyamine 1.0~2.0 mg/L for stable performance under varying conditions. Polyamine reacts slower than PAC, because the molecular weight is higher than PAC and it has a sub-reaction (Branching between the flocs).

At a high dosage of PAC (30 mg/L) and polyamine (1.0 mg/L), performance is poor due

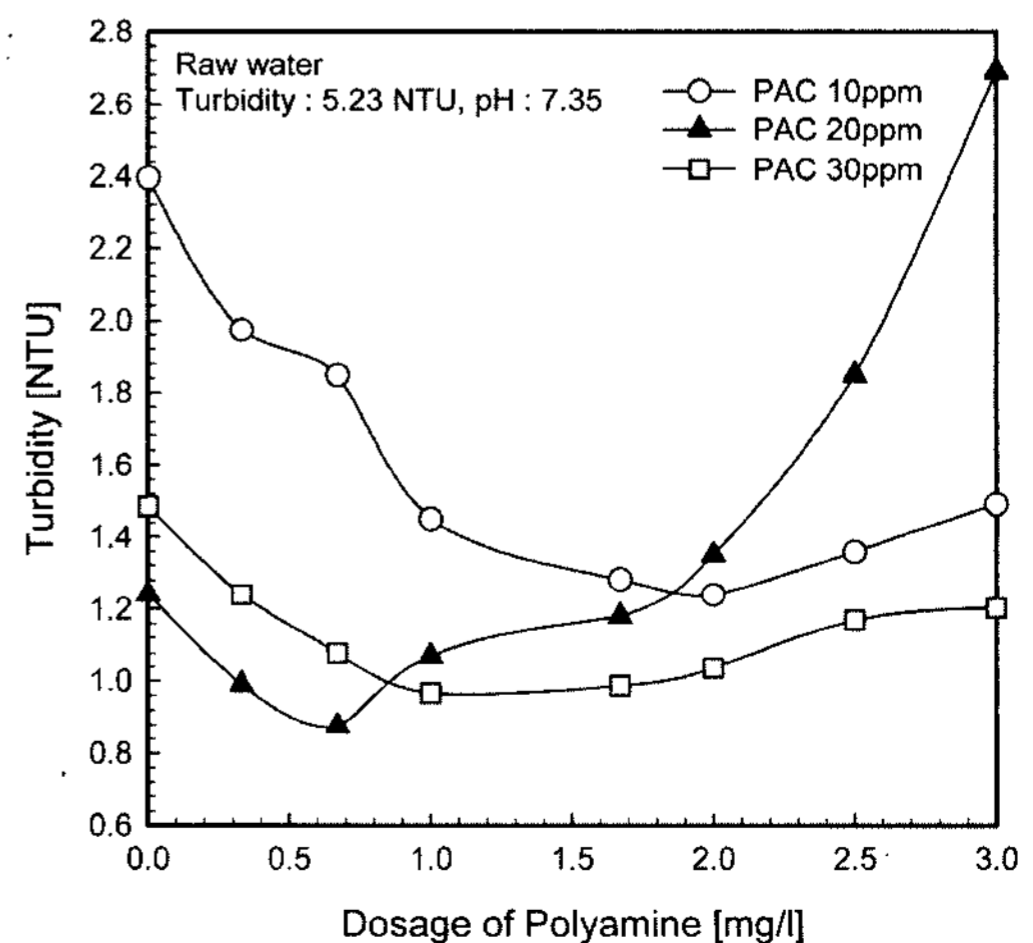


Figure 3. The relationship between PAC and polyamine on turbidity and TOC removal efficiency.

to a high charge density. Therefore, the net charge is reversed.

The Effect of Polyacrylamide Flocculant

Polyacrylamide is used for sedimentation, flotation and dewatering to improve efficiency in wastewater treatment.

In the treatment of potable water, cationic organic coagulants can be used alone as a primary coagulant or together with an inorganic coagulant to help with coagulation efficiency. Also, it is possible to use a polyacrylamide flocculant to accelerate the settling speed. An inorganic and organic coagulant combination is used for potable water treatment in Korea. Polyacrylamides are not permitted by regulation license. However, there are many countries in which polyacrylamide-type flocculant is used to improve the efficiency of potable water treatment.

The addition of a flocculant after coagulation may be necessary, depending on the settling time used at the treatment plant. The shorter the settling time, the more important it is to use a flocculant. In many cases, addition of a flocculant can notably increase the input flow without comprising quality.

Figure 4 shows the effect of ionic charge density of PAA for turbidity and TOC removal

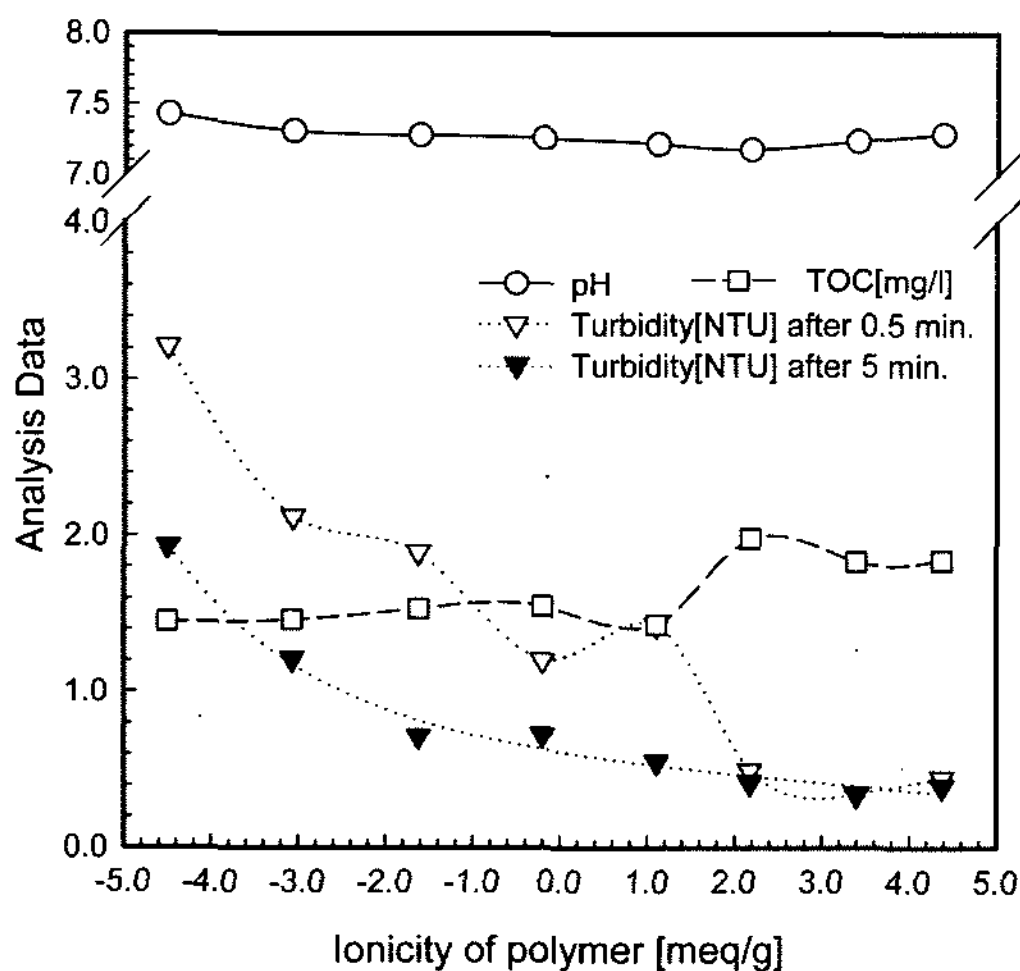


Figure 4. The effect of ionic charge density of PAA for turbidity and TOC removal efficiency with PAC.

efficiency with PAC coagulation. For this study, the dosages of PAC and PAA were 30 mg/L and 0.1 mg/L, respectively. The quantity of PAA was large enough to agglomerate the flocs that were destabilized during the coagulation step with PAC, and it was large enough to avoid a sedimentation rate that was too fast.

Therefore, cationic flocculant (CPAA) generated a large floc with sludge due to the bridging effect from a free electric attraction. PAA had more than 2.0 meq/g for its charge density and had a superior turbidity removal efficiency rate at 0.5-minutes and 5-minutes (Figure 4). The optimal ionicity for removal of TOC and turbidity ranged from 0.0 to 1.5 meq/g for CPAA in this study.

This means that the conditioning of raw water by coagulants has a partial negative charge, so the cationic flocculant is useful due to the random free cationic functional group it generates, and the dosage of PAM is universal at around 0.1 mg/L. The dosage of flocculant to be added is very small—in the range of 0.01 mg/L to 0.5 mg/L—in many advanced countries to prevent the risk of a residual acrylamide monomer.

The chemical treatment of potable water for this study can be divided into two steps by dual chemicals systems. The first step is coagulation, which consists of the destabilization of colloids and the formation of flocs by coagulants, and the second step is flocculated sedimentation as a solid-liquid separation stage by PAA.

It is important that the process using a flocculator involve significant mechanical agitation in order to increase the collision of reagents - coagulant and flocculant - and the colloidal particles in suspension. However, the cutting action must not be too strong, otherwise it will destroy the flocs that are formed.

The effect of PAC dosage in a dual system is illustrated in Figure 5. PC-218 and FO4240PWG are medium molecular weight (approximately 9 million Dalton) and cationic PAAs. The turbidity removal efficiency is poor at low PAC dosage for both PC-218 and FO4240PWG. These results illustrate that when the PAA charge is optimal

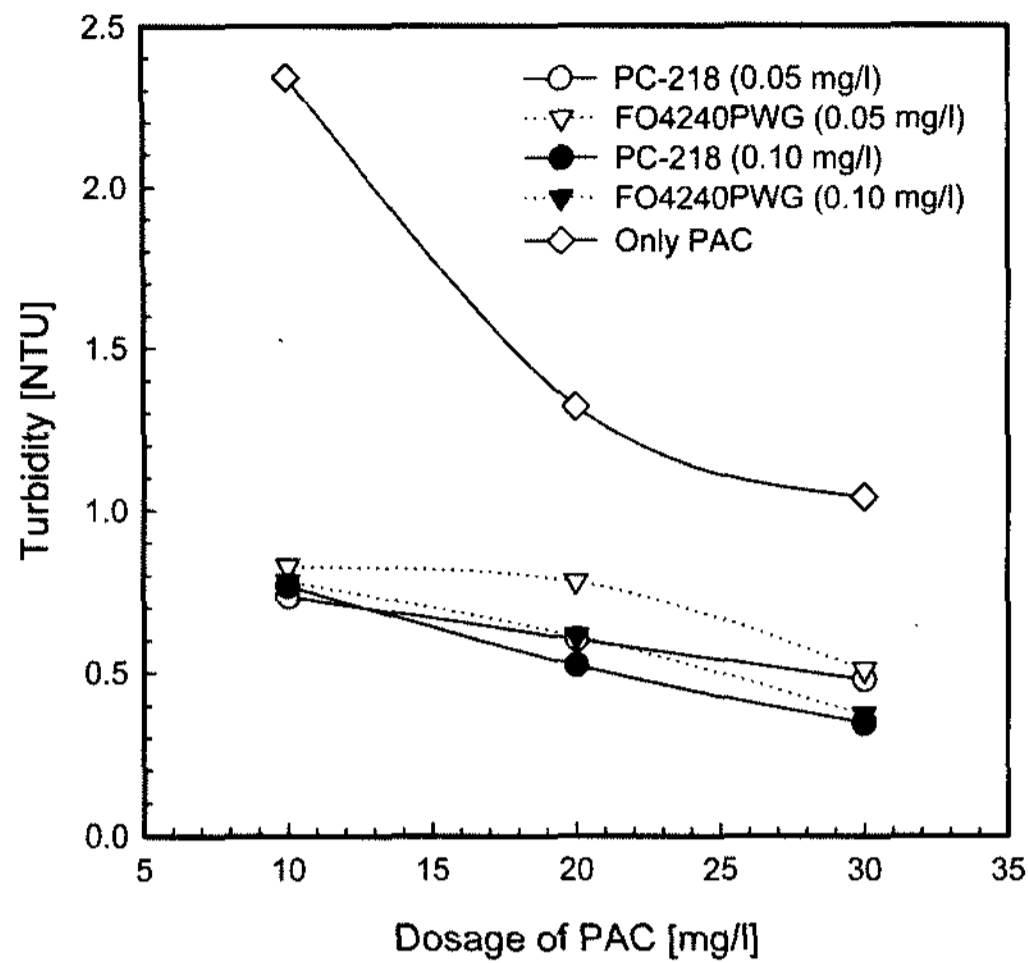


Figure 5. The effect of combination between PAA and PAC on turbidity and TOC against PAC.

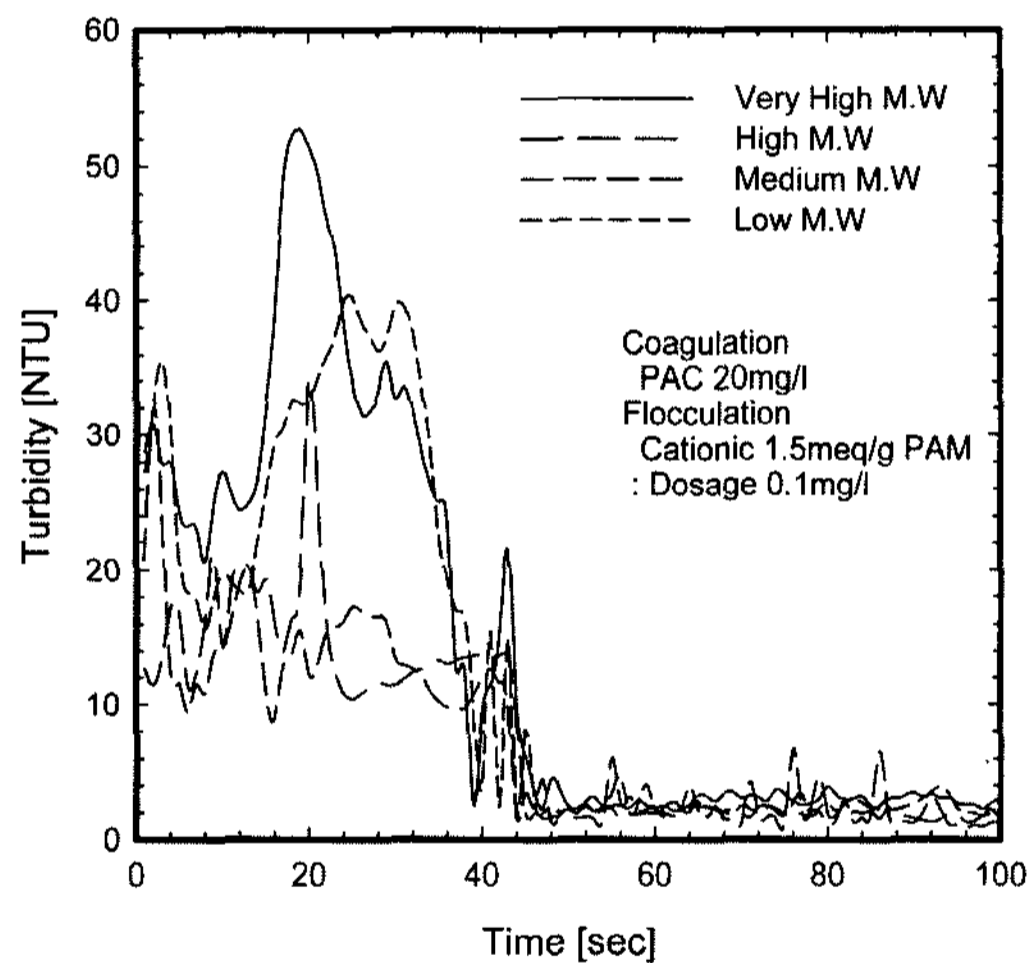


Figure 6. The turbidity spectrum for inorganic coagulants with polyacrylamide using 6 channel turbidimeter.

in a dual chemicals system, charge neutralization is accomplished at a much smaller dosage of inorganic coagulant than in a single coagulation system.

Figure 6 illustrates the turbidity spectrum for PAC as inorganic coagulants with polyacrylamide. In general, PAC was used as an inorganic coagulant to form the micro floc. But the varying molecular weight of PAA could be considered an enhanced flocculant for generation and branching of micro floc. The higher the molecular weight of PAA, the higher the

turbidity during flocculation, due to larger floc. The difference in turbidity means the size of the generated floc is interfering with absorption. The flocculation time of PAA is about 40 sec, which is much faster than both inorganic and organic coagulants.

The molecular weight of PAAs as million Dalton is followed; low M.W. (4) less than 5, medium M.W. (9) from 5 to 10, high M.W. (14) from 10 to 20 and very high M.W (25) higher than 20.

The removal of turbidity was consistent with reductions of TOC. Therefore, the higher reductions of TOC, with attendant lower residual turbidity, in the treated water were achieved by introducing the dual system of PAA and PAC into the potable water treatment process.

The results of the polyacrylamide base flocculant could provide options for water treatment plant operators seeking appropriate chemicals to replicate the results suggested by the enhanced drinking water treatment technique.

The optimal effect of PAA is mainly determined by the surface potential acting on the particle. These results are dependent both on the particle itself and on the conditions in the environment, i.e., on the ionic strength of the water and the resulting properties, such as pH value, electrical conductivity, and hardness.

Natural pollutants in raw water generally contain large quantities of negatively charged colloidal matter. Therefore, it is possible to change the optimal conditions, including the dosage, kind, and structure of chemicals inherent in the natural properties of raw water. But, inorganic and organic coagulants have an even higher cationic charge and a lower molecular weight than PAA.

Because many kinds of PAA with varying ionic charges (from anionic 100 mol% to cationic 100 mol%), molecular weights, and structures are produced by commercial manufactures, PAA is useful for optimizing the various treatment conditions for drinking water.

The Residual Monomer of Polyacrylamide Flocculant

Residual acrylamide monomer was extracted from the PAA into a mixture of water and acetone, which softened the polymer but did not dissolve it. The extract was analyzed by high-performance liquid chromatography (HPLC) using ultraviolet detection. Identification was made by comparison with an external standard and concentration determined by peak area measurement and ratio.¹⁰⁾

The detection times of residual monomers in PAA [acrylic acid (3.448 min), acrylamide (5.242 min) and cationic monomer (> 5.5 min)] were dependent on the kind of monomer in that experimental condition. The concentration of external acrylamide standard solution was 100 mg/L and the analysis results were 9018.886 (mV×sec) as a function of concentration by analysis area.

The analysis results of commercial PAA and lab products are illustrated in Table 2.

The concentration of residual acrylamide in treated water is very low and can be ignored when compared with other sources (Regular: Max. conc. in polymer: 250 mg/L, Max. Dosage: 0.2 mg/L, that is 0.05 ppb in water by EPA calculation methods). More than 0.2 mg/L of PAA is a bit of an overdose. And, if the quantity of flocculant is too large, the flocs will

be too large to settle quickly and will not catch all the small colloids.

From the analysis results, human exposure to acrylamide through drinking-water is less than 0.5 µg/day, and human exposure to acrylamide in the diet is estimated to be about 70 µg/day on average by WHO. These levels are low when compared to the No Observed Adverse Effect Level (NOAEL) in animal studies of 500 µg/kg/day.

Therefore, it is possible to ignore the toxic effects on the human body of residual monomers in PAA.

CONCLUSION

To optimize and improve the performance efficiency of chemical treatment for potable water processes, we polymerized the polyacrylamide base flocculant as acrylamide-co-acrylic acid polymer, acrylamide-co-trimethyl ammonium ethyl acrylate chloride polymer and acrylamide homopolymer. This led to the following conclusions:

The turbidity removal efficiency rate of dual chemicals system using PAC and PAA as a post-treatment is more than 93% at 20~30 mg/L of PAC and 0.05 mg/l of PAA (Raw water: 5.21 NTU, Treated-PAA+PAC: 0.34 NTU, Treated-PAC: 1.04 NTU).

Table 2. The analysis results for residual acrylamide in PAA

Sample name	Ionic charge	Manuf.	Retention Time (sec)	Area (mV×sec)	Conc. (mg/L)
PA-211	Anionic 20 mol%	Lab.	5.113	2178.275	241.52
FA-934PWG	Anionic 20 mol%	EYANG	5.211	1134.324	125.74
PC-218	Cationic 20 mol%	Lab	5.200	3211.549	356.09
FO-4240PWG	Cationic 20 mol%	EYANG	5.198	1421.111	157.57

Table 3. Exposure source of acrylamide

Exposure source	Basis	Type	Uptake	Daily exposure
Food	1.4 µg/kg/day	Average (WHO)	100%	70 µg
Cigarette smoking	2 µg/cigarette	20 cigarettes/day	50%	20 µg
Drinking-water (USA)	0.5 µg/L	2 liters/day	100%	1.0 µg
Drinking-water (EU)	0.1 µg/L	2 liters/day	100%	0.2 µg

The floc size and the turbidity of treated water can be controlled by M.W. and ionicity of PAA to improve the filtration efficiency. A slow reaction uses a lower than medium cationic charge: 2.0 meq/g, and high M.W. PAA. A fast reaction is higher and has both a low M.W. and PAA.

The PAA and organic coagulant are not affected by the pH change to flocculate in suspended solids, but PAC is, and the reactivity of PAA is much faster. Therefore, prior to doing a jar test, a plant operator should first calculate the delay times between each chemical addition point and check the reaction time to optimize a dual chemical system.

With the changing properties for every treatment plant, including pH, turbidity, and amount of algae, conductivity also changes. Organic polymers, therefore, should be augmented with inorganic coagulants.

The concentration of residual acrylamide in treated water is very low and can be ignored, especially when compared with that from other sources. More than 0.2 mg/L of PAA is a bit of an overdose. And, if the quantity of flocculant is too big, the flocs will be too big (fast settling rate) to catch all the small colloids. In the filtration process, the lower the turbidity and the smaller the floc, the longer the filtration time. Therefore, the control of turbidity and floc size for treated water is important. We should manage the dosage of PAA in order to get a good settling rate and to keep consistent performance in the sand filter step.

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