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Effect of cationic polyacrylamide with cationic microblock structure on dual-flocculant sludge conditioning: performance and mechanism

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ABSTRACT

In this study, sludge conditioned by single and dual flocculants was comprehensively investigated. polyaluminium chloride (PAC) was used as inorganic flocculant, whereas TP (cationic polyacrylamide [CPAM] with cationic microblock structure) and SP (CPAM with random distributed units) were used as organic flocculants. First, the effect of single and dual flocculant dosage on sludge conditioning performance was investigated using turbidity, filter cake moisture content (FCMC), and specific resistance of filtration. The effect of dosing sequence on sludge conditioning performance was also studied. Dual-flocculant conditioning of sludge with PAC and then with TP not only improved sludge dewaterability but also decreased flocculant dosage. Settling behavior was determined for studying the floc characteristics. The effect of charge neutralization on sludge conditioning was investigated by zeta potential. The mechanism of dual-flocculant conditioning mechanism was further summarized with the experimental results.

Keywords: Sludge conditioning; Flocculation mechanism; Dual-flocculant conditioning

1. Introduction

Given that activated sludge (AS) process is the most important treatment technology for a wide range of wastewaters, the large, annually increasing excess sludge, which represents 1% or 2% of treated wastewater, contains 50% to 80% of pollution that must be disposed of. Sludge contains various kinds of contaminants, such as heavy metals, pathogens, and organics, which can cause serious environment pollution. In addition, sludge is extremely difficult to dewater because of small negatively charged particles distributed evenly in the form of a stable colloidal suspension. For destabilizing the particulate system, sludge conditioning prior to mechanical dewatering is generally required [1–3]. Flocculation, a conventional sludge conditioning method, is advantageous particularly in sludge separation based on different particle weights. Numerous widely used inorganic and organic chemical flocculants with high conditioning performance and low cost play a vital role in sludge conditioning [4]. Good control of flocculant dose is critical in sludge conditioning because overdosing increases cost and reduces sludge dewaterability. The optimal polyelectrolyte dosage is usually associated with the colloidal surface of the minimum surface charge and a tendency to aggregate to form large flocs [5].

In recent years, cationic polyacrylamide (CPAM) has become one of the most commonly used polyelectrolytes for sludge conditioning because of its capacity to neutralize the surface charge of solid particles and bridge particles through its long polymer chains; these properties enable the formation

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of large flocs, which can reduce sludge specific resistance and diminish cake compressibility [6–8]. Therefore, molecular weight and charge density are two of the most important indexes for CPAM. Sludge conditioned with CPAM generates a less solid cake after sludge dewatering because of the lower required dosage compared with fly ash and inorganic chemicals. The cost of the polymer accounts for almost half of the overall sludge dewatering and disposal costs. Therefore, effective sludge conditioning is important in sludge dewatering. However, CPAM is considerably more expensive than inorganic polymers.

Inorganic flocculants exhibit their own characteristics and advantages in sludge conditioning. First, inorganic coagulants act as skeleton builders and reduce solid compressibility. Therefore, these materials are particularly beneficial in high-pressure dewatering, such as that employing a filter press, because the high porosity of the solids cake needs to be maintained under high pressure [9]. Second, coagulants exhibit excellent charge neutralization performance owing to their high charge density. Furthermore, most soluble extracellular polymeric substances (S-EPS) with high molecular weight (>2,000 Da), which influences sludge dewatering performance, can be effectively removed after inorganic flocculant conditioning [10,11]. Finally, the low price of inorganic flocculants reduces sludge treatment cost.

Thus, we can reasonably infer that sludge conditioned with dual chemicals may yield excellent conditioning performance with effectively reduced treatment cost. Since the end of the 20th century, dual-chemical conditioning has become an important technique for conditioning and dewatering sludge. Chitikela and Dentel [12] experimentally determined the differences in the effects of single- and dual-chemical conditioning of anaerobically digested sludge. The optimal dosage of each additive in dual-chemical conditioning is significantly lower compared with that of each chemical used individually. Chen and Wang [13] used polymeric ferric sulfate (PFS) and flocculants in single- and dual-chemical conditioning to treat AS. Inorganic flocculant conditioning facilitates the compactness of AS flocs/aggregates. The PFS hydrogel and CPAM molecules can serve as backbones of the newly formed AS flocs/aggregates [13]. Peeters et al. [14] indicated that the effect of the dual-chemical conditioning on the efficiency of sludge treatment results in an increasingly open sludge structure when this complex interacts with sludge particles, hence facilitating the dewatering and drying steps. Even with molecular weight and charge density, the sequence distribution of organic flocculants affects flocculation mechanism and performance. However, the polyelectrolytes used in the aforementioned studies were all randomly distributed.

CPAM, which possesses a cationic block structure, can flocculate suspended particles more efficiently than random distributed particles [15–17] owing to its capability to strengthen adsorption sites between copolymer and negatively charged particle segments and facilitate the efficient use of cationic charges. Thus, flocculation performance increased [18–21]. In our previous research [22], template copolymers (TP) with cationic block structure were prepared through template polymerization. TP flocculation performance and mechanism in kaolin suspension were investigated in comparison with SP (polymer with random distribution prepared through solution polymerization). For sludge conditioning, Zheng et al. [23] found that the cationic microblock structure strongly enhanced charge neutralization, patching, and bridging, thus improving the AS flocculation performance. However, no previous study has investigated the effect of CPAM with cationic microblock structure on dual-coagulation sludge conditioning.

In this study, polyaluminium chloride (PAC) was used as inorganic flocculant, and TP and SP were used as organic flocculants. The effect of dosage on sludge conditioning performance was extensively investigated through turbidity, filter cake moisture content (FCMC), and specific resistance of filtration (SRF) studies. Floc characteristics were investigated by settling performance. The mechanism of charge neutralization was evaluated by zeta potential. The conditioning mechanisms were analyzed.

2. Materials and methods

2.1. Materials

TP and SP were prepared by a method used by Guan et al. [22]. The characteristics of the copolymers can be seen in Table 1. PAC was bought from Baolai Water Treatment Material Plant (Gongyi, China).

Raw waste sludge from the thickener of Jiangning Drainage Co., Ltd. (Nanjing, China) was used for this study. The designed daily sewage treatment capacity was 80,000 cubic meters and Oble oxidation ditch process was applied as the main treatment technology. The samples after collection were stored in a refrigerator maintained at 4°C to minimize the microbial activity and analyzed within 2 d. The characteristics of the sludge are listed in Table 2.

2.2. Sludge conditioning at lab scale

Buchner funnel test, the most common method for dewatering ability measurement, was used to investigate the conditioning ability of the polymers. Qualitative filter paper was used in Buchner test. A certain dosage of one flocculant was added into 200 mL sludge at a stirring of 200 rpm for 0.5 min. And the other kind of flocculant was then added into

Table 1
Characteristics of TP and SP

Flocculant	Intrinsic viscosity (dL/g)	Charge density (%, m/m)
TP	11.42	25%
SP	11.68	25%

Table 2
Characteristics of raw waste sludge

Moisture content (%)	98.9 ± 0.15
SRF (m/kg)	52.6×10^{12}
Mass density (mL/g)	1.021
pH	7.41 ± 0.12
Zeta potential (mV)	–18.6 ± 0.4
Appearance	Dark gray

at a stirring of 200 rpm for another 0.5 min. After 10 min settling period, turbidity sample was collected at 1 cm below the supernatant surface (TB-2000, Sinsche, China). Zeta potential was measured by Zetasizer Nano ZS90 (Britain) and the procedure was the same with that was conducted by Guan et al. [22] and Thapa et al. [24]. A specific amount of conditioned sludge was transferred immediately to the laser particle size analyzer dispersion unit to determine the floc size distribution (Winner 2000 laser particle size analyzer, Jinan Winner Particle Technology Co., Ltd., Jinan, China). The sample under investigation was gently stirred using the stirrer integrated in the dispersion unit to avoid agglomeration of flocs. The dispersion force in the unit was significantly milder compared with that used during the flocculation experiments; therefore, the integrity of the flocs was not affected. The average size of flocs was automatically recorded by computer.

The conditioned sludge was poured into a Buchner funnel to filter under a vacuum pressure of 0.05 MPa for 30 min or until the vacuum could not be maintained (in <30 min). The filterability of the sludge is measured by Eq. (1):

$$\mathrm{SRF} = \frac{2bPA^2}{\mu\omega} \quad (1)$$

where SRF is the specific resistance of the sludge (m/kg); P is the filtration pressure (N/m^2); A is the filter area (m^2); μ is the viscosity of the filtrate (Ns/m^2); ω is the weight of cake solids per unit volume of filtrate (kg/m^3, $\omega = (1/C_i)/((100C_i - C_f)/100C_f)$; C_i is the initial moisture content (%); C_f is the final moisture content (%); b is the slope of filtrate discharge curve (t/V vs. V) (s/m^6), where t is the filtration time (s); and V is the volume of the filtrate [25].

FCMC (%) of the conditioned sludge was determined using Eq. (2):

$$\mathrm{FCMC} = \frac{M_T - M_f}{M_T} \quad (2)$$

where M_T is the weight of filter cake after filtration (g) and M_f is the weight of filter cake after drying at 105°C. The experiment was repeated three times and average results were reported.

The floc settling rates were determined in a graduated cylinder with a 1,000 mL sludge sample. The total height of the graduated cylinder is 30.0 cm. After rapid agitation, the sludge suspension was transferred into a graduated cylinder and precipitated without disturbance. The height of the sludge–liquid interface corresponding time was recorded within 45 min. The settling rate of the sludge flocs was calculated in terms of the height of the sludge–liquid interface as a function of settling time in the first 2 min.

3. Results and discussion

The effect of any flocculant on flocculation is influenced by dosage; flocculation efficiency decreases at decreased or increased dosages relative to the optimum values. Therefore, dosage was selected as the variable. Turbidity, an indication of suspended particle content in the liquid phase, is widely used to explain the performance of a polymer sample in various flocculation fields. The water content of sludge cake is an important aspect to be considered in dewatering compressible sludge. The high moisture content of the filter cake is not desirable because of the large volume of generated sludge, with increases followed by increasing treatment costs. SRF is widely used to evaluate sludge filtration performance, and the decreased specific resistance to filtration reflected improved sludge dewatering. In this study, residual turbidity, FCMC, and SRF were used to evaluate polymer sludge dewatering performance at various dosages. Settling performance was used to characterize floc size and floc compactness. Zeta potential is a useful index for evaluating the charge neutralization performance of the flocculant. The conditioning mechanism of dual flocculant was analyzed through settling performance and zeta potential [26].

3.1. Conditioning performance of single-flocculant sludge conditioning performance

3.1.1. Effect of PAC dosage on sludge conditioning performance

The effect of single PAC dosage on supernatant turbidity, FCMC, and SRF after sludge conditioning is shown in Fig. 1. Residual turbidity, FCMC, and SRF markedly decreased with increasing dosage (Fig. 1). The negative surface charge of sludge particles can be eliminated, and interparticle bridging can occur through PAC addition, resulting in particle destablization and aggregation. Large flocs can be formed, and residual turbidity decreased. Decreased SRF is due to formed flocs facilitating sludge dewatering because water between flocs are easily eliminated. With PAC addition, water can be stripped from aggregates by double electric-layer compression, and water trapped in EPS can be released into sludge bulk solution [11]. Therefore, the FCMC of conditioned sludge decreased. However, after reaching the minimum values at a dosage of 500 mg/L, all three parameters decreased slightly or remained constant. Deterioration of sludge dewaterability was observed with organic flocculant dosing; organic flocculants were overdosed due to the presence of unabsorbed polyelectrolyte in the aqueous phase, which consequently increased the sludge viscosity [27]. However, overdose did not occur with continued increase

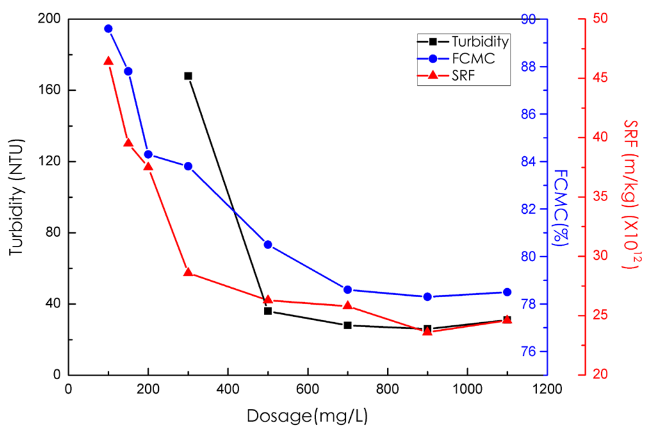

Fig. 1. Effect of PAC dosage on supernatant turbidity, FCMC, and SRF after sludge conditioning.

in PAC dosage because the intrinsic viscosity of PAC was markedly lower than that of the organic flocculants. Residual turbidity, FCMC, and SRF were 28 NTU, 78.6%, and 25.8 × 10^{12} m/kg, respectively, at a PAC dosage of 700 mg/L, indicating poor conditioning performance.

3.1.2. Effect of SP and TP dosage on sludge conditioning performance

Figs. 2 and 3 show the effect of SP and TP dosage on supernatant turbidity, FCMC, and SRF after sludge conditioning,

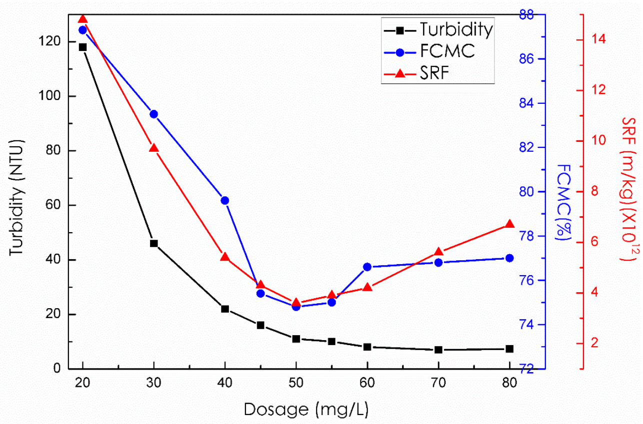

Fig. 2. Effect of SP dosage on sludge conditioning performance.

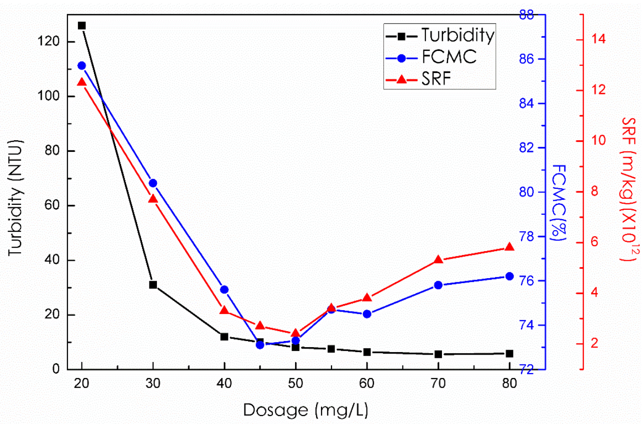

Fig. 3. Effect of TP dosage on supernatant turbidity, FCMC, and SRF after sludge conditioning.

respectively. A sharp residual downtrend with increasing polycation dose was observed, reaching a minimum and then remaining constant for both SP and TP. The optimum dosages at which the lowest turbidity was achieved were about 50 and 40 mg/L for SP and TP, respectively. As indicated by our previous study [26], residual turbidity was closely related to flocculant charge neutralization performance. Therefore, we can conclude that TP showed superior charge neutralization performance to SP. The FCMC and SRF curve shape of TP and SP was similar. As shown in Figs. 2 and 3, FCMC and SRF decreased significantly with increasing polymer dosage, and the lowest FCMC and SRF were attained at the dosages of about 45–55 mg/L for TP and SP, respectively. Restabilization occurred after the optimal dosage was reached, and FCMC and SRF increased with increasing polymer dosage. In Table 3, the FCMC and SRF of TP are lower than those of SP at optimum dosages, meaning that sludge conditioned by TP is less compressible and easier dewatered compared with sludge conditioned by other flocculants. The sequence distribution of cationic monomers explained the varying flocculation performance. TP neutralized the particle surface more effectively than SP, and denser flocs can be formed with TP. Polymers in solution are coiled in shape [28], and chain expansion can be induced through repulsion between charged segments. Repulsion forces among cationic microblock segments should exceed those among random cationic segments. Therefore, the linear molecular chains of TP favored more significant bridging than those of SP. In consequence, denser, larger flocs can be formed through superior charge neutralization, bridging performance, better sludge conditioning performance of TP than that of SP.

3.2. Dual-flocculant sludge conditioning performance

3.2.1. Effect of PAC dosage on dual-flocculant sludge conditioning performance

Sludge was first conditioned with controllable PAC dosage, followed by a fixed SP or TP dosage of 45 mg/L. The effect of PAC dosage on dual-flocculant sludge conditioning performance is shown in Figs. 4 and 5. The optimum dosages and corresponding conditioning performance are summarized in Table 3. Evidently, the curve shape of Figs. 4 and 5 is similar with those in Figs. 2 and 3, respectively, instead of that in Fig. 1, with the three values decreasing significantly in Figs. 2 and 3 but remaining unchanged in Figs. 4 and 5. This outcome may be due to PAC not being the main control

Table 3
Optimum dosages of flocculants and corresponding conditioning performance

Flocculant	Optimum dosage (mg/L)	Residual turbidity (NTU)	FCMC (%)	SRF (m/kg) (×10^{12})
Single PAC	700	28	78.6	25.8
Single SP	50	11	74.8	3.6
Single TP	45	10	73.1	2.7
PAC–SP (45 mg/L)	300	11.6	73.5	3.1
PAC–TP (45 mg/L)	300	6.5	70.3	0.6
PAC (300 mg/L)–SP	40	11.6	72.6	2.1
PAC (300 mg/L)–TP	35	6.8	70.6	0.8

factor for sludge conditioning performance. By contrast, the optimum PAC dosage was 300 mg/L for both dual-flocculant systems, with residual turbidity, FCMC, and SRF of dual-flocculant conditioning being lower than those of single PAC conditioning. Sludge conditioning performance was already acceptable with the addition of the organic flocculant. In single PAC conditioning, the process mostly relies on charge neutralization mechanism, the bridging mechanism cannot function well, and the molecular weight of PAC is notably lower than that of organic polymers. Bridging performance can be promoted by the addition of TP or SP. Thus, sludge dewatering performance was enhanced in dual-flocculant conditioning.

3.2.2. Effect of SP and TP dosage on dual-flocculant sludge conditioning performance

Sludge was first conditioned with a fixed dosage of PAC and controllable SP or TP dosage. Figs. 6 and 7, respectively, show the effect of SP and TP dosage on the sludge conditioning performance of dual-flocculant treatment. The optimum dosages and corresponding conditioning performance are summarized in Table 3. The curved shape in Figs. 6 and 7 is similar to that in Figs. 2 and 3. Residual turbidity, FCMC, and SRF all decreased significantly with increasing polycation dose from 20 to 40 mg/L. These phenomena indicated that organic flocculants play a vital role in dual-flocculant conditioning. Notably, not only the optimum dosages of SP and TP were decreased; conditioning performance was superior to that of single SP and TP conditioning. As mentioned above, PAC conditioning of sludge mainly occurred through charge neutralization mechanism. One advantage of PAC is the considerably lower molecular weight in comparison with organic polymers. Charge neutralization and double electric-layer compression can be conducted more sufficiently compared with organic polymers. Therefore, in dual-flocculant conditioning, primary flocs were already formed through PAC charge neutralization, and the positive charges needed from SP or TP can diminish. Therefore, all optimum dosages decreased. Moreover, dense, small flocs can form with the addition of PAC, and the bridging mechanism of SP and TP can function completely. Dense, large flocs were subsequently formed, and decreased FCMC and SRF was observed.

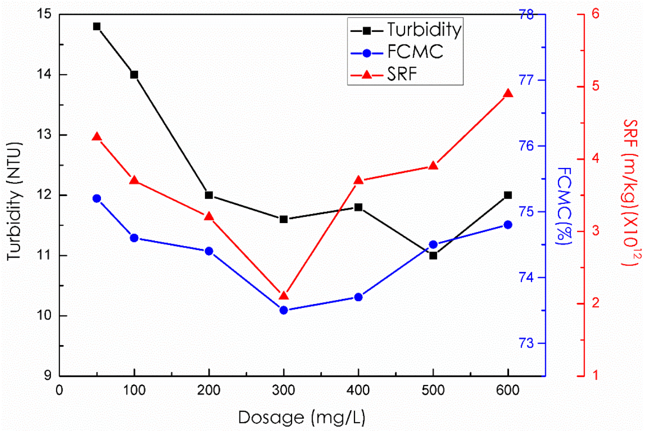

Fig. 4. Effect of PAC dosage on supernatant turbidity, FCMC, and SRF after PAC–SP (45 mg/L) sludge conditioning.

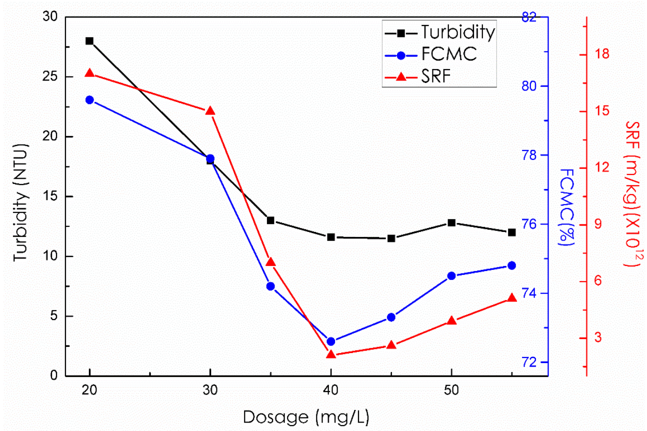

Fig. 6. Effect of SP dosage on supernatant turbidity, FCMC, and SRF after PAC (300 mg/L)–SP sludge conditioning.

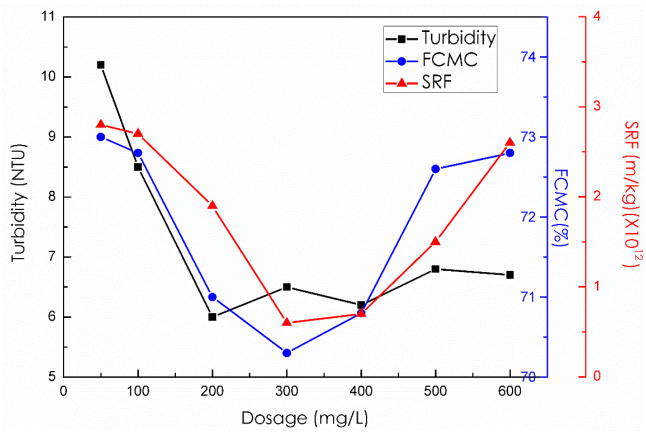

Fig. 5. Effect of PAC dosage on supernatant turbidity, FCMC, and SRF after PAC–TP (45 mg/L) sludge conditioning.

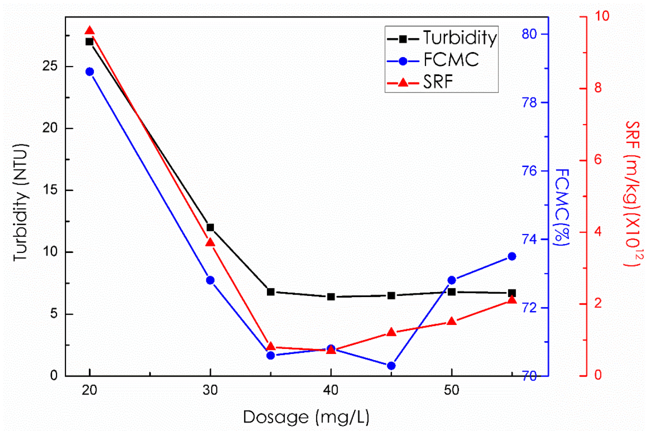

Fig. 7. Effect of TP dosage on supernatant turbidity, FCMC, and SRF after PAC (300 mg/L)–TP sludge conditioning.

3.2.3. Effect of dosing sequence on sludge conditioning performance

Previous studies recorded that dosing sequence affects sludge conditioning performance [5]. Therefore, we investigated the performance of sludge preconditioning with SP or TP and then with PAC (Table 4). In Tables 3 and 4, sludge preconditioned with organic polymers show poorer conditioning performance than that preconditioned with PAC because the advantages of both organic and inorganic flocculants cannot contribute to flocculation. Large flocs had already formed with charge neutralization and bridging mechanism when sludge was preconditioned with organic flocculants. At such times, PAC did not show excellent bridging capacity and cannot promote the formation of large, dense flocs. Therefore, the sludge conditioning performance was unacceptable. In this study, the flocculation mechanism of sludge preconditioned with organic flocculant was not explored.

3.3. Dual-flocculants conditioning mechanism

3.3.1. Settling behavior

Sludge settling behaviors are widely used to evaluate the flocculation performance and sludge dewatering capacity of flocculants. In general, the fast settling rate and large size of flocs indicate good dewatering capability [29]. In this study, the settling behaviors of flocs conditioned with various flocculants were investigated in terms of settling rate and condensed sludge floc volume (Fig. 8).

Table 4
Residual turbidity, FCMC, and SRF after sludge conditioned with SP (40 mg/L)–PAC (300 mg/L) and TP (40 mg/L)–PAC (300 mg/L)

Flocculant	Residual turbidity (NTU)	FCMC (%)	SRF (m/kg) ($\times10^{12}$)
SP (40 mg/L)–PAC (300 mg/L)	16	75.3	4.8
TP (40 mg/L)–PAC (300 mg/L)	11	74.7	3.1

In Fig. 8, sludge preconditioned with PAC and then TP resulted in the best ultimate sediment height and the corresponding settling rate among the flocculants, and the order was as follows: PAC (300 mg/L)–TP (35 mg/L) (15.0 cm; 2.85 cm/min) > PAC (300 mg/L)–TP (40 mg/L) (16.6 cm; 1.80 cm/min) > TP (45 mg/L) (18.4 cm; 1.25 cm/min) > SP (50 mg/L) (19.3 cm; 1.15 cm/min) > PAC (700 mg/L) (20.3 cm; 0.90 cm/min). First, PAC yielded the largest condensed sludge floc volume. Floc density and floc diameter exert a certain effect on settling behaviors. However, floc size is primarily important in determining settling rate. Sludge conditioned with organic flocculants resulted in the formation of relatively large flocs owing to improved bridging performance. By contrast, relatively small, dense flocs were formed for PAC through charge neutralization and double electric-layer compression [11], thereby resulting in the occurrence of the above phenomena. Second, the settling performance and condensed sludge floc volume of dual-flocculant conditioning were superior to those of single-flocculant conditioning. This result is closely related to conditioning mechanism, which can be illustrated through understanding of zeta potential.

3.3.2. Zeta potential

The effects of PAC and SP and TP dosages on zeta potential are shown in Figs. 9 and 10, respectively. In Fig. 9, the zeta potential of single PAC conditioning did not reach 0 mV in the dosage range, indicating that the negative surface of sludge particles cannot be completely neutralized. In addition, Watanabe et al. [30] reported that negative segments of suspended particles were only partially neutralized even at the zeta potential of 0 mV. However, the zeta potentials of TP and SP reached 0 mV when TP and SP dosages of 56.5 and 58.5 mg/L, respectively. The increasing trend of organic flocculant zeta potential continued with further increase in dosage. This trend cannot imply that the charge neutralization of TP and SP was superior to that of PAC because bridging can also contribute to the increasing zeta potential for flocculants with high molecular weight. Von Homeyer et al. [31] reported similar findings. The curved shape of the dual-flocculant zeta potential was similar to those of SP and TP. Upon the addition

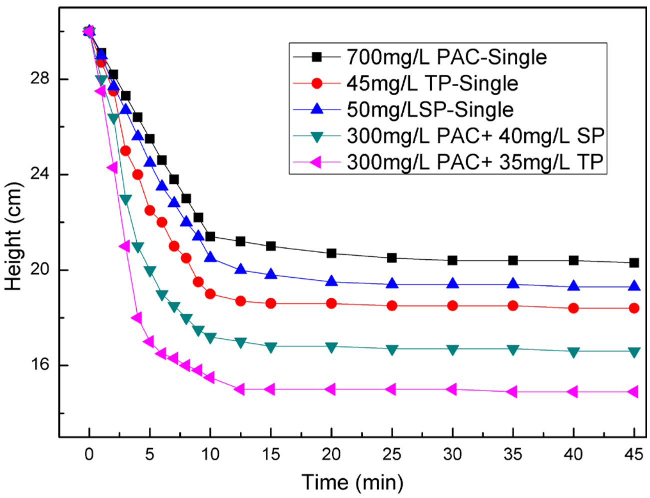

Fig. 8. Flocs interface heights of the flocculants.

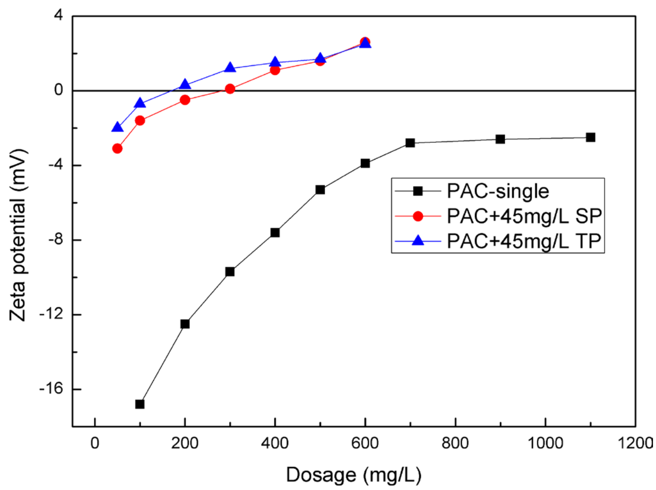

Fig. 9. Effect of PAC dosage on zeta potential.

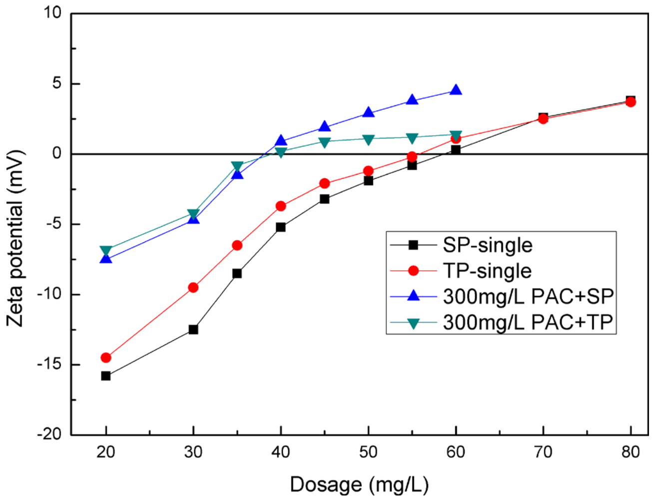

Fig. 10. Effect of SP and TP dosage on zeta potential.

of 180 and 280 mg/L PAC in dual-flocculant conditioning, the zeta potentials of PAC-SP and PAC-TP reached zero (Fig. 9). Isoelectric dosages where zeta potential reached 0 mV were about 38.6 mg/L for SP and 39.1 mV for TP, which were all near the optimum dosages in Table 4. These results indicated that after primary flocs formed through PAC addition, electrostatic force strongly contributed to the formation of large, dense flocs in dual-flocculant conditioning. The detailed conditioning mechanism can be illustrated as follows.

3.3.3. Sludge conditioning mechanism

In single SP and TP conditioning, a bridging mechanism played a major role in forming large particles. The zeta potential of TP was higher than that of SP, indicating that charge neutralization also contributed to conditioning and that TP showed superior conditioning performance. PAC sludge conditioning mainly relied on charge neutralization and double electric-layer compression. In dual-flocculant treatment, flocs formed by PAC addition were small, dense, and negatively charged. In addition, water can be stripped from aggregates and EPS, and free water content can be increased, thus aiding mechanical dewatering. After the addition of SP and TP, electrostatic force formed between flocs and the organic flocculant. Therefore, large and dense flocs can be formed, and the conditioning performance can be promoted. Large electrostatic force was observed between flocs and TP owing to the cationic microblocks in the TP chain. Thus, the performance of PAC–TP dual conditioning was superior to that of PAC–SP.

4. Conclusion

Sludge conditioning by single and dual flocculants was conducted in this study. In dual flocculant conditioning, not only sludge dewaterability was improved but also flocculant dosage was decreased. Moreover, sludge preconditioned with PAC was superior to that preconditioned with TP or SP. Settling behavior proved that flocs formed by PAC addition was small but dense, whereas those formed by TP and SP were large but loose. The zeta potential of PAC indicated that flocs remained negatively charged after PAC conditioning. The electrostatic force between formed flocs and the organic flocculant is the driving force of large and dense flocs. Thus, TP, a flocculant with cationic microblocks, showed superior performance in dual-flocculant sludge conditioning owing to its larger electrostatic force than that of SP.

Acknowledgments

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