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Influences of Various Aluminum Coagulants on Algae Floc Structure, Strength and Flotation Effect

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***Abstract*—Floc structure and strength play important roles in particles separation in water treatment. In a jar test of coagulation/flocculation/dissolved air flotation (C/F/DAF) process, aluminium sulphate (AS) and polyaluminium chloride (PAC) were used as coagulants to treat the algal-rich water which had a high concentration of *Microcysits aeruginosa* (MA) cells. Two-dimension fractal dimension (D_2) and strength factor (*SF*) were used to represent the floc structure and strength respectively. The experimental results showed: meeting the appropriate dose range (measured in Al^{3+}) simultaneously, dose of PAC was less than AS; and treatment effect obtained by PAC was better than AS. Adding the moderate PAC, the algae flocs with highly branched structure and higher strength formed by bridging flocculation; this kind of algae flocs could firmly adhere to more tiny bubbles and be significantly more resistant to the hydraulic shear, so the performance of flotation separation using PAC could be improved compared with adding AS.**

Keywords- Cogulation/flocculation/dissolved air flotation (C/F/DAF); Polyaluminium Chloride (PAC); Aluminium Sulphate(AS); Floc Structure; Floc Strength

I. INTRODUCTION

It has been proved that coagulation/flocculation/DAF (dissolved air flotation) is an efficient and practical process in water treatment to remove suspended particles and colloidal impurities. Since the separation of algae from water is under intensive study recently, C/F/DAF has been receiving more and more attention. The process is generally more effective than coagulation/flocculation/sedimentation for treating algal-rich water [1-3].

Coagulation/flocculation (C/F) prior to DAF can destabilize negative algal cells and flocculate small particles into larger flocs which can more easily adhere to tiny bubbles [4]. In the C/F process, the selection and dose of coagulant, as well as operating conditions (the velocity gradient and time), are critical for better overall treatment efficiency. Coagulants such as aluminium sulphate, ferric sulphate, ferric chloride and polymerised coagulants as polyaluminium chloride have been successfully used for treating algal-rich waters in the C/F/DAF. Using alum and PAC as coagulants, over 80% of 18 types of algae can be removed from water by DAF. It has been found that both alum and $FeCl_3$ are effectivein removing *Chlorella vulgaris* from water, and aluminum coagulants have some advantages over other coagulants in the removal of *Microcystis aeruginosa* and *Chlorella vulgaris* [5].

Flocs, generated in C/F process, have been shown to be fractal structure [6], which has been considered that the flocs properties, such as form and density. Fractal dimensions can be described as one of the significant properties of fractal aggregates. In addition to fractal structure, floc strength is another important parameter to describe floc properties, and it is dependent upon the inter-particle ponds between the components of the floc [7]. Furthermore, the properties of floc, such as form and strength, are very important impact factors in C/F/DAF for the efficient removal of aggregated particles [8].

The aim of this paper is to investigate the impacts of two types of aluminum coagulants on algae-floc structure and strength, and the influences of various aluminum coagulants on floation effect are analysed from microcosmic viewpoint.

II. MATERIALS AND METHODS

Coagulants

The aluminum coagulants used in the experiment were aluminium sulphate (or AS, $(Al_2(SO_4)_3 \cdot 18H_2O$, analytic reagent) and polyaluminium chloride (or PAC, industrial product). PAC is a pre-polymerised aluminium coagulant with 28% Al_2O_3 and a relative basicity of 65~85%.

Each coagulant was made up into 10mg/L solutions with deionized water. The adding dose was measured in Al^{3+} (mmol/L).

Raw Water

The raw water was algal-rich water by using tap water to dilute the high concentration suspension contained *Microcysits aeruginosa* (short for MA). MA cells were cultured in laboratory by BG11 medium at 23~24 ◦C under a light regimen of 12 h light and 12 h dark. The quality of raw water is presented in Table 1.

Table 1. Quality of the raw water quality

Water Quality Index	Algal amount ($\times 10^9$cells/L)	Turbidity (NTU)	pH	Chromaticity (PCU)
Range	2.57~3.45	5.2～5.8	8.0~8.4	17.69~18.28

Experimental Equipments

The experimental equipment was comprised of two same reactors, a coagulation instrument and a set of dissolved air system. The reactor was a test jar made of transparent organic glass, the dimensions of which were 100×100×300 mm. A sample hole and a dissolved-air water inlet were installed at the bottom of the reactor. A paddle was installed in the reactor, and a rapid (coagulation) and a slow (flocculation) stirring were controlled by the coagulation instrument. So coagulation, flocculation and flotation separation were all performed in the reactor.

The dissolved air system consisted of an air compressor, a dissolved air tank, a high-pressure pump and a dissolved air water releaser.

Experimental Procedures

- Coagulation and flocculation experiments were simultaneously performed in the same two reactors. The experimental procedures were: (1) the first was the setting of mixing procedures: coagulation was at G-value (value of hydrodynamic velocity gradient) of 500 s^{-1} (correspondingly the turning rate of paddle was at 250rpm) for 1 min, and then flocculation was at G-value of 70 s^{-1} (50rpm) for 8 min. (2) 2 liters of raw water was respectively poured in each reactor; (3) the same dose of coagulant was added in each one by pipet at the beginning of rapid mixing.
- Flotation separation experiment was processing immediately in one of the two reactors after flocculation operation. The experimental procedures were: (1) 200ml dissolved-air water (a recycle ration of 10%) was immediately introduce into the reactor from the inlet for 2min, meanwhile, the paddle was slowly stirring at 20rpm in order to make flocs contact with and adhere to tiny bubbles adequately; (2) flotation separation was performed on standing for 5min after release of dissolved air water; (3) finally, clarified water sample was collected from the sample hole to measure water quality.
- Algae flocs were formed at the addition of two kinds of coagulants in a series of jar test to examine the flocs form and strength. Flocs samples were collected from the other reactor. The experimental procedures were: (1) having slowly drawn the flocculated water by a sampler with a wide mouth, the sample was diluted by ultra-pure water in a small glass channel (40×15×2 mm) for avoiding drying crack and gathering of flocs, noticing that the act of sampling was cautious and slow so as not to break up the flocs; (2) after the first sampling was finished, the mixing rate of paddle was immediately adjusted to 150 rpm (G-value of 107s^{-1}) for 1 min to break the flocs formed in flocculation into pieces. (3) the second sampling was repeated to collect the cracked flocs in accordance with the first step; (4) the fractal dimension and strength factor of floc were measured with photomicrography and image manipulation software.

Analytical Methods

- The water samples were analyzed for turbidity, chromaticity and A_{440nm} (absorbance at the light wavelength of 440 nm). Turbidity was measured by a HACH 2100N turbidity meter of high resolution (0.001 NTU). A_{440nm} was measured by absorbance method using a HACH DR-4000 spectrophotometer. Absorption peak of algal-rich water occurred at 440 nm by wavelength scan, so A_{440nm} can indirectly reflect the algal level in synthetic water.
- The pictures of flocs were filmed by Olympus photomicrography system, and then they were analyzed by image processing software (Image Pro Plus).
- Two-dimension fractal dimension was used to describe floc form. The two-dimension fractal dimension is determined by a power law relation between projected area (A) and the characteristic length (l) of the aggregate, and the relationship between them is [9]:

$$A \propto l^{D_2} \qquad (1)$$

where D_2 is the two-dimensional fractal dimension, and the perimeter (P) of floc image indicated the characteristic length. The D_2 value was calculated by regression analysis of the logarithm of A versus the logarithm of P as showed by Eq. (1). The lower D_2 value results from the large, highly branched and loosely bound structure, while densely packed flocs have a high fractal dimension.

- In order to compare flocs of different strengths obtained by adding various coagulants, strength factor (SF) was used to represent floc strength. SF was determined by exposing floc to single level of increased shear within a containing vessel and comparing the ratio of the floc size before and after breakage [7]. The theoretical value of floc strength can be calculated by Eq. (2): where, d_1 was the average size of the flocs formed by flocculating reaction, and d_2 was that of the smaller broken flocs generated under shear-induced breakage; floc size was indicated with equivalent-area circle diameter of floc image. The higher SF value is, the stronger floc strength is.

$$SF = \frac{d_2}{d_1} \times 100 \qquad (2)$$

III. Results and Discussion

A. Coagulation/flocculation/DAF Experiments

Adding two types of aluminum coagulants, variation trends of treatment effects with dose were shown in Fig. 1 and Fig. 2. The two curves in each figure showed the similar tendency that the removal rates increased first and then decreased with increase of dose. The dose range of 0.08~0.14 mmol Al^{3+}/L was appropriate with PAC addition, and the treatment effects were satisfactory: removal rates of turbidity and A_{440nm} were more than 70% and 80%. The optimal dose of PAC was 0.11mmol Al^{3+}/L where removal rates of the above two indexes reached 80.4% and 89.3% respectively. Under adding AS, both turbidity and algal level were removed well with the dose range of 0.30~0.41mmol Al^{3+}/L, and the removal efficiencies of the two indexes reached 72.5% and 77.3% respectively at the optimal dose (0.41mmol Al^{3+}/L).

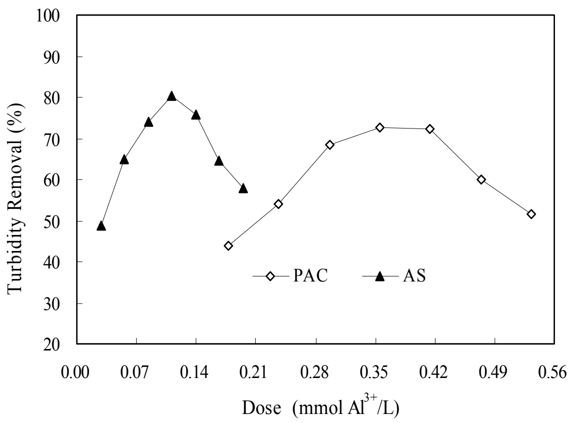

Figure 1. Influences of AS and PAC on removal of turbidity

The treatment effects achieved by adding PAC were significantly superior to those of AS in their respective appropriate dose range. Furthermore, dose of PAC was less than that of AS under gaining better effects. Meeting the optimal dose, aluminum dose of PAC was 73 percent less than that of AS.

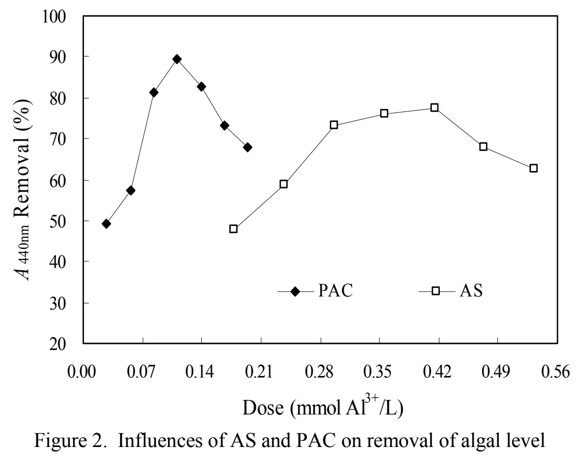

Figure 2. Influences of AS and PAC on removal of algal level

B. Floc Fractal Dimension

At the optimal dose of PAC and AS, Fig. 3 showed the relationship between projected area (A) and perimeter (P) of floc image in double logarithmic coordinate by image analysis. According to the power law, the slope of the straight line obtained by linear regression was the two-dimensional fractal dimension (D_2). The D_2 value was 1.3399 (R^2=0.95) for AS-MA flocs at the optimal dose, and it was 1.2246 (R^2=0.93) for PAC-MA flocs.

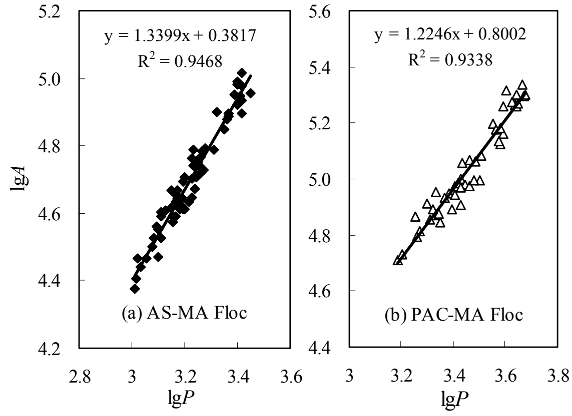

Figure 3. D_2 determined by image analysis

In Fig. 4, the relationships between D_2 and dose were shown adding PAC and AS. Both of the two curves presented that D_2 values decreased first and then increased with increasing dose. The changing tendency was influenced obviously by various coagulation mechanisms [10].

The charge neutralization dominated at the moderate dose for AS; at that time, the algae cells were destabilized to form micro flocs, and then the micro flocs gathered into bigger and branched flocs by flocculation. When AS was added excessively, plenty of amorphous aluminum hydroxides played sweep role; algae cells were embedded in the amorphous aluminum hydroxide, so the flocs structure was more compact than those formed by charge neutralization. Therefore Fig. 4 reflected that the D_2 values of flocs formed with adding the moderate AS (0.30~0.41mmol Al^{3+}/L) were lower than those generated with overdose (>0.47 mmol Al^{3+}/L).

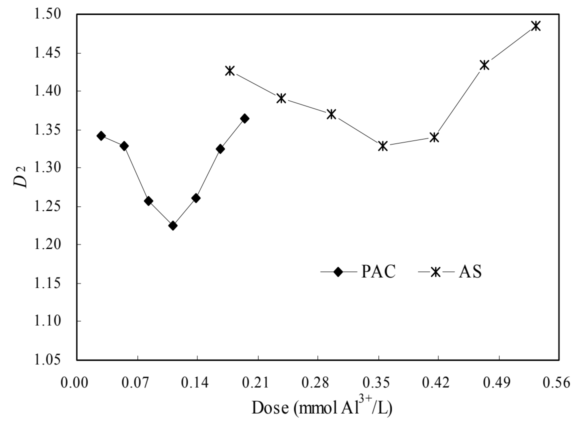

Figure 4. Influences of AS and PAC on structure of algae floc

PAC is a kind of inorganic polymeric coagulant. When PAC is dissolved in water, it can not only destabilize negatively charged particles in water by charge neutralization but also link the particles together by bridging mechanism. Shown as Fig. 4, at low dose PAC, the charge neutralization was a major mechanism of algae destabilization; and meanwhile the flocs generated by charge neutralization domain had higher D_2 values, which meant that the flocs structure was densely packed. The bridging mechanism dominated at the moderate PAC (0.08~0.14 mmol Al^{3+}/L); by contrast, the flocs structure was large, highly branched and loosely bound as a result of bridging, since active groups of a polymer chain adsorbed a lot of algae cells by ionic bonds, covalent bonds, hydrogen bonds and electrostatic interactions. When excessive PAC (> 0.17mmol Al^{3+}/L) was added, the more dense flocs formed by the sweep of amorphous aluminum hydroxide, which had higher fractal dimensions. When both dosages of AS and PAC were moderate, the D_2 values of flocs generated with PAC were lower than those with AS, on account of the flocs having relatively open structure by bridging.

C. Floc Strength

The variations of flocs strength with dosages of PAC and AS were presented in Fig. 5. The two curves showed same variations that *SF* values increased and then decreased with the increase of the dose. The variation was also related to different coagulation mechanisms closely.

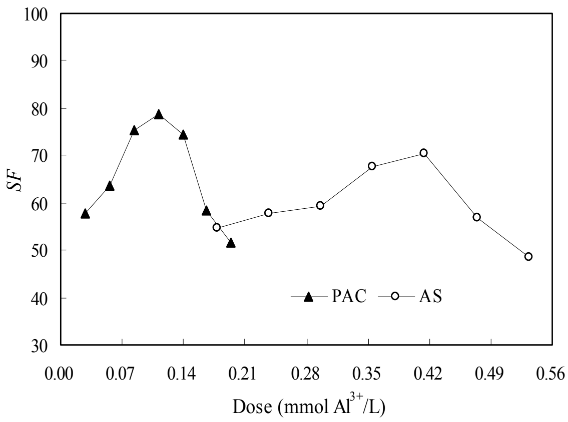

Figure 5. Influences of AS and PAC on strength of algae floc

Adding AS, the results showed that the floc strength formed under charge neutralization was stronger than that generated by sweep coagulation, because the interparticle bonds in precipitate of aluminum hydroxide were weaker, besides, there were electrostatic repulsions among the algae cells enmeshed in the precipitate. When PAC was added, the results indicated that the floc strength formed under bridging flocculation was the strongest in three coagulation mechanism, and the reason could be ascribed to the strong bonds of polymer chains [11].

During the process of flotation separation, both floc structure and strength have important impact to flotation effect. The flocs with larger size and highly branched structure have more adhesion points, which can attach to more tiny bubbles; furthermore, the flocs with higher strength have more powerful resistance to the hydraulic shear force due to rising bubbles and currents. On the basis of the above reasons the performance of flotation separation can be improved. Synthesizing the above figures, when moderate PAC was added, the satisfactory treatment effects resulted from the algae flocs with highly branched structure and higher strength by polymer chain serving as a bridge.

IV. CONCLUSION

Floc structure and strength play important roles in rich-algal water treatment by coagulation/flocculation/DAF process, and they are apparently affected by the selection of coagulants and dose. The experimental results showed: meeting the appropriate dose range (measured in Al^{3+}) simultaneously, dose of PAC was less than AS; and treatment effect obtained by PAC was better than AS. Adding the moderate PAC, polymers could adequately play bridging role, and thereby the algae flocs with highly branched structure and higher strength formed by bridging flocculation; this kind of algae flocs could firmly adhere to more tiny bubbles and be significantly more resistant to the hydraulic shear, so the performance of flotation separation using PAC could be improved compared with adding AS. Given treatment effect and harm of residual aluminum in water, PAC is is superior to AS in rich-algal water treatment by coagulation/flocculation/DAF process.

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