Water Coagulation Using Electrostatic Patch Coagulation (EPC) Mechanism

Ya-Ling Cheng [a], Ruei-Jyun Wong [a], Justin Chun-Te Lin [a b], Chihpin Huang [b], Duu-Jong Lee [a] & Arun S. Mujumdar [c]

[a] Department of Chemical Engineering, National Taiwan University, Taipei, Taiwan

[b] Institute of Environmental Engineering, National Chiao Tung University, Hsinchu, Taiwan

[c] Department of Mechanical Engineering, National University of Singapore, Singapore
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Ya-Ling Cheng,[1] Ruei-Jyun Wong,[1] Justin Chun-Te Lin,[1,2] Chihpin Huang,[2] Duu-Jong Lee,[1] and Arun S. Mujumdar[3]

[1]*Department of Chemical Engineering, National Taiwan University, Taipei, Taiwan*
[2]*Institute of Environmental Engineering, National Chiao Tung University, Hsinchu, Taiwan*
[3]*Department of Mechanical Engineering, National University of Singapore, Singapore*

Coagulation of fine particles to form large flocs is an essential step to reach efficient dewatering of wet materials. The electrostatic patch coagulation (EPC) mechanism was applied in this study to remove turbidity and natural organic matters in water using four different dosing-mixing schemes. Sufficient rapid mixing and slow mixing or applying two-stage coagulation benefit removal of turbidity. The EPC mechanism could only efficiently remove organic matters of molecular weight of 7500–10,000 Da. Only sufficient rapid mixing is relevant to organic matter removal. The mechanisms for natural organic matters (NOM) removal are proposed to be complexation of ions and NOM to form insoluble aggregates to be separated by membrane filtration.

INTRODUCTION

Moisture content is a critical parameter governing the feasibility of various chemical and mineral processes. Prior to thermal drying, most moisture in wet materials is removed via mechanical means for the sake of energy savings.[1–14] An enhanced coagulation process was commissioned to remove natural organic matters (NOM),[15] which are precursors of disinfection by-products (DBP) and other synthetic organic compounds in the water.[16] The enhanced coagulation process optimizes coagulant dosage and pH values to achieve total organic carbon (TOC) removal targets. Mechanisms corresponding to the coagulation removal of organic substances had been discussed.[17–19]

Colloidal destabilization can be achieved by adding aluminum or ferrous salts to form cations to be adsorbed onto the negatively charged surfaces or to form excessive hydroxide precipitation to enmesh fine particles. Chowdhury and Amy[20] proposed that precipitation is the main

mechanism in the coagulation of alumina colloids with alum. Studies on polyaluminum chloride (PACl) focused on the mode of charge neutralization.[21,22] Dentel[23] proposed a combined coagulation model considering precipitation charge neutralization (PCN). Narkis and Rebhun[24] showed that when both mineral particles and dissolved humic substances are present in raw water, the latter controls the coagulation process. Coagulation of humic substance-containing suspension would produce highly turbid supernatant.[25,26] Su et al.[27] showed how the levels of mineral particles and of humic acid affect the stability of the blanket in full-scale floc blanket clarifiers.
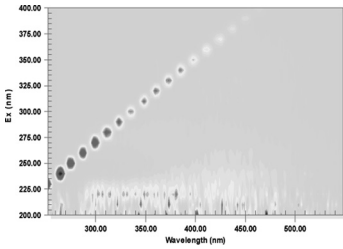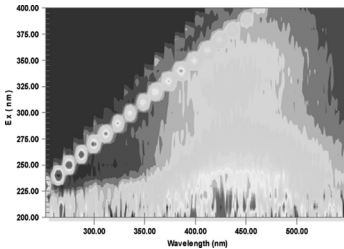
Gregory[28] proposed the electrostatic patch coagulation (EPC) mechanism to interpret his experimental data on efficient turbidity removal before surface charge of particles was neutralized. Restated, when part of the charged surface is neutralized by adsorbing oppositively charged ions, particle coagulation could be induced by local contact between neutralized "patches" of surface could induce coagulation. Wang et al.[29] applied EPC mechanism to interpret the coagulation behaviors of particles using PACl. Duan and Gregory[30] reviewed the mechanisms of coagulation processes.

A high dosage of coagulants leads to corrosion of iron pipe.[31] Excess coagulants in water are potential foulants to membranes if membrane filtration is the step following the coagulation step. Applying the EPC mechanism, if feasible, can save excess coagulant dosage and produce less sludge. This study aims at studying the coagulation behavior of PACl on removal of constituents of NOM of different molecular weights. Four dosing–mixing schemes were tested. The organic matters in the raw waters were fractionated by a high-performance size exclusion chromatography (HP-SEC) system and were characterized using the excitation–emission matrix (EEM) fluorescence spectra. Two-stage alum coagulation for enhancing turbidity removal was studied.[32] This study compared the coagulation–settling performance for NOM removal using one-stage or two-stage coagulation.

Correspondence: Duu-Jong Lee, Department of Chemical Engineering, National Taiwan University, Room 205, No. 1, Sec. 4, Roosevelt Rd., Taipei 10617, Taiwan; E-mail: djlee@ntu.edu.tw

TABLE 1
Characteristics of studied water samples

Sample	Raw water A	Raw water B
Sampling date	March 26, 2008	April 2, 2008
Water temperature (°C)	25	20
pH	7.49	7.29
Turbidity (NTU)	4.28	19.9
DOC (mgL^{-1} as C)	1.62	0.73
Alkalinity (mgL^{-1} as CaCO$_3$)	32	30
EEM plot		

MATERIALS AND METHODS
Raw Water Characteristics

Two types of raw water, namely, water A and B, were sampled from the intake port of a major water treatment plant in ChangShin Waterworks (Table 1). The turbidity and dissolved organic matter (DOM) of the two raw waters were 4.28 and 19.9 NTU and 1.62 and 0.73 mgL^{-1} as C, respectively. The EEM fluorescence spectra of the two raw water samples are also shown in Table 1.

Experimental Setup and Procedures

Jar tests were performed in 2-L square tanks equipped with a six-paddle stirrer. Prior to jar tests, the raw water samples were adjusted to pH 7 using sulphuric acid and sodium hydroxide. The water sample was then placed in the jar tester to evaluate the coagulation efficiencies with

dosed PACl under the scheme listed in Table 2. In processes 1–3 the coagulant was added at the start of the rapid mixing, whereas in process 4 the coagulant was added at half of the dosed quantity at the start of the first stage of rapid mixing and the other half at the second stage of rapid mixing. Process 1 was a typical coagulation process adopted in waterworks that comprised a rapid mixing step at 100 rpm for 3 min followed by a slow mixing at 25 rpm for 20 min. The suspension was kept still for 30 min before sampling. Process 2 consisted of a rapid mixing stage of 180 rpm for 3 min followed directly by 30-min settling. Process 3 had a rapid mixing step at 180 rpm for 3 min plus slow mixing at 25 rpm for 20 min plus 30-min settling. Process 4 comprised two rapid mixing steps, each at 100 rpm for 3 min, followed by slow mixing (25 rpm, 20 min) plus 30-min settling. The suspension at two-thirds height of the tanks was sampled after 30-min settling. The turbidities

TABLE 2
Coagulation scheme for the four coagulants

Coagulation process	Coagulant adding		Way of mixing				
			Rapid			Slow 25 rpm 20 min	Setting time 30 min
	Single-stage	Two-stage	180 rpm 3 min	100 rpm 3 min	100 rpm 3 min		
0							•
1	•				•	•	•
2	•		•				•
3	•		•			•	•
4		•		•	•	•	•

of the collected samples were measured by a turbidimeter (2100P, Hach Company, Loveland, Co., USA). The NOM in the collected samples were fractionated using high-performance size exclusion chromatography (HP-SEC).

Analysis

Zeta potential measurement of particles in suspensions was carried out by a Zetasizer (Nano-ZS, Malvern Co., Worcestershire, UK). The cell repeated flushing by deionized water and ethanol before measurement. The pH of the samples was measured using a calibrated pH meter (WTW pH-315, Weilheim, Germany). The TOC data of water samples were analyzed by Aurora Model 1030 TOC analyzer (OI Analytical, Co., College Station, TX). The nonpurgable dissolved organic carbon (NPDOC) is an index of organic composition concentration. The DOC data were averaged over triplicate analysis.

The HP-SEC system was comprised of a degasser (DEGASYS DG-1310, Uniflows Co. Ltd., Tokyo, Japan), a feed pump (BETA 10 Gradient pump, Ecom spol. s.r.o., Prague, Czech Republic), a chromatography column (TSK G2000SWx1, TOSOH Co., Tokyo, Japan) and two on-line detectors. The first detector is a UV-vis variable wavelength detector (SAPPHIRE 600, Ecom spol. s.r.o.) and the second is a refractive index detector (IOTA 2, Precision Instruments, Marseille, France). Both detectors were connected in series and monitored simultaneously via the software Peak-ABC on the computer. Due to mobile phase affecting the resolution of HP-SEC system significantly; priority concern of solvent is that it will not interfere with analytical samples. A phosphate mobile phase ($0.0024 \, mol \, L^{-1}$ NaH_2PO_4 and $0.0016 \, mol \, L^{-1}$ Na_2HPO_4, pH 6.8) was chosen for low UV absorbance and low interference with samples and freshly prepared for each experiment. Flow rate of the feed pump was set as $1 \, mL \cdot min^{-1}$. The mobile phase and samples were conditioned by pH 7.0 ± 0.1 and filtered by 0.45-μm polytetrafluoroethylene (PTFE) membranes prior to all HP-SEC measurements. A 200-μL sample was manually injected into the sample injector of the feed pump. Polyethylene glycols (PEG, 200, 1000, 4000, 8000, and 20,000 Da) were used for apparent molecular weight (AMW) calibration of chromatograms. The absorption wavelength of the UV detector was set at 254 nm.

The EEM analysis was measured by a Cary Eclipse fluorescence spectrophotometer (Varian Inc., Palo Alto, CA). Because fluorescence measurements are easily affected by instruments, sample preparation methods, sample pH, concentration, temperature, ionic strength, and solvent, it is necessary to control samples in the same condition prior to all measurements. All samples were conditioned by pH 7.0 ± 0.1 and filtered by 0.45-μm PTFE membranes prior to all EEM measurements. EEM spectra were gathered with subsequent scanning emission spectra from 250 to 550 nm at 1-nm increments by varying the excitation wavelength from 200 to 400 nm at 10-nm increments. The spectra were recorded at a scan rate of $1200 \, nm \, min^{-1}$, using excitation and emission slit bandwidths of 5 nm. The voltage of the photomultiplier tube (PMT) was set to 800 V for low-level light detection. Data were saved as ASCII format and removed Rayleigh scattering by Perl code editing. The EEM fluorescence spectra listed in Table 1 revealed that the raw water B contains more fulvic acid–like DOM than the raw water A.

RESULTS

Coagulation of Raw Waters Using Process 1

Process 1 (100 rpm rapid mixing for 3 min plus 25 rpm slow mixing for 20 min plus settling) was conducted to determine the dosage quantity needed in the coagulation
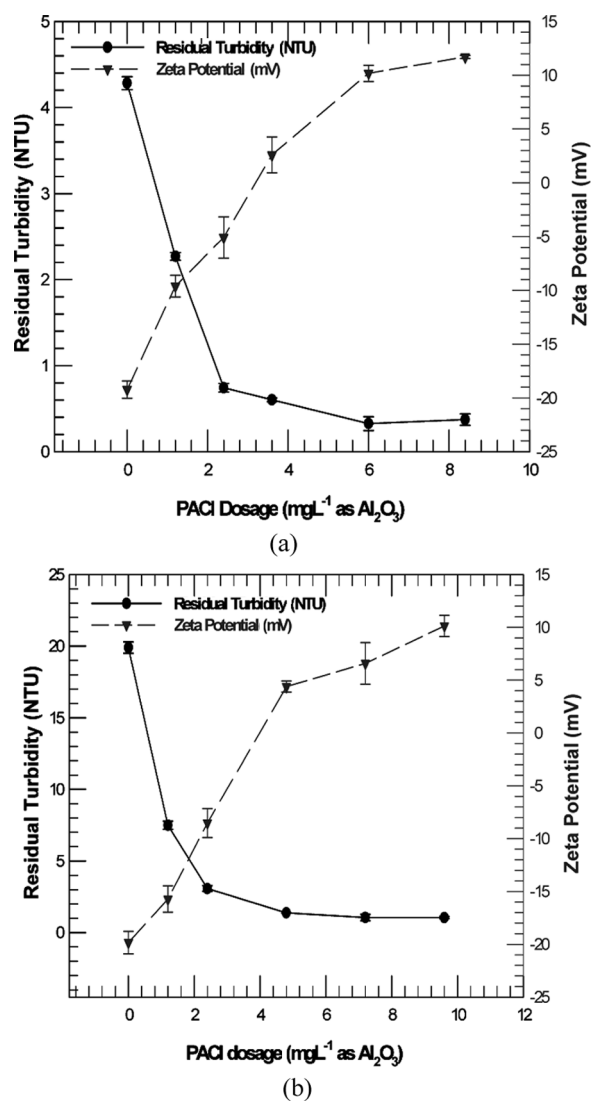

(a)

(b)

FIG. 1.   Residual turbidities and zeta potentials of raw and coagulated water samples. PACl. (a) Water A; (b) water B.

tests. Figures 1a and 1b show the residual turbidities and zeta potentials for the PACl-coagulated waters A and B, respectively. For water A, PACl at $3.6\,mgL^{-1}$ as Al_2O_3 was needed to neutralize the surface charge of the suspended particles in water. However, adding PACl of only $1.2\,mgL^{-1}$ as Al_2O_3 reduced the residual turbidity from 4.3 to 2.25 NTU. Further increasing the PACl dose to $2.4\,mgL^{-1}$ reduced further residual turbidity to 0.75 NTU. For water B, PACl at $4.4\,mgL^{-1}$ as Al_2O_3 was needed to neutralize the surface charge of the suspended particles in water. However, adding merely $1.2\,mgL^{-1}$ as Al_2O_3 reduced the residual turbidity from 20 to 7.5 NTU. Further increasing the PACl dose to $2.4\,mgL^{-1}$ reduced residual turbidity to 3.0 NTU. We denoted the coagulation tests with PACl doses of 1.2, 3.6, and $8.4\,mgL^{-1}$ as ECP, CN, and EM points studied for water sample A. The corresponding dosages for water B were 1.2, 4.4, and $9.6\,mgL^{-1}$ for ECP, CN, and EM points.

Restated, the ECP mechanisms correspond to the PACl coagulation process for turbidity removal in waters A and B. Charge neutralization was not an adequate mechanism in charge of the turbidity removal from low-alkalinity, low- to medium-turbidity raw waters.

There exist six UV_{254} absorption peaks for the SEC diagram for waters A and B (Figs. 2a and 2b), with apparent molecular weights at 8600 (peak 1), 7200 (peak 2), 5600 (peak 3), 3700 (peak 4), 2200 (peak 5), and 1800 Da (peak 6). The sharp peak at around 1500 Da is the water peak containing most dissolved ions.

In both waters, addition of $1.2\,mgL^{-1}$ PACl nearly completely removed peak 1 but only partially removed peak 2. With PACl dose of $2.4\,mgL^{-1}$, peak 2 was effectively removed. Then the intensity of peak 3 was gradually removed with PACl dose further increasing up to $8.4\,mgL^{-1}$. The PACl coagulation had negligible effects on peak 4. On peak 5, the peak intensity increased rather
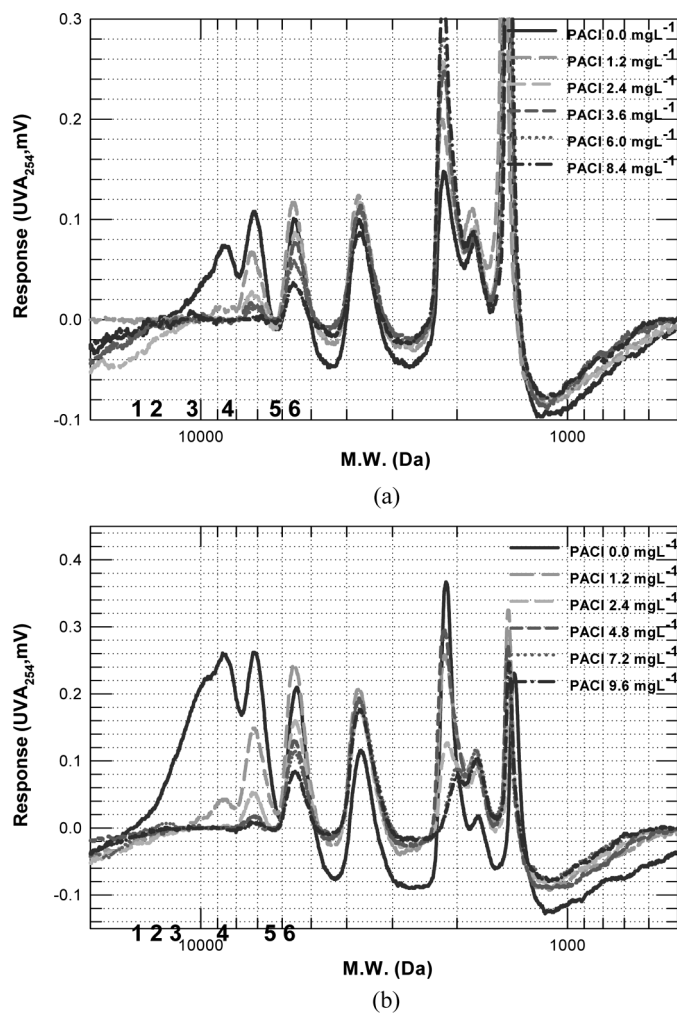

FIG. 2. SEC-UV_{254} spectra for raw and coagulated water samples. PACl. Process 1. (a) Water A; (b) water B.
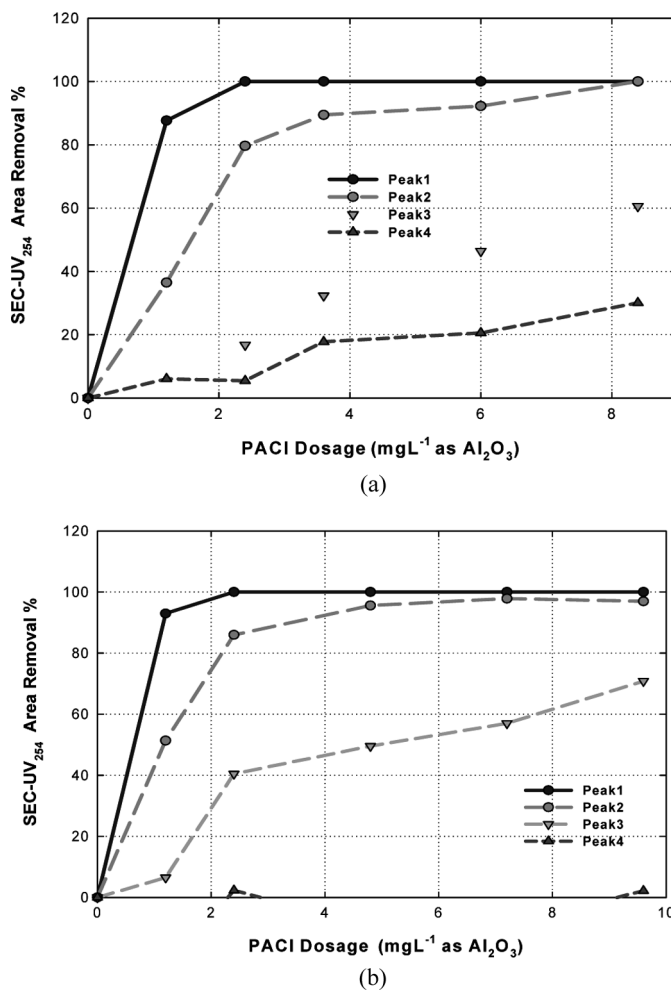
FIG. 3. SEC-UV_{254} areal removal versus PACl dosage curves for peaks 1–4. Process 1. (a) Water A; (b) water B.

than decreased with increasing PACl dose. Restated, the impurity in the dosed PACl contributed to the 2100 Da peak in the tested samples. The peak intensity of peak 6 was comparatively low and the removal efficiency by PACl coagulation was not conclusive.

The areas under UV_{254} (in mV) versus AMW (in Da) curves were numerically evaluated by Simpson's algorithm (Fig. 3). The PACl dosage at $> 2\,mgL^{-1}$ completely removed peak 1. With $> 7\,mgL^{-1}$ PACl, peak 2 could be completely removed. On peak 3, at PACl dose $> 2\,mgL^{-1}$, the areal removal increased linearly with increasing PACl dosage. For instance, the relationship for water A is (areal removal (in %) = 6.49 PACl dosage (in mgL^{-1}) + 6.00 (in %)), with $r^2 = 0.989$, whereas that for water B is (areal removal (in %) = 4.34 PACl dosage (in mgL^{-1}) + 27.8 (in %)), with $r^2 = 0.996$. On peak 4, the PACl dosage yielded negligible areal removal.

The linearity between UV_{254} areal removal (%) and the PACl dosage on peak 3 indicates a stoichiometric relationship between the removal of UV_{254}-sensitive compounds and the dosed PACl. Restated, the PACl-NOM complex may be formed for peak 3. This linearity was observed at dosage $> 2\,mgL^{-1}$ only, indicating that the dosed PACl tends to react with compounds in peak 1, and then peak 2 first, and then the excess dosed PACl interacts with those compounds in peak 3.

Coagulation of Raw Waters Using Processes 1–4 by EPC Mechanism

The ECP coagulation efficiencies for turbidity removal using processes 1–4 at PACl of $1.2\,mgL^{-1}$ as Al_2O_3 are demonstrated in Figs. 4a and 4b. Processes 1–4 at ECP produced the turbidity removal rates for water A as 47, 0.2, 75, and 67% and for water B, 78, 55, 91, and 90%, respectively. The residual turbidities for processes 1–4 for coagulated waters A and B at $1.2\,mgL^{-1}$ followed Process 2 > Process 1 > Process 4 = Process 3.

The ECP coagulation efficiencies for NOM removal using processes 1–4 at PACl of $1.2\,mgL^{-1}$ as Al_2O_3 are demonstrated in Figs. 5 and 6. Processes 1–4 effectively removed peak 1 and partially removed peak 2. Specifically, process 1 removed 88% of peak 1 for water A and 93% of
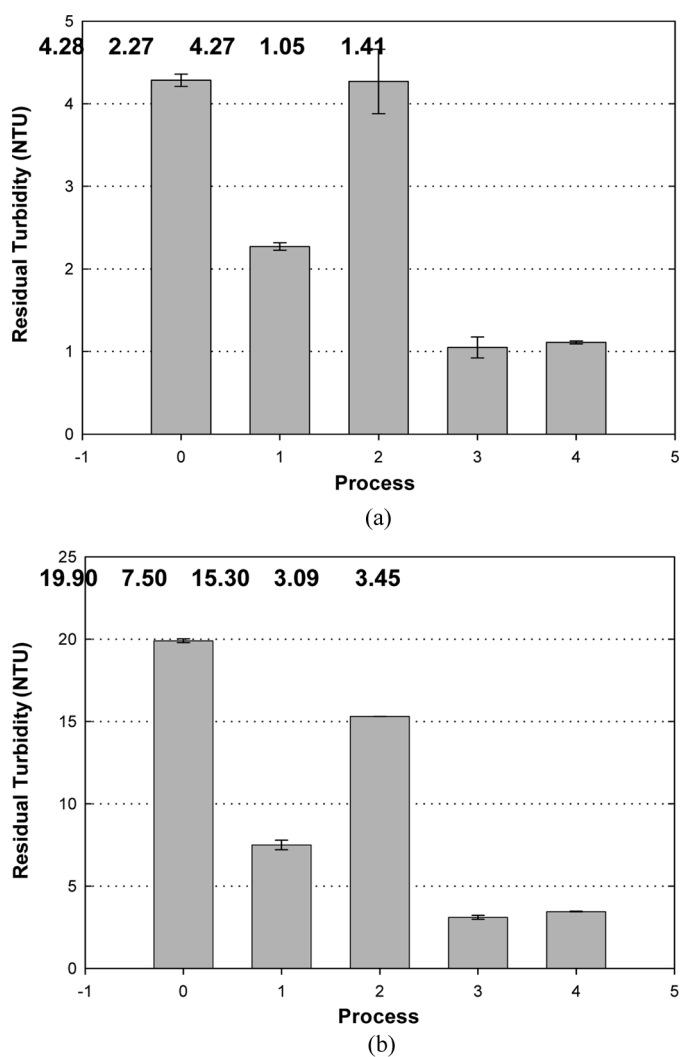

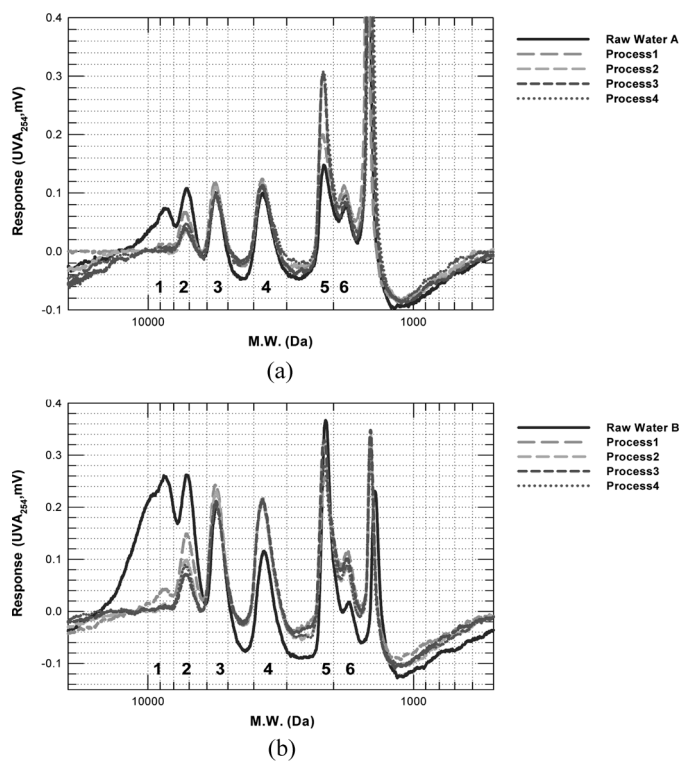FIG. 4. Residual turbidities for raw and coagulated water samples. PACl. Processes 1–4. 0 denotes the raw water. (a) Water A; (b) water B.

FIG. 5. SEC-UV_{254} spectra for raw and coagulated water samples. PACl. Processes 1–4. (a) Water A; (b) water B.
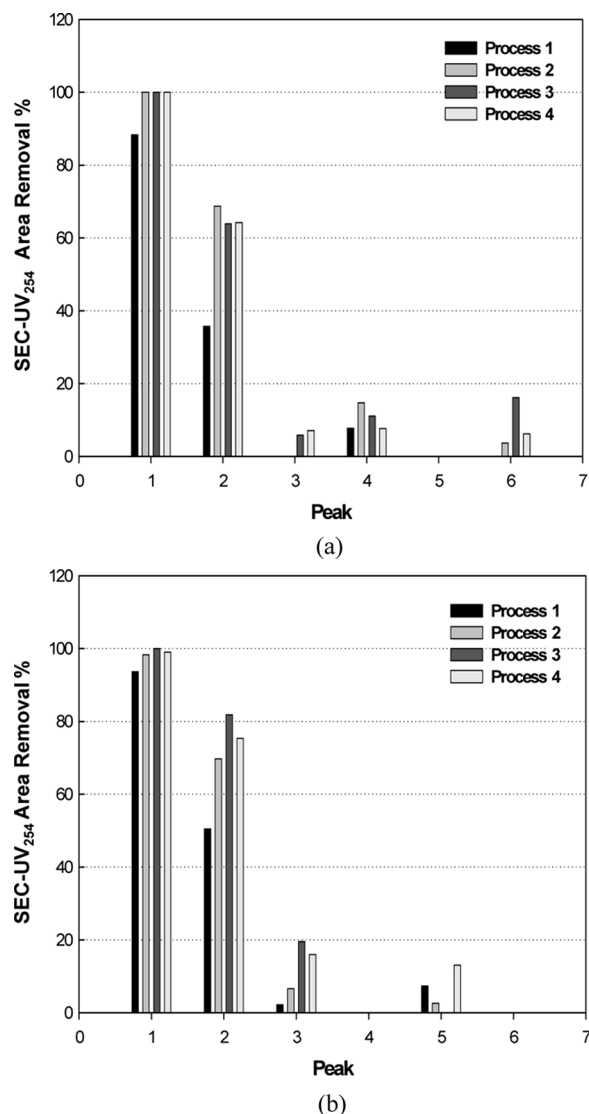
FIG. 6. Areal removal rates for raw and coagulated water samples. PACl. Processes 1–4. (a) Water A; (b) water B.

peak 1 for water B. The corresponding removal rates for processes 2–4 were all 100%. On peak 2, process 1 removed 36% of peak 2 for water A and 50% of peak 2 for water B. Processes 2–4 removed 63–69% of peak 2 for water A and 69–81% of peak 2 for water B. On peak 3, processes 1–4 removed <20% of NOM, with process 1 leading to the least removal rate. A few NOM in peak 4 were removed by the ECP mechanism.

DISCUSSION
Coagulation Removal of Turbidity and NOM

The characteristics of raw water are profiled by fluorescence EEM and HP-SEC, respectively, in Table 1 and Fig. 2. Raw water B represents the case of high turbidity (19.9 NTU), low DOC (0.73 mgL^{-1} as C), with fulvic

acid–like species, whereas raw water A served as a reference of low turbidity (4.28 NTU), high DOC (1.62 mgL^{-1} as C), and a relatively less contaminated sample. The molecular weight distributions of two raw waters are shown in Figs. 2a and 2b. The two baselines provided reference for comparisons of removal efficiency of subsequent experiments with various coagulant species and operation conditions. The NOM of water samples had a molecular weight between 1000 and 11,000 Da. Five peaks appeared on the HP-SEC graphs, including (1) 7500–11,000, (2) 5500–7500, (3) 4000–5500, (4) 2000–4000, and (5) 1000–2000 Da (water peak). A chemical coagulation process adopted in drinking waterworks was reported only efficient to remove high-molecular-weight dissolved organic matter (5500–11,000 Da). However, low-molecular-weight dissolved organic matters are normally precursors of carcinogenic DBPs. Moreover, several trace contaminants, such as endogenic disruption chemicals that are of high health concern, are normally of low molecular weights (150–500 Da) and are not readily removed in conventional water treatment process.

EPC Mechanism

The surface charge of suspended particles in water A and water B was not neutralized with the presence of 1.2 mgL^{-1} PACl (Fig. 1); however, this dosage removed most of peak 1 and about 50–80% of peak 2. During the EPC stage the surface charges of suspended particles are far from neutralized. The proposal is that the patches of surface of particles are becoming non-charged so coagulation can occur locally.[30] Overall charge neutralization does not correspond to the turbidity removal from the present, low-alkalinity, low- to medium-turbidity raw waters.

Comparing the performances between process 1 and process 3 in Table 2 revealed that intensive rapid mixing benefits turbidity removal, likely being attributable to the so-induced more uniform dispersion of dosed PACl over the entire suspension and the more frequent particle collision to form nuclei flocs at extensive mixing. Comparing the turbidity removal rates between process 2 and process 3 reveals the significant role of slow mixing to allow large flocs to form for separation. Rapid mixing without subsequent slow mixing left numerous fine particles that could not settle well. Comparing the turbidity removal between process 1 and process 4 indicated that the use of a two-stage coagulation produced nuclei flocs in the first 3-min rapid mixing phase assisted capture of fine particles to reach higher turbidity removal in the slow mixing and the settling stages.

The 1.2 mgL^{-1} PACl almost completely removed peak 1 and about 50–80% of peak 2. The corresponding removals of peaks 3–6 were negligible. For both water samples, the SEC-UV$_{254}$ areal removal rates follow Processes 2–4 >

Process 1 for both peaks 1 and 2. Restated, the strong and prolonged rapid mixing enhances NOM removal, whereas the presence of slow mixing is irrelevant to NOM removal rate. Additionally, the practice of two-stage coagulation reveals no noticeable benefit to the NOM removal. Restated, the mechanisms corresponding to remove peak 1 and peak 2 NOM differs from those for turbidity removal. It is likely that the mechanisms for NOM removal include complexation of ions and NOM to form insoluble flocs to be removed in the settling stage.

The mechanisms corresponding to NOM removal differ from those for turbidity removal. The proposal assuming adsorption of NOM on the suspended particles or on the coagulated particle flocs is not relevant. It is likely that the mechanisms are complexation of ions and NOM to form insoluble aggregates that could be removed in filtration before TOC tests.

CONCLUSIONS

To achieve efficient coagulation with minimum coagulant dose, the EPC mechanism was applied herein to remove turbidity and natural organic matters in water using four different dosing–mixing schemes. The organic matters in the water samples were fractionated by the HP-SEC system and by the EEM fluorescence spectra. There exist six UV_{254} absorption peaks for the SEC diagram for two raw water samples. In both waters, addition of $1.2\,\mathrm{mgL^{-1}}$ PACl nearly completely removed the peak 1 (7500–10,000 Da) but only partially removed the peak 2 (5500–7500 Da). A strong and prolonged rapid mixing benefits the NOM removal, whereas slow mixing is irrelevant to NOM removal. Additionally, the practice of two-stage coagulation has no benefits to the noted NOM removal. The mechanisms for NOM removal are proposed to be complexation of ions and NOM to form insoluble flocs to be removed in the separation stage. On turbidity removal, slow mixing significantly assists formation of large flocs for gravity separation. The use of two-stage coagulation produced a higher turbidity removal rate compared with that using one-stage coagulation. The mechanisms corresponding to NOM removal differ from those for turbidity removal.

ACKNOWLEDGMENT

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