The characteristics of flocs and zeta potential in nano-TiO₂ system under different coagulation conditions

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HIGHLIGHTS

- Recovery factors with CaCl₂ were larger in TiO₂ system.
- Second addition of Ca²⁺ increased the floc size in TiO₂ system.
- Nano-Al₂O₃ decreased the fractal dimension in TiO₂ system.
- Zeta potential decreased in the breakage period.

GRAPHICAL ABSTRACT

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ABSTRACT

Three coagulants were used to investigate the characteristics of flocs formed in TiO₂ system, and the effects of humic acid (HA) on the coagulation process were also investigated. The results indicated that flocs could not regrow to former size in all coagulation conditions. The floc sizes after re-growth decreased with increasing the breakage period. Polyacrylamide (PAM) could significantly increase the floc size, and the fractal dimension decreased with increasing the concentration of N-PAM. Zeta potentials of flocs formed without N-PAM as coagulant decreased when the flocs were broken at 400 rpm. Zeta potential using N-PAM as flocculant did not significantly change during breakage period, and this maybe caused by the adsorption of N-PAM molecules on new exposed TiO₂ surfaces. The results of coagulation process using nano-Al₂O₃ + AS as coagulant showed that nano-Al₂O₃ particles could increase the floc size and fractal dimension in TiO₂ system, and the recovery factor was larger than only using AS. The further addition of Ca²⁺ and Al₂O₃ particles could enhance the floc size after regrowth. In the enhanced coagulation process (Al 1.0 mmol/L), PACI could produce larger flocs due to electrostatic patch effects.

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1. Introduction

More and more nano particles found in the aquatic environment caused more attentions [1]. Commercial TiO₂ particles were widely used in the paint and paper industries for their unique performance [2]. Coagulation has played an important role in water

treatment for decades [3,4]. Some researchers have applied poly-aluminum chloride (PACI) and cationic polyacrylamide to remove nanoparticles [5]. The breakage and re-growth of flocs have been studied by many researchers [6–8]. The floc size, strength and compaction may significantly affect the efficiency of solid/liquid separation [9]. The strength factors depends on floc structure, floc formation mechanisms and the inter-particle bonds. Floc strength could be measured by applying an increasing shear rate to the pre-formed flocs and relating the energy dissipation to the broken floc size [10].

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TiO₂ has caused more and more attentions in recent years due to its photo-catalytic effects. TiO₂ particles could cause degradation of organic compounds effectively with UV light. Some papers demonstrated the good antifouling properties of membranes with TiO₂. The application of nano-Al₂O₃ as adsorbents has become an interesting area of research due to their high removal efficiency. However, the effects of nano-Al₂O₃ particles on coagulation performance have not been paid enough attention. Nano-Al₂O₃ particles were added to the AS solution in order to understand the effects on the characteristics of flocs formed in the coagulation process, and the Al-based coagulants (AS and PACl) were used as traditional coagulants.

Humic acid (HA) has played an important role in the water treatment process. Numerous studies have shown the stabilizing impacts of humic acids on inorganic colloids due to the net negative surface charge [11,12]. The aggregation mechanisms of NPs may be significantly different in the presence of organic matters. Divalent cations were widely reported to enhance particle flocculation by facilitating development of bridges between anionic poly-electrolytes and negatively charged between Al and negatively charged colloids [13,14]. Chen et al. showed that Ca²⁺ strongly enhanced the aggregation rate of hematite NPs [15]. Polyacrylamide (PAM) is a polymer formed from acrylamide subunits [16,17]. Different volumes of PAM solution were added to the model water to measure the floc size and fractal dimension in the coagulation process.

The objective of this study was: (1) to investigate the characteristics of flocs formed in TiO₂ system; (2) to investigate the breakage rate and period on the characteristics of flocs; (3) to investigate the effects of HA molecules on the characteristics of flocs formed in TiO₂ system; (4) to investigate the effects of second addition of Ca²⁺ and Al₂O₃ on characteristics of flocs; (5) to investigate the effects of different volumes of PAM solution on characteristics of flocs.

2. Materials and methods

2.1. Methods and model water

All reagents used were of analytical grade. Deionized water was used to prepare all solutions. The stability of nano-TiO₂ suspensions was obtained through the addition of NaOH which generated anionic nano-TiO₂ surface. Particles would thus repel each other, providing a good stability. The model water was diluted to certain concentration (10.0 mg/L) with deionized water. The water samples were added 5.0 mM NaNO₃ and 4.0 mM NaHCO₃. The pH value of the solution was adjusted to 7.00 ± 0.03 by the addition of 0.1 M HCl and 0.1 M NaOH. When HA molecules were added to the model water, the water samples were stirred for 12.0 h for the equipment of HA adsorption on TiO₂ particles.

The zeta potentials of water samples were measured by Zetasizer 2000 (Mavlern Instruments, UK). PH was measured at room temperature with a pH-meter. Turbidity was measured with a turbidity meter. The size and fractal dimension analysis in the coagulation process were done by Mastersizer 2000 (Malvern Instrument, UK).

2.2. Coagulants and flocculants

Three coagulants were applied: Al₂(SO₄)₃·18H₂O (AS) was used as a metal salt, and PACl was used as pre-hydrolyzed metal salt. Nano-Al₂O₃ (100 mg/L) + AS was used to investigate the effects of nano Al₂O₃ particles on the characteristics of flocs. Nano-Al₂O₃ was added to AS solution and the solution was shaken well in order to disperse the composited systems as uniformly as possible before

Table 1

Al species for three coagulants used in this study.

	Al _a	Al _b	Al _c
PACl	31.33%	16.09%	52.58%
AS	93.78%	6.22%	0
Nano-Al ₂ O ₃ + AS	87.13%	11.23%	1.64%

dosing. Different PAM solutions were prepared by dissolving a certain amount of solid PAM (1.0 g/L) to de-ionized water.

The Al-species distribution was measured by Ferron method [18]. Total Al concentration (Al_T) for PACl was determined using ICP-OES. The results are summarized in Table 1.

2.3. Jar tests

Coagulation and flocculation experiments were conducted in 500 ml glass beakers using conventional Jar-test apparatus at room temperature.

A four-stage mixing process was conducted in this particle, including rapid mixing (200 rpm for 1.5 min) and slow mixing (40 rpm for 10 min) was followed for growth. After the breakage, the speed was reduced to 40 rpm to re-grow. Each experiment was repeated 10 times and the average zeta potential for every sample was determined based on the 10 measurements. The zeta potential of flocs after breakage by high stirring speed was also measured to investigate the effects of breakage on surface charges. The strength factor (*S_f*), recovery factor and fractal dimension were calculated [18,19] to better understand the mechanism. Zeta potential of flocs was measured in the coagulation process, and the breakage rate was 400 rpm.

3. Results and discussion

3.1. Characteristics of the model water

The concentration of TiO₂ in model water was 10.0 mg/L. The physico-chemical characteristics for the water samples were summarized in Table 2. It is generally considered that the zeta potential values greater than +30 mV or smaller than −30 mV, resulting in a stable suspension. The results showed that the zeta potential of TiO₂ suspension was −38.5 mV, and the addition of Al₂O₃, CaCl₂ and NaCl could increase the zeta potential. In the aquatic environments, chemical properties of the solution strongly influence the aggregation process. Previous studies have shown that electrolytes would promote the aggregation of nano materials [20–22].

3.2. Dosage optimum

Zeta potential and residual turbidity in TiO₂ system under different coagulant dosage (using AS) were measured. The results showed that the residual turbidity decreased dramatically and the zeta potentials remained negative but approached zero. When dosage was larger than 0.1 mmol/L of Al, the zeta potential was relatively constant with coagulant dosage. As dosage increased,

Table 2

Physico-chemical properties for the model water.

	Particle size <i>d</i> ₅₀ (μm)	Zeta potential (mV)
TiO ₂	1.004	−38.5
Nano-Al ₂ O ₃ (0.2 mg/L) + TiO ₂	1.423	−27.34
CaCl ₂ (6.0 mmol/L) + TiO ₂	2.257	−14.38
NaCl (6.0 mmol/L) + TiO ₂	1.814	−20.59

Fig. 1. The floc size in the coagulation process under different conditions (A: TiO_2 , B: $\text{TiO}_2 + 0.2 \text{ mg/L Al}_2\text{O}_3$, C: $\text{TiO}_2 + 6.0 \text{ mmol/L CaCl}_2$, D: $\text{TiO}_2 + 6.0 \text{ mmol/L NaCl}$).

the removal efficiency for turbidity reached a plateau and further increase of dosage would lead to a slight re-stabilization of particles, which was contributed to the charge reverse. Thus, 0.1 mmol/L was chosen as low dosage (close to 0 mV), and 1.0 mmol/L was chosen as high dosage for enhanced coagulation.

3.3. Flocs produced by AS ($\text{Al} - 0.1 \text{ mmol/L}$) in TiO_2 system

The results in Fig. 1 indicated that the floc size after re-growth decreased with increasing the breakage period and rate. However, flocs could not re-grow to former size in all conditions. The addition of CaCl_2 decreased the floc size from 600 to 550 μm due to charge reversal, but the recovery factors were larger (Table S1). Although the dosage of NaCl was the same as CaCl_2 (6.0 mmol/L), the bridging effect was smaller than CaCl_2 . The addition of 0.2 mg/L Al_2O_3 had no significant effects on the floc size, but it could increase the floc size after re-growth. The results in Fig. S1 showed that the fractal dimension after breakage increased with increasing the breakage period and breakage rate. The smaller flocs had larger fractal dimension. When nano- Al_2O_3 particles were added, the fractal dimensions decreased when the breakage rate was 400 rpm. When the breakage rate was 400 rpm, the bridges between TiO_2 particles formed by Al_2O_3 , CaCl_2 and NaCl were broken, so the fractal dimension decreased. The results in Table S1 showed that the

addition of Al_2O_3 , CaCl_2 and NaCl could increase the strength factor and recovery factor due to the bridging effects.

In order to better understand the effects of Ca^{2+} on the coagulation process, Ca^{2+} was added to the broken flocs as the second addition. The breakage rate was 200 rpm, and the breakage period was 5.0 min. The addition time was adjusted at the beginning of breakage and after 3.0 min. The results (Fig. 2) showed that the addition of Ca^{2+} could increase the floc size after re-growth due to the bridging effects. The results of fractal dimension indicated that the second addition of Ca^{2+} could increase the fractal dimension from 2.74 to 2.86 for flocs after breakage and 2.66 to 2.76 for flocs after re-growth. Ca^{2+} was adsorbed on the new surface exposed in the breakage period, and bridged the particles together. The results in Table S2 indicated that the second addition of Ca^{2+} (6.0 mmol/L) increased the strength factor from 36.16% to 40.00% and 45.00%, and the recovery factor was increased from 34.72% to 50.00% and 57.58%. Al_2O_3 particles were added to the solution to investigate the effects of Al_2O_3 on the flocs, and the results are summarized in Fig. 3 and Table S3. The results in Figs. 1A and 3 showed that the second addition of Al_2O_3 increased the floc size after re-growth. The results in Tables S1 and S3 showed that the strength factor and recovery factor were larger when Al_2O_3 was added before the addition of coagulant. When the Al_2O_3 was added firstly, the Al_2O_3 and TiO_2 could react absolutely,

Fig. 2. Floc size and fractal dimension using Ca²⁺ (6.0 mmol/L) as second addition.

so the strength and recovery factor were larger than the second addition.

3.4. Flocs produced by PACl and nano-Al₂O₃ + AS (Al – 0.1 mmol/L) in TiO₂ system

The results (Fig. 4, Table S4) showed that the recovery factor of flocs formed by PACl was larger than AS though the zeta potential of solution was positive. The fractal dimension of flocs formed by AS was larger. This could be explained that the flocs with positive charge would repel to each other, so the fractal dimension decreased. The results in Fig. S2(A) indicated that fractal dimension increased with increasing the breakage period and rate, and the smaller flocs had the larger fractal dimension. When nano-Al₂O₃ + AS was used as coagulant, the floc size was larger than using AS. The results of fractal dimension indicated that the addition of nano-Al₂O₃ would significantly decrease the fractal dimension when the breakage rate was 400 rpm. The results in Figs. 1(A) and 4(B) showed that the recovery factor of flocs using nano-Al₂O₃ + AS was larger. After breakage, the existence nano-Al₂O₃ particles could bridge the small flocs together.

Fig. 3. Floc size using Al₂O₃ (0.2 mg/L) as second addition.

Fig. 4. Floc size in coagulation process (A: PACl; B: nano- Al_2O_3 + AS).

3.5. Flocs produced by AS ($\text{Al} = 0.1 \text{ mmol/L}$) in TiO_2 –HA system

Many studies have reported on the stabilization of inorganic colloidal particles in presence of NOM [23,24,25,26]. Some researchers showed that organic anions increased the negative charge density adjacent to the mineral particle surface [27].

The results in Fig. 5 indicated that the addition of three materials could increase the floc size. The floc size after growth was $325 \mu\text{m}$, $350 \mu\text{m}$, $300 \mu\text{m}$, respectively. Results in Fig. 5(A) and (C) showed that Ca^{2+} could neutralize the repulsive negatively charged HA chain and formed bridges between adjacent molecules, and hence promoted the aggregation. During the breakage process, the newly exposed surfaces of aggregates could have a net negative, positive or neutral charge, which may depend on the property of the initial coagulation process. In the initial coagulation process, the charge neutralization ability of AS in HA- Ca^{2+} - TiO_2 water was stronger than that in HA- TiO_2 water. The fractal dimension in Fig. S3 showed that the addition of three materials increased the fractal dimension, and this may be caused by the increasing adsorption of HA on the surface of TiO_2 particles. When the breakage rate was large

(400 rpm), the bridging effects produced by HA molecules with TiO_2 particles were broken, and the fractal dimension decreased. The results in Table S5 indicated that the addition of three materials decreased the recovery factor due to the increasing adsorption of HA molecules on the particles.

3.6. Flocs produced by PACl and nano- Al_2O_3 + AS ($\text{Al} = 0.1 \text{ mmol/L}$) in TiO_2 –HA system

The results in Fig. S4 showed that nano- Al_2O_3 particles could increase floc size from 265 to $400 \mu\text{m}$. This could be explained by the bridging effects of nano particles. The nano particles adsorbing Al^{3+} could bridge the small flocs together quickly, and the flocs grow rate was larger than using AS. The bridging points of flocs were easily to be broken, so that the strength factor decreased. The results of fractal dimension (Fig. S5) indicated that fractal dimension decreased when the breakage rate was 400 rpm. The bridging effects formed by HA molecules were damaged after breakage, so the broken flocs had small fractal dimension. The results using PACl were similar.

Fig. 5. The floc size in coagulation process under different conditions (A: TiO_2 ; B: $\text{TiO}_2 + 0.2 \text{ mg/L Al}_2\text{O}_3$; C: $\text{TiO}_2 + 6.0 \text{ mmol/L CaCl}_2$; D: $\text{TiO}_2 + 6.0 \text{ mmol/L NaCl}$).

3.7. Flocs produced by AS ($\text{Al} - 0.1 \text{ mmol/L}$) and different PAM

Nonionic PAM (N-PAM), anionic PAM (A-PAM) or cationic PAM (C-PAM) was used in this study. These flocculant solutions were prepared by dissolving 1.0 g solid PAM in 1 L de-ionized water. The floc sizes with PAM were significantly larger than flocs coagulated by Al-based coagulant. In this study, four volumes of PAM solution were applied. The results in Fig. 6 indicated that the floc size increased with increasing the volume of PAM. Fractal dimension decreased from 2.52 to 2.13 with increasing the dosage using N-PAM as flocculant. When C-PAM and A-PAM were used as flocculant, the fractal dimensions at four dosages were almost the same (2.07). Because the zeta potential using AS (0.1 mmol/L) were close to 0 mV, the addition of C-PAM (with positive charges) and A-PAM (with negative charges) could decrease the floc size compared to N-PAM (without charges).

3.8. Zeta potential in the coagulation process

The results in Fig. 7 indicated that the zeta potential of flocs decreased when the flocs were broken in the nano- TiO_2 system and TiO_2 -HA system using AS. This may be caused by the new exposed surfaces which had $-\text{OH}$, $-\text{COOH}$ functional groups. When HA molecules were introduced to the system, zeta potential after re-aggregation was smaller than dis-aggregation. Zeta potential decreased from -35.2 to -39.3 mV , which was different from TiO_2 system. The HA molecules existed in solution could prevent the TiO_2 particles to aggregate together, so the zeta potential of

re-aggregation was smaller than dis-aggregation. When PACI was used, zeta potential (22 mV) was larger than using AS. This is due to the pre-hydrolyzed Al-species has larger charge ability. When N-PAM was used as flocculant, zeta potential of flocs after breakage had no significant differences, and this was caused by adsorption of N-PAM molecules on the new exposed surfaces of TiO_2 .

3.9. Enhanced coagulation ($\text{Al} - 1.0 \text{ mmol/L}$)

In order to investigate different characteristics of flocs under different coagulant dosages, larger amount of coagulant ($\text{Al} - 1.0 \text{ mmol/L}$) was added to the solution.

3.9.1. Without HA

The results in Fig. S6 showed that the larger amount of AS could produce smaller flocs ($400 \mu\text{m}$) due to charge reversal. The recovery factor (Table S6) decreased with increasing the dosage of coagulant, and this trend was more significant using PACI which had more positive charges. When PACI was used as coagulant, larger flocs ($835 \mu\text{m}$) were produced. These results may be caused by the different Al species in PACI and AS solutions. The pre-hydrolyzed Al species in PACI (Al_{13}) could be adsorbed on the surface of TiO_2 particles, and then it bridged the TiO_2 particles together.

3.9.2. With HA ($\text{DOC} 2.5 \text{ mg/L}$)

The results in Fig. S7 indicated that larger amount of PACI could produce larger flocs, but the floc size decreased within the coagulant process. These results proved that electrostatic patch effect

Fig. 6. Floc size and fractal dimension using different PAM as coagulant.

Fig. 7. Changes of zeta potential in the coagulant process under different conditions.

played a significant role in the coagulation process. The results of strength factor and recovery factor (Table S7) indicated that the HA molecules in the solution played a significant role in the recovery of flocs. The recovery factor of flocs decreased with increasing the dosage of coagulant.

4. Conclusion

Coagulation processes in TiO_2 system under different coagulation conditions were studied. Floc size, strength factor, recovery factor or fractal dimension were investigated. The main conclusions from this work were listed as follows:

- (1) The addition of CaCl_2 decreased the floc size due to charge reversal, but the recovery factors were larger than TiO_2 system. The fractal dimension after breakage increased with increasing the breakage period and rate in TiO_2 system. The further addition of Ca^{2+} increased the strength factor from 36.16% to 40.00% and 45.00%.
- (2) Nano- Al_2O_3 + AS increased the floc size. The addition of nano- Al_2O_3 decreased the fractal dimension when the breakage rate was 400 rpm.
- (3) When the breakage rate was 400 rpm, the HA molecules which were adsorbed on the surface of TiO_2 particles were desorption, and the fractal dimension decreased.
- (4) The zeta potential of flocs decreased using AS and PACI after breakage. The HA molecules existed in solution could prevent the TiO_2 particles to aggregate together.
- (5) The pre-hydrolyzed Al species in PACI (Al_{13}) could be adsorbed on the surfaces of TiO_2 particles, and then it bridged the TiO_2 particles together.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.colsurfa.2014.03.073>.

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