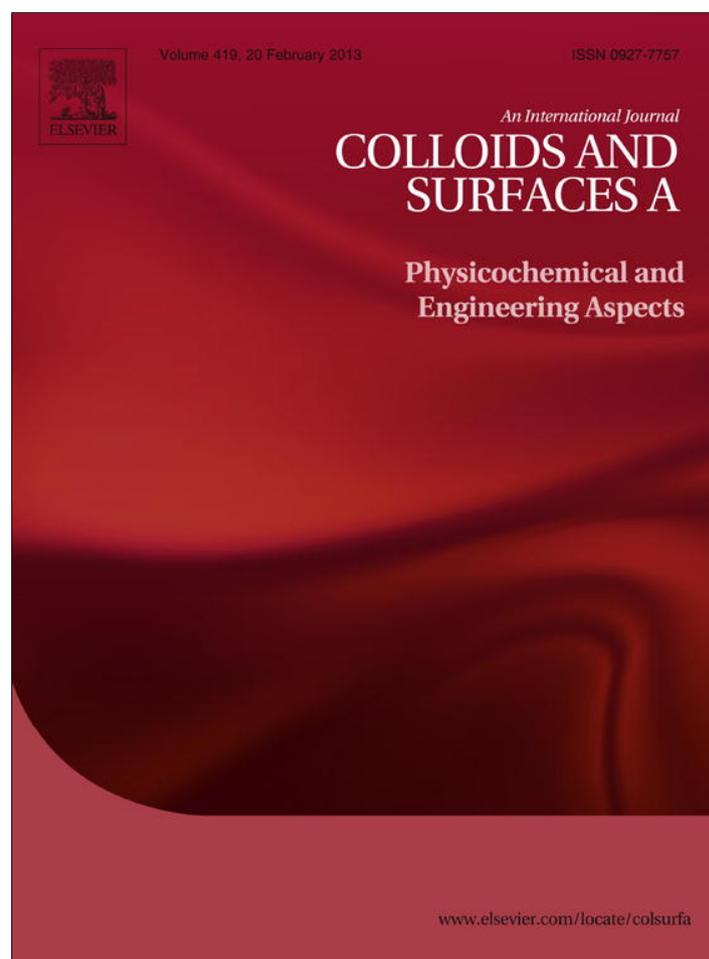


Provided for non-commercial research and education use.
Not for reproduction, distribution or commercial use.



This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

<http://www.elsevier.com/copyright>

cases. High turbidity also increases the contamination of influent waters, as most organic contaminants are adsorbed onto suspended particles. The jar test is not appropriate for waters with highly fluctuating water quality [6–8].

Studies have been performed to improve the jar test for the effective coagulation. For example, a matrix approach of coagulant feeding based on the results of a conventional jar test in real time has been adopted [9–11]. Zeta-meter control by measuring the electrical instability of particles during the coagulation process has also been effective [12–14]. However, these methods still require further optimization.

A streaming current detector (SCD) can be used to rapidly and accurately determine the proper coagulant dose. SCD devices operate in real time to quantify the amount of negatively charged particles in the bulk phase. Injections of coagulant controlled by a SCD system offset the negative charges. Given that aggregation occurs due to electrical neutrality, this system can effectively separate particles in real time. Previous studies on the application of SCD systems have been performed [3,15–22]. However, control of the coagulant dose by the SCD device remains challenging in highly turbid water with large fluctuations [3].

Recently, statistical methods such as response surface methodology, factorial design analysis, and multiple linear regression (MLR) has been applied to optimize dose-related conditions [18,20,23–27]. Using these methods, it is possible to prepare for unknown influent conditions at water treatment plants. By applying these approaches to a SCD system, it should be possible to control purification processes.

In this study, the feasibility of SCD application was evaluated in a lab-scale coagulation experiment. Using a SCD system, the effective removal of turbidity for especially highly turbid waters, was examined.

2. Materials and methods

2.1. Chemicals

Kaolin particles ($\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$; Sigma–Aldrich) were used to prepare synthetic water sample suspensions. NaHCO_3 (Sigma–Aldrich) was injected to ensure sufficient alkalinity. Commercial 17% polyaluminum chloride (PAC: $[\text{Al}_2(\text{OH})_n\text{Cl}_{6-n}]_m$; Samgoo Chemical, Korea) and 0.001 N poly-diallyldimethyl-ammonium chloride (poly-DADMAC: $(\text{C}_8\text{H}_{16}\text{NCl})_n$; AFG, Germany) were used as coagulants.

2.2. Preparation of synthetic water

Kaolin particle was mixed with distilled water to make various turbid suspensions. Five kaolin concentrations of 100, 250, 500, 750, and 1000 mg/L were used to provide the turbid waters in a range from 57.1 to 619.9 NTU. pH of the solution was adjusted to 7 by adding NaHCO_3 .

2.3. Analytical instruments

A streaming current detector (CAS; AFG Analytic GmbH, Leipzig, Germany) was used for charge titrations during the coagulation process. This system interfaces with a computer to titrate the charge by controlling the injection of coagulant. The suspension is titrated in the cylinder cell of CAS system. When the Teflon piston is moving up and down four times per second, streaming current readings were monitored by electrodes at the upper and lower ends of cylinder cell. Usually, 10 mL of kaolin suspension was titrated in the cylinder cell, and 10 μL of coagulants are injected into the cell every 30 s until the achieving electrical neutrality.

To verify the results of the aggregation conditions, a jar test (SF6; Chosun Instruments, Seoul, Korea) was performed simultaneously. For the jar test, a sample volume of 1 L was stirred rapidly at 140 rpm for 5 min and then at 70 rpm for 20 min, followed by settling for 1 h. After the settling period, the supernatant turbidity was measured three times using a turbidimeter (2100P; Hach, Loveland, CO, USA). A pH meter (IstekModel 52 A; Orion, Reno, NV, USA) was used to maintain proper pH levels. The solution temperature was maintained at 25 °C.

2.4. Statistical analysis

Statistical analyses were conducted to verify the data. Student's *t*-test, the *F*-test, Tukey's Honestly Significant Difference test (HSD), analysis of variance (ANOVA), and multiple linear regression (MLR) were used to evaluate statistical significance. Suitability of study model was verified in the 95% significance level ($\alpha = 0.05$). R-commander was used as a statistical package, and Minitab 14 was used to crosscheck the accuracy.

3. Results and discussion

3.1. Adjustment of pH in the jar test

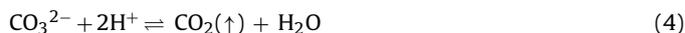
As the pH plays an important role in coagulation [24,28–35], the pH changes during PAC and poly-DADMAC injections were monitored. The pH change during PAC injection was larger than that during poly-DADMAC injection (Fig. 1a).

With increasing doses of coagulant, the pH decreased. This may be due to the fact that Al^{3+} ions can donate protons through a hydration reaction, resulting in a pH decrease as shown in Eq. (1). Under acidic conditions, the electric repulsion between $\text{Al}(\text{OH})^{2+}$ ions and the protons surrounding the particles inhibit particle aggregation. Recent studies have reported the optimal coagulation conditions to maximize $\text{Al}(\text{OH})_3$ formation in a bulk solution [24,25,34,36–39], as shown Eqs. (1) and (2):



At water treatment plants (WTPs), the pH of the influent is usually acidic during the pre-coagulation stage, and the pH is adjusted by adding NaOH or CaCO_3 . When the appropriate pH condition is achieved at WTPs, particle aggregation is not significantly affected [30,31,33,40–43]. Thus, the pH adjustment was conducted when PAC was used as a coagulant.

To provide alkalinity, NaHCO_3 , Na_2CO_3 , and CaCO_3 were injected into samples containing PAC (100 mg/L), and the changes in pH were observed (Fig. 1b). Bicarbonate (HCO_3^-) ion, which was the dominant ion at neutral pH, and carbonate (CO_3^{2-}) ion, which was dominant at basic pH, resulted in neutralization owing to the proton donated by PAC, as shown in Eqs. (3) and (4). The neutralization of carbonate is greater than bicarbonate, and can affect the pH.



Previous coagulation studies with PAC have shown that the optimal pH is 6–7. At neutral pH, bicarbonate injection had more significant effect than carbonate, as shown in Fig. 1(b) [6,7]. Therefore, NaHCO_3 was used to control the pH during coagulation process. PAC coagulation at different pH in this study was similar to that in previous studies (Fig. 1c).

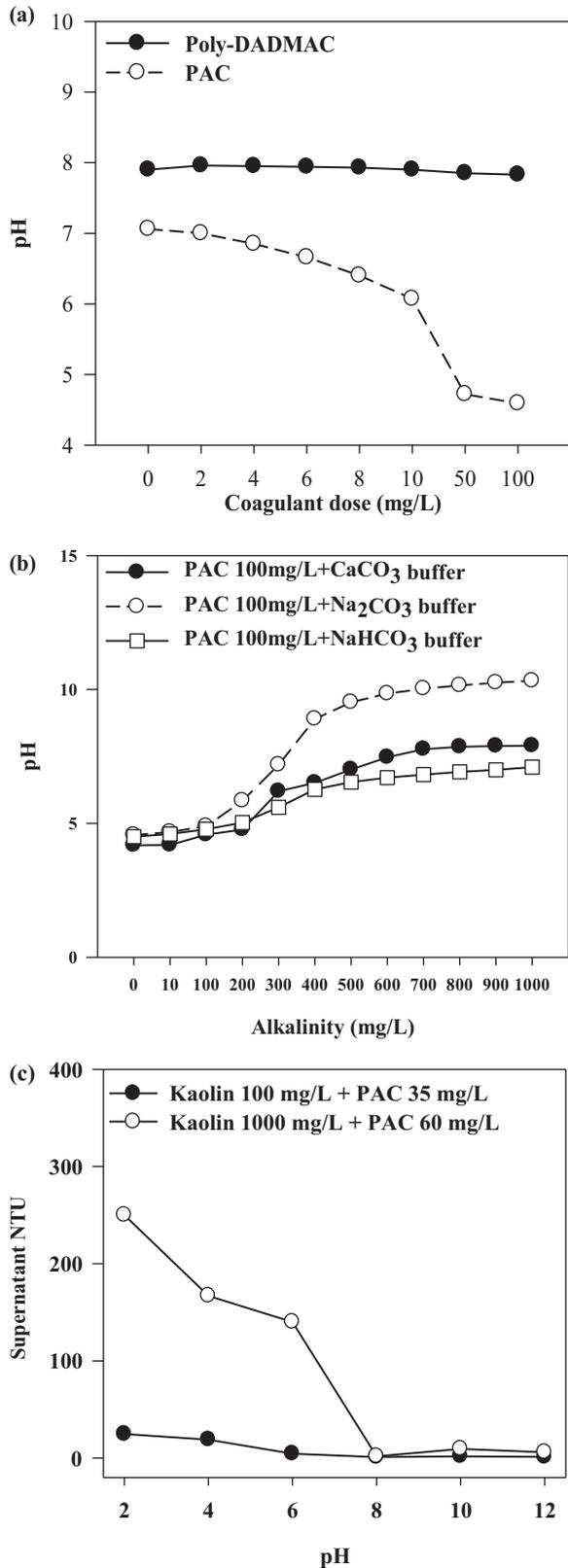


Fig. 1. Range of pH and alkalinity adjustments: (a) coagulant acidic effects of PAC and poly-DADMAC, (b) carbonate series buffering with CaCO₃, Na₂CO₃, and NaHCO₃, and (c) pH of jar test with PAC in kaolin suspension: 100 mg/L, and 1000 mg/L.

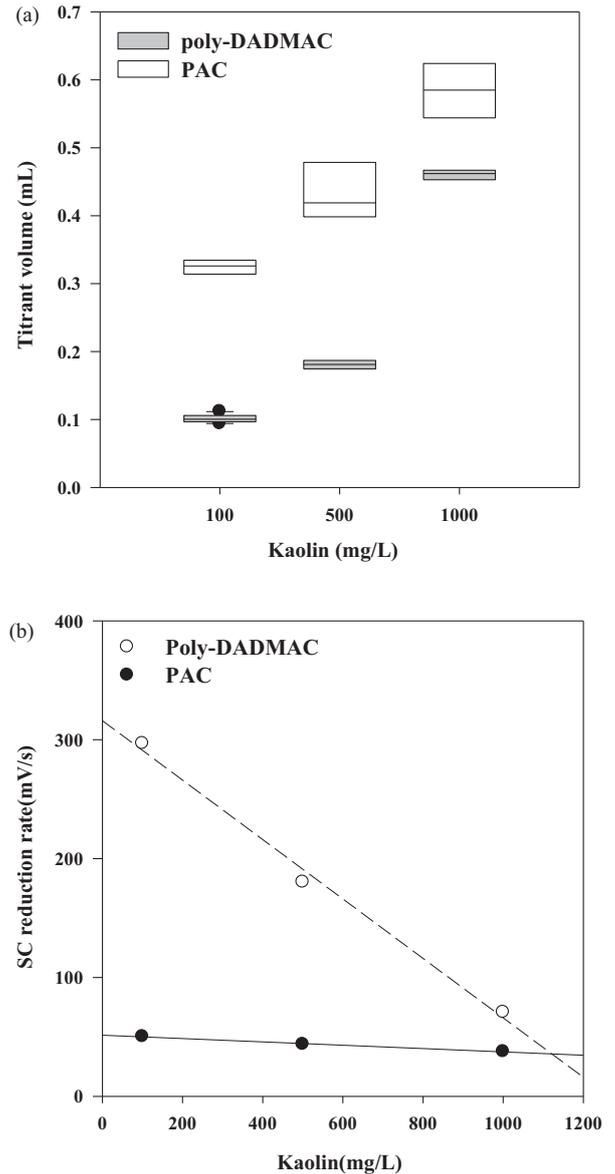


Fig. 2. Charge titration and streaming current reduction using PAC and poly-DADMAC: (a) CAS titration, and (b) streaming current (SC) charge reduction rates in kaolin suspension: 100, 500, and 1000 mg/L (pH=7).

3.2. Characteristics of charge titration

PAC and poly-DADMAC were added as titrants, and the charge of solutions at kaolin concentrations of 100, 250, 500, 750, and 1000 mg/L was measured using a CAS system. Each measurement was repeated ten times. According to ANOVA and Tukey's HSD, the results did not differ significantly among kaolin concentrations of 100 ~ 250 and 500 ~ 750 mg/L (P -value > 0.05). Therefore, final kaolin concentrations of 100, 500, and 1000 mg/L (50 ~ 600 NTU) were used for subsequent titrations.

Fig. 2(a) shows the PAC and poly-DADMAC titration results using the SCD system. As increasing kaolin concentration, higher titrant doses were needed, implying that increasing turbidity in the bulk solution increased the required doses of PAC and poly-DADMAC coagulant. This result might be from the inter-particle bridging processes between particles and polyelectrolyte, which is a critical factor for achieving electrical neutrality [6,44]. Interestingly, the PAC titrant dose was significantly higher than the one in poly-DADMAC, indicating that much less doses of poly-DADMAC was

Table 1
Turbidity obtained by Jar-test result with the dose obtained by CAS

Kaolin concentration (mg/L)	Coagulant			
	PAC		poly-DADMAC	
	Dosage (mg/L)	Residual turbidity (NTU)	Dosage (mg/L)	Residual turbidity (NTU)
100	3.24	7.93 ~ 8.02	1.02	81.9 ~ 82.5
500	4.34	7.46 ~ 7.47	1.82	328 ~ 335
1000	5.84	5.47 ~ 5.55	4.62	623 ~ 626

Table 2
Conversion to calculated coagulant dosage

Kaolin concentration (mg/L)	Calculated dosage (titrant → coagulant)			
	PAC → PAC (mg/L)		Poly-DADMAC → PAC (mg/L)	
100	35		35	
500	46.88	($\cong \frac{4.34}{3.24} \times 35$)	62.58	($\cong \frac{1.82}{1.02} \times 35$)
1000	63.08	($\cong \frac{5.84}{3.24} \times 35$)	158.50	($\cong \frac{4.62}{1.02} \times 35$)

required compared to dose of PAC for reducing negative charges [18–19].

Fig. 2(b) shows the unit dose of titrant required to reduce the streaming current reading, which represents the charge reduction, with an injection interval of 30 s. The streaming current (SC) reduction rate in Fig.2(b) was expressed by Eq. (5).

$$SC_{reduction\ rate}(mV/s) = \frac{\text{reduced } SC(mV)}{\text{measurement time}(s)} \quad (5)$$

A jar test was also performed with CAS titration, and the supernatant turbidity of the treated water after coagulation was measured. With the CAS titration, PAC was more effective than poly-DADMAC in removing the kaolin particles (Table 1). However, the supernatant turbidities at increased kaolin concentrations were higher than 1 NTU, indicating that the PAC dosage obtained by CAS titration did not achieve turbidity levels which are sufficient for satisfying US EPA drinking water standard (<1 NTU) [45].

Although electrical neutrality was achieved by CAS titration, a supernatant turbidity higher than 1 NTU might result from not only by interparticle bridging effect which is insufficient for diffusion during flocculation, but also by the effect of depletion force [46]. Therefore, the sweep flocculation of aluminum hydroxide precipitation might not have occurred, and the increased coagulant dosage would be required [46–50]. To maintain low supernatant turbidity, the jar test result (at equivalent doses) can be applicable to the CAS titration, before converting the coagulant dose.

When considering the conversion of coagulant dosage, the negative charge distribution in the bulk solution can be interpreted based on the titrant dose ratio [18,19]. The coagulant dose can be calculated according to the SCD titration ratio, as shown in Eq. (6).

$$K_{amp} = \frac{i_M}{i} \quad (6)$$

where K_{amp} is an amplifier constant, i is conventional titrant dose which were already known turbidity and coagulant dose (mg/L), and i_M is the measured titrant dose in the unknown sample. Eq. (6) indicates the ratio of titrant dose of an unknown sample (i_M) divided by conventional titrant dose (i).

When applied to a sample of an unknown influent, the optimal coagulant dosage is expressed as Eq. (7):

$$C_{dose} = K_{amp} \times J_{condition} \quad (7)$$

where C_{dose} is the optimal coagulant dosage of unknown sample, and $J_{condition}$ is the jar test condition which was already known turbidity and coagulant dose. Based on Eq. (7), the coagulant dose was calculated according to the kaolin concentration. In the jar test, the PAC dose was 35 mg/L at 100 mg/L of kaolin.

Considering the jar test results under titration dose (Table 1), poly-DADMAC did not function well as a coagulant. Therefore, the results of poly-DADMAC titration were converted to PAC dose. The titrant dose ratios in the poly-DADMAC and PAC jar tests were applied to the dosage calculation using Eq. (7), and the converted results are shown in Table 2.

3.3. Multiple linear regression analysis

Multiple linear regression (MLR) analysis was used to verify the suitability of the calculated values (Table 2), and to construct a significant optimal dosage model. The basic regression equation of the model is shown in Eq. (8):

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_1 X_2 + \beta_4 X_1^2 + \beta_5 X_2^2 + \epsilon \quad (8)$$

where Y is the optimal dosage which is calculated from the model (mg/L), X_1 is the titrant dose (mg/L), X_2 is the kaolin concentration (mg/L), $X_1 X_2$ is the interaction term (titrant dose times kaolin concentration), X_1^2 is the quadratic term of titrant dose, X_2^2 is the quadratic term of kaolin concentration, β is the regression parameter, and ϵ is the measurement error.

Titrant dose (X_1) and the kaolin concentration of the suspension (X_2) were chosen as the independent variables, and the calculated dose was selected as the response variable (Y). Also, the quadratic term and the interaction of X_1 and X_2 were added.

Individual correlations of Y , X_1 , and X_2 are presented in Table 3. Table 2 shows that the independent variables were positively correlated. Fig. 3 also shows that the two plots were crossed, indicating the interaction between two variables [51,52]. The suitability of all independent variables in the model was determined using the step-wise selection method, and inappropriate variables were excluded. Some values in the final model were applied to perform interpolation. The results were used to conduct a jar test in order to confirm that the model was accurate.

3.3.1. PAC titration model

The overall fitness of the model was confirmed with the F -test, and the significance of coefficients was examined using Student's

Table 3
Pearson correlation coefficients of variances

Relation	Titrant dose (X_1)	
	PAC	poly-DADMAC
Optimal dose (Y)	0.9714	0.9938
Kaolin concentration (X_2)	0.9707	0.965

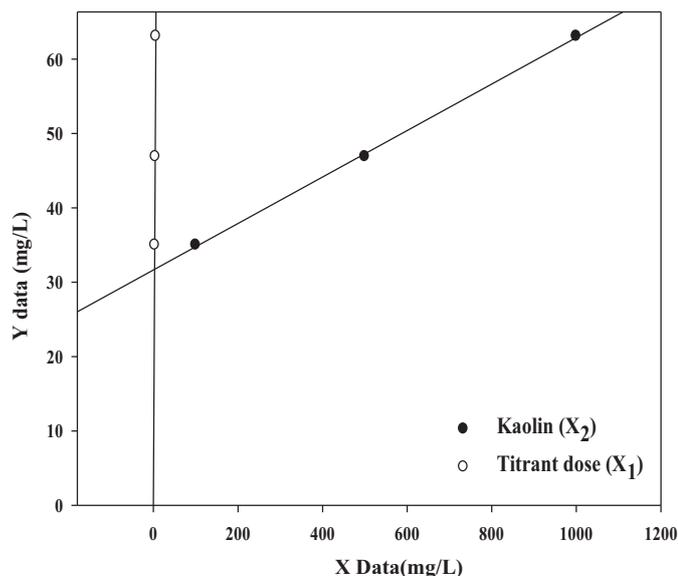


Fig. 3. Correlation between kaolin concentration and PAC titration. Y data indicates coagulation dose, and X data represents kaolin concentration (●) and titrant dose (○).

t-test. Measurements were conducted at 100, 500, and 1000 mg/L of kaolin, and were repeated 10 times. A total of 30 samples were analyzed, and the central limit theorem significantly reduced the labor to estimate model for large measurements of *n*. Based on the measurements, MLR analysis was performed. The results of the *F*-test showed that the model explained the overall values, and the *t*-test confirmed that each coefficient was statistically significant (*P*-values < 0.05).

The quadratic terms of the equation were excluded during the stepwise selection, and Shapiro–Wilk’s normality test showed that residual errors were normally distributed in the model. The statistical result indicated that the data were properly explained by the model. The model equation is expressed as Eq. (9)

$$Y = 34.06 - 0.6304X_1 + 0.0268X_2 + 0.001011X_1X_2 \quad R^2 = 0.99 \quad (9)$$

A comparison between the observed values and the modeled values is shown in Fig. 4(a). Titration results for calculating coagulant dosages were repeated 30 times, and the results were confirmed using the jar test. The supernatant turbidity of all predictive values was less than 1 NTU. The result was also confirmed by Student’s *t*-test (*P*-values < 0.05), implying that the proposed model effectively predicted the coagulation process. The distribution of the predicted values is also presented as a contour plot (Fig. 4(b)). Fig. 4(b) indicates that the PAC model was linear with constant variation, and the coagulant dose tended to increase gradually.

3.3.2. Poly-DADMAC titration model

Since poly-DADMAC is ineffective as a coagulant in the kaolin aggregation (Table 1), the titration results using the poly-DADMAC were converted to the PAC doses, and then applied into the regression model. As for PAC titrations, 30 samples were analyzed, and the central limit theorem was satisfied. The MLR analysis showed that the model accurately described the data overall, and each coefficient was significant based on the *t*-test (*P*-values < 0.05). Stepwise selection results, excluding the quadratic terms, are shown in the PAC model and the residual error satisfied normality by Shapiro–Wilk’s test. The model showed that the CAS data

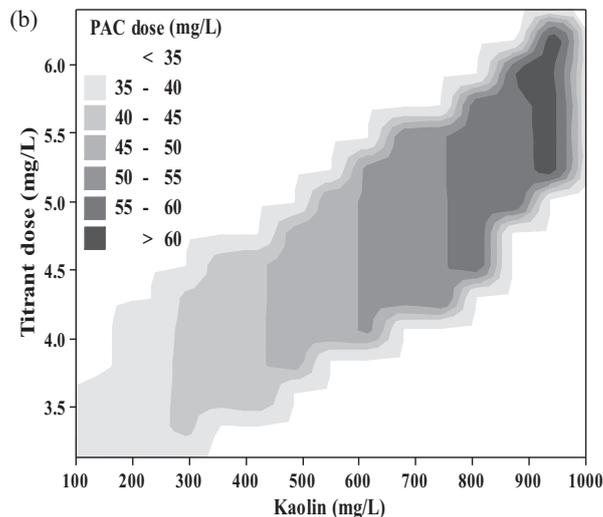
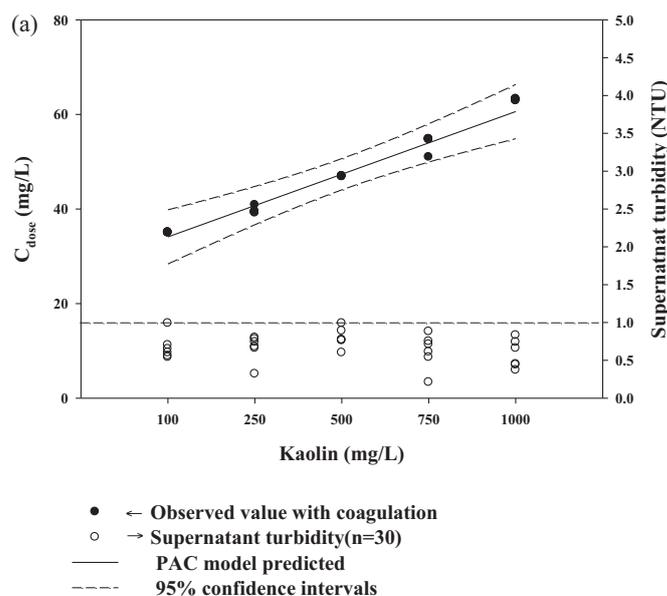


Fig. 4. PAC titration model profiles (a) jar test results of the model, and (b) contour plot of the dosage. The height of contour plots indicates the variation of charge titration in kaolin suspension: 100, 250, 500, 750, and 1000 mg/L (pH = 7).

properly explained the results. The expression of the model was shown in Eq. (10):

$$Y = 59.7277 - 33.8285X_1 + 0.0481X_2 + 0.0448X_1X_2 \quad R^2 = 0.99 \quad (10)$$

To test the applicability of this model, total 30 poly-DADMAC titrations were performed. The titration results using poly-DADMAC were converted to PAC doses and the jar test was performed to check the turbidity using these PAC doses. The coagulation results are shown in Fig. 5(a), showing that the measured supernatant turbidities in all samples were less than 1 NTU.

The distribution of the predicted values is also expressed as a contour plot (Fig. 5(b)). The poly-DADMAC model was linear with a gradually increasing dose and showed less variation than the PAC model. Interestingly, the result indicates that the poly-DADMAC model predicted significantly larger doses than the PAC model.

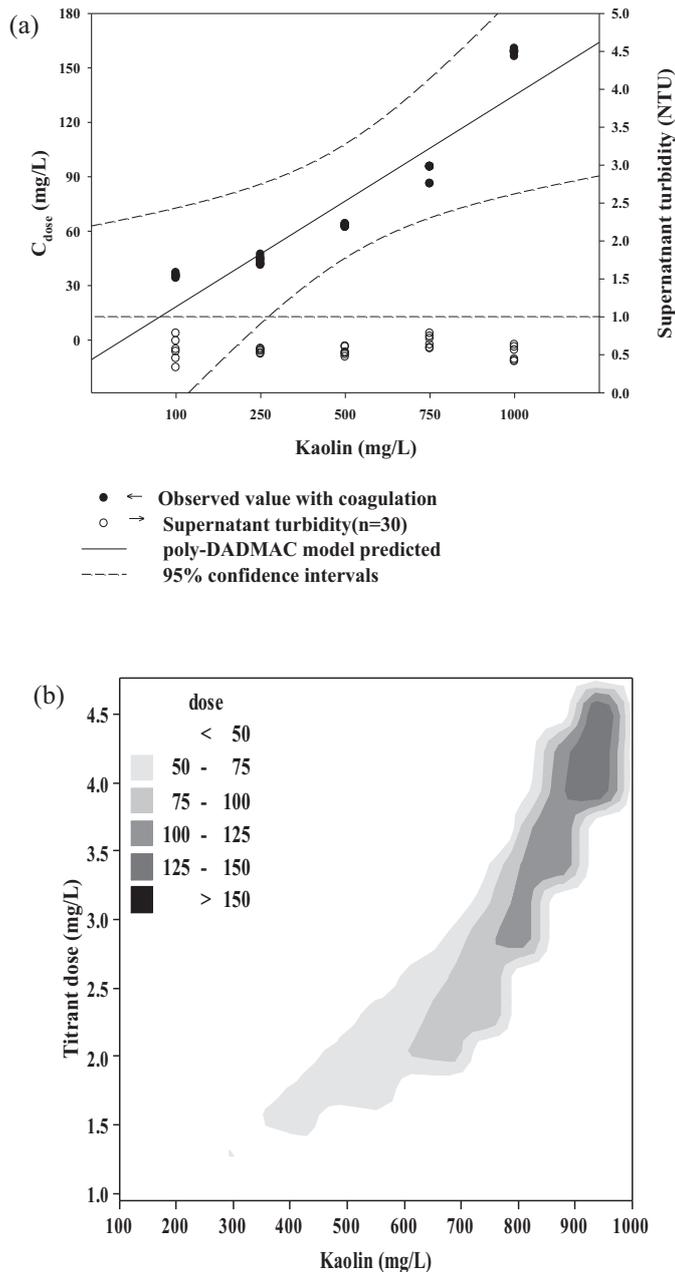


Fig. 5. Poly-DADMAC titration model profiles: (a) Jar-test results of the model (b) Contour plot of dosage in kaolin suspension: 100, 250, 500, 750, and 1000 mg/L (pH=7). The height of contour plots indicates the variation of charge titration in kaolin suspension: 100, 250, 500, 750, and 1000 mg/L (pH = 7).

3.4. Comparison of titration models

It is important to optimize the dose of coagulants used in the coagulation process in WTPs in order to reduce costs and minimize sludge formation. In addition, the optimal coagulation can decrease the fouling in the filtration step [7,52]. When the PAC and poly-DADMAC models developed here were compared by MLR analysis, lower coagulant doses were required in the PAC model under highly turbid conditions (Fig. 6(a)).

Trial and error methods identified the PAC doses required at kaolin concentrations of 100, 250, 500, 750, and 1000 mg/L, and these doses were compared with the model results. The result shows that the PAC model and jar test results were very similar, with a maximum dose difference of less than 3 mg/L. In contrast, the

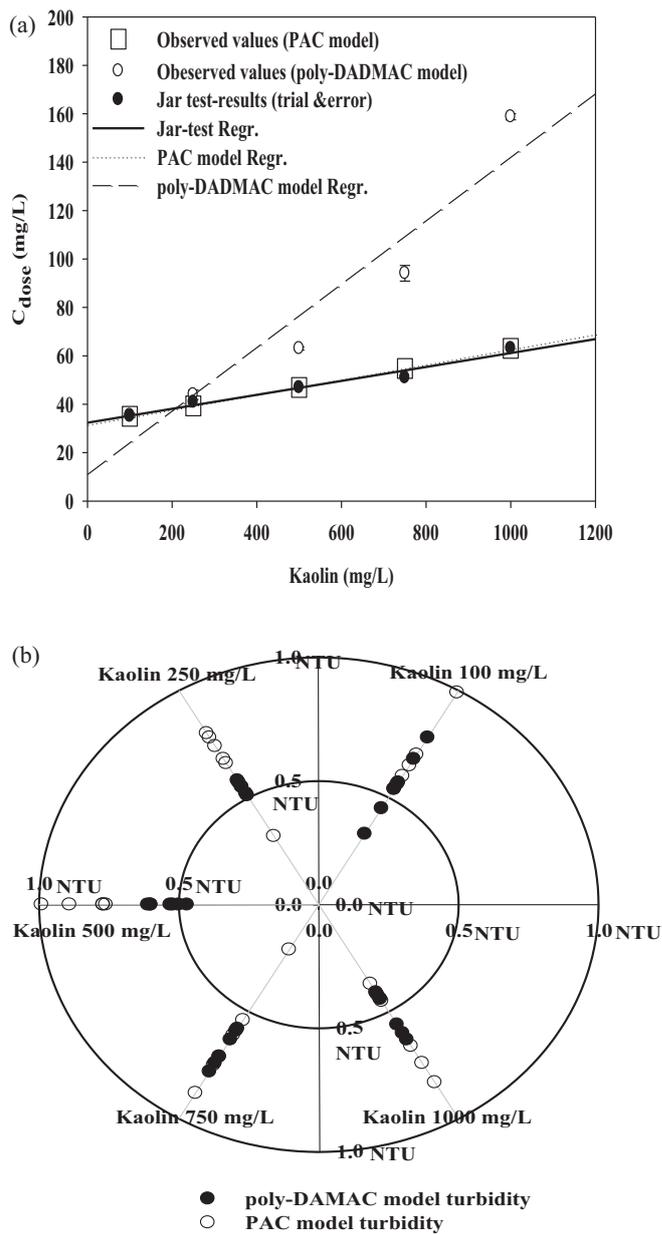


Fig. 6. Comparison of the effects of the PAC and poly-DADMAC models: (a) optimal dosage difference in the multiple linear regression (PAC vs. poly-DADMAC vs. jar test), and (b) polar plots of supernatant turbidity (PAC vs. poly-DADMAC). Points in polar plots circles represent the jar test results within supernatant turbidity levels (0, <0.5, and <1 NTU).

coagulation doses were significantly different between the poly-DADMAC model and jar test results. This result implies that the PAC dosage is overestimated when using the poly-DADMAC model. When the poly-DADMAC dosage was converted to the PAC dosage, overdosing was observed due to the large charge reduction with poly-DADMAC. Thus, heterogeneous titration-coagulation required careful evaluation of coagulant dose. On the other hand, lower turbidity was observed in the poly-DADMAC model compared with the PAC model (Fig. 6(b)).

4. Conclusions

In this study, homogeneous (PAC only) and heterogeneous (poly-DADMAC-PAC) charge titration-coagulation models were examined using a streaming current detector. The kaolin particle

solution formed highly turbid suspensions. Compared with the poly-DADMAC model, the PAC titration-coagulation model controlled coagulation more effectively. The maximum difference in PAC dose between the PAC model and jar test was less than 3 mg/L, and the supernatant turbidity with the model was less than 1 NTU, suggesting that the high turbidity influent waters in WTPs can be effectively controlled. In addition, the application of SCD can reduce the time required for the jar test and the cost of overdosing coagulant. With the appropriate charge titration condition and initial coagulation dosage for sweep flocculation, SCD can be used to determine the optimum coagulant dosage in WTPs.

Acknowledgement

This research was supported by the Seoul R&BD Program (No. JP100072) through the Seoul Metropolitan Government of Korea.

References

- [1] Z.H. Yang, J. Huang, G.M. Zeng, M. Ruan, C.S. Zhou, L. Li, Z.G. Rong, Optimization of flocculation conditions for kaolin suspension using the composite flocculant of MBFGA1 and PAC by response surface methodology, *Bioresour. Technol.* 100 (2009) 4233–4239.
- [2] A. Campbell, The potential role of aluminium in Alzheimer's disease, *Nephrol. Dial. Transplant* 17 (2002) 17–20.
- [3] S. Xia, X. Li, Q. Zhang, B. Xu, G. Li, Ultrafiltration of surface water with coagulation pretreatment by streaming current control, *Desalination* 204 (2007) 351–358.
- [4] J.C. Vickers, M.A. Thompson, U.G. Kelkar, The use of membrane filtration in conjunction with coagulation processes for improved NOM removal, *Desalination* 102 (1995) 57–61.
- [5] R. Bryant, Optimizing coagulation with the Streaming Current Monitor, *J. N. England Water Works Assoc.* 110 (1996) 268–271.
- [6] J.L. Lin, C. Huang, J.R. Pan, D. Wang, Effect of Al(III) speciation on coagulation of highly turbid water, *Chemosphere* 72 (2008) 189–196.
- [7] J.H. Kweon, H.W. Hur, G.T. Seo, T.R. Jang, J.H. Park, K.Y. Choi, H.S. Kim, Evaluation of coagulation and PAC adsorption pretreatments on membrane filtration for a surface water in Korea: A pilot study, *Desalination* 249 (2009) 212–216.
- [8] D. Di Bernardo, Performance of two-stage filtration system for treating high turbidity water, *J. Water Supply Res. Technol., AQUA* 55 (2006) 499–515.
- [9] P. Aragonés-Beltrán, J. Mendoza-Roca, A. Bes-Piá, M. García-Melón, E. Parra-Ruiz, Application of multicriteria decision analysis to jar-test results for chemicals selection in the physical-chemical treatment of textile wastewater, *J. Hazard. Mater.* 164 (2009) 288–295.
- [10] S. Park, H. Bae, C. Kim, Decision model for coagulant dosage using genetic programming and multivariate statistical analysis for coagulation/flocculation at water treatment process, *Korean J. Chem. Eng.* 25 (2008) 1372–1376.
- [11] W. Xiaojie, J. Yunzhe, L. Xiaojing, Research on the prediction of water treatment plant coagulant dosage based on feed-forward artificial neural network, in: *IEEE* (2011) 1615–1617.
- [12] H. Moayedi, B.B.K. Huat, S. Kazemian, S. Daneshmand, D. Moazami, H. Niroumand, Electrophoresis of suspended kaolinite in multivalent electrolyte solution, *Int. J. Electrochem. Sci.* 6 (2011) 6514–6524.
- [13] A. Morfesis, A.M. Jacobson, R. Frollini, M. Helgeson, J. Billica, K.R. Gertig, Role of zeta (ζ) potential in the optimization of water treatment facility operations, *Ind. Eng. Chem. Res.* 48 (2008) 2305–2308.
- [14] Z. Tang, M.A. Butkus, Y.F. Xie, Enhanced performance of crumb rubber filtration for ballast water treatment, *Chemosphere* 74 (2009) 1396–1399.
- [15] P.H. Cardwell, Adsorption studies using a streaming current detector, *J. Colloid Interface Sci.* 22 (1966) 430–437.
- [16] S.K. Dentel, A.V. Thomas, K.M. Kingery, Evaluation of the streaming current detector—I. Use in jar tests, *Water Res.* 23 (1989) 413–421.
- [17] S.K. Dentel, A.V. Thomas, K.M. Kingery, Evaluation of the streaming current detector—II. Continuous flow tests, *Water Res.* 23 (1989) 423–430.
- [18] S.K. Kam, J. Gregory, Charge determination of synthetic cationic polyelectrolytes by colloid titration, *Colloids Surf., A* 159 (1999) 165–179.
- [19] J. Chen, Dependency of polyelectrolyte complex stoichiometry on the order of addition. 1. Effect of salt concentration during streaming current titrations with strong poly-acid and poly-base, *Colloids Surf., A* 223 (2003) 215–230.
- [20] R. Vespalec, J. Neca, Z. Simek, Measurement of streaming current and calculation of zeta potential for beds of wide-dispersion microparticulate solids interacting with water, *Colloids Surf., A* 92 (1994) 147–157.
- [21] C.A. Walker, J.T. Kirby, S.K. Dentel, The streaming current detector: a quantitative model, *J. Colloid Interface Sci.* 182 (1996) 71–81.
- [22] M. Zaucha, Z. Adamczyk, J. Barbasz, Zeta potential of particle bilayers on mica: a streaming potential study, *J. Colloid Interface Sci.* 360 (2011) 195–203.
- [23] F. Ito, G. Ma, M. Nagai, S. Omi, Study of particle growth by seeded emulsion polymerization accompanied by electrostatic coagulation, *Colloids Surf., A* 201 (2002) 131–142.
- [24] J.P. Wang, Y.Z. Chen, X.W. Ge, H.Q. Yu, Optimization of coagulation-flocculation process for a paper-recycling wastewater treatment using response surface methodology, *Colloids Surf., A* 302 (2007) 204–210.
- [25] C. Ye, D. Wang, B. Shi, J. Yu, J. Qu, M. Edwards, H. Tang, Alkalinity effect of coagulation with polyaluminum chlorides: role of electrostatic patch, *Colloids Surf., A* 294 (2007) 163–173.
- [26] J. Nan, W. He, X. Song, G. Li, Impact of dynamic distribution of floc particles on flocculation effect, *J. Environ. Sci.* 21 (2009) 1059–1065.
- [27] Q. Kang, B. Gao, Q. Yue, W. Zhou, D. Shen, Residual color profiles of reactive dyes mixture during a chemical flocculation process, *Colloids Surf., A* 299 (2007) 45–53.
- [28] T. Chen, B. Gao, Q. Yue, Effect of dosing method and pH on color removal performance and floc aggregation of polyferric chloride-polyamine dual-coagulant in synthetic dyeing wastewater treatment, *Colloids Surf., A* 355 (2010) 121–129.
- [29] J. Hempoonsert, B. Tansel, S. Laha, Effect of temperature and pH on droplet aggregation and phase separation characteristics of flocs formed in oil-water emulsions after coagulation, *Colloids Surf., A* 353 (2010) 37–42.
- [30] P.K. Holt, G.W. Barton, M. Wark, C.A. Mitchell, A quantitative comparison between chemical dosing and electrocoagulation, *Colloids Surf., A* 211 (2002) 233–248.
- [31] W.H. Kuan, C.Y. Hu, Chemical evidences for the optimal coagulant dosage and pH adjustment of silica removal from chemical mechanical polishing (CMP) wastewater, *Colloids Surf., A* 342 (2009) 1–7.
- [32] T.K. Liu, C.J.M. Chin, Improved coagulation performance using preformed polymeric iron chloride (PICI), *Colloids Surf., A* 339 (2009) 192–198.
- [33] U. Nowostawska, S.G. Sander, K.M. McGrath, K.A. Hunter, Effects of coagulants on the surface forces of colloidal alumina under water treatment conditions, *Colloids Surf., A* 266 (2005) 214–222.
- [34] M. Özacar, I.A. Sengil, Evaluation of tannin biopolymer as a coagulant aid for coagulation of colloidal particles, *Colloids Surf., A* 229 (2003) 85–96.
- [35] S. Pal, D. Mal, R.P. Singh, Synthesis, characterization and flocculation characteristics of cationic glycogen: a novel polymeric flocculant, *Colloids Surf., A* 289 (2006) 193–199.
- [36] M.A. Kim, P.J. Panak, D.C. Breban, A. Priemyshev, J.I. Yun, A. Mansel, J.I. Kim, Interaction of actinides(III) with aluminosilicate colloids: part IV. Influence of humic acid, *Colloids Surf., A* 296 (2007) 206–215.
- [37] U. Nowostawska, S.G. Sander, K.M. McGrath, K.A. Hunter, Effect of trivalent metal sulfates on the coagulation and particle interactions of alumina colloids, *Colloids Surf., A* 266 (2005) 200–206.
- [38] D.I. Verrelli, D.R. Dixon, P.J. Scales, Effect of coagulation conditions on the dewatering properties of sludges produced in drinking water treatment, *Colloids Surf., A* 348 (2009) 14–23.
- [39] X. Wu, D. Wayeyey, Ge, H. Tang, Coagulation of silica microspheres with hydrolyzed Al(III)—Significance of Al13 and Al13 aggregates, *Colloids Surf., A* 330 (2008) 72–79.
- [40] A.A.A. Aziz, D.R. Dixon, S.P. Usher, P.J. Scales, Electrically enhanced dewatering (EED) of particulate suspensions, *Colloids Surf., A* 290 (2006) 194–205.
- [41] M. Bob, H.W. Walker, Enhanced adsorption of natural organic matter on calcium carbonate particles through surface charge modification, *Colloids Surf., A* 191 (2001) 17–25.
- [42] M. Ishiguro, W. Tan, L.K. Koopal, Binding of cationic surfactants to humic substances, *Colloids Surf., A* 306 (2007) 29–39.
- [43] M.J. McGuire, J. Addai-Mensah, K.E. Bremmell, The effect of polymer structure type, pH and shear on the interfacial chemistry, rheology and dewaterability of model iron oxide dispersions, *Colloids Surf., A* 275 (2006) 153–160.
- [44] Z. Chen, B. Fan, X. Peng, Z. Zhang, J. Fan, Z. Luan, Evaluation of Al30 polynuclear species in polyaluminum solutions as coagulant for water treatment, *Chemosphere* 64 (2006) 912–918.
- [45] M. Dozier, M.L. McFarland, in: *Drinking Water Standards*, 2006.
- [46] C.L. de Vasconcelos, M.R. Pereira, J.L.C. Fonseca, Polyelectrolytes in solution and the stabilization of colloids, *J. Dispersion Sci. Technol.* 26 (2005) 59–70.
- [47] T.K. Trinh, L.S. Kang, Response surface methodological approach to optimize the coagulation-flocculation process in drinking water treatment, *Chem. Eng. Res. Des.* 89 (2011) 1126–1135.
- [48] J. Gregory, J. Duan, Hydrolyzing metal salts as coagulants, *Pure Appl. Chem.* 73 (2001) 2017–2026.
- [49] J. Shin, R. Spinette, C. O'Melia, Stoichiometry of coagulation revisited, *Environ. Sci. Technol.* 42 (2008) 2582–2589.
- [50] W.J. Snodgrass, M.M. Clark, C.R. O'Melia, Particle formation and growth in dilute aluminum (III) solutions: characterization of particle size distributions at pH 5.5, *Water Res.* 18 (1984) 479–488.
- [51] W.W. Daniel, W.D. Wayne, in: *Biostatistics: A Foundation for Analysis in the Health Sciences*, Wiley, New York, 1978.
- [52] B.B. Lee, K.H. Choo, D. Chang, S.J. Choi, Optimizing the coagulant dose to control membrane fouling in combined coagulation/ultrafiltration systems for textile wastewater reclamation, *Chem. Eng. J.* 155 (2009) 101–107.