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# The Combined Action of Surfactant Mixture and Flocculants for Black Carbon Removal

Jingke Song,<sup>1,2</sup> Caiting Li,<sup>1,2</sup> Pei Lu,<sup>1,2</sup> Qingbo Wen,<sup>1,2</sup> Qi Zhan,<sup>1,2</sup> and Yapei Zhao<sup>1,2</sup>

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In this paper, two commercial surfactants with excellent performance nonionic fatty alcohol polyoxyethylene ether (AEO-9), and anionic sodium dodecyl benzene sulfonate (SDBS) were mixed in different mole ratios to enhance the solution's wetting ability and the flocculating efficiency of black carbon (BC). Results showed that when the mole ratio of AEO-9: SDBS was 1:7, and the concentration of the solution was 0.57 mM, and the solution obtained the lowest surface tension. Furthermore, the existence of AEO-9 in the surfactant mixture reduced the influences of temperature and polyelectrolytes with different electrical properties on the solution's surface tension. In the flocculation system, the flocculation behavior of BC suspensions by anionic polyacrylamide (PAM-A) and polyaluminium chloride (PAC) in the presence of a surfactant mixture were investigated. The results suggested that when BC suspensions were pretreated with the surfactant mixture, the existence of SDBS made the surface of BC own more negative charges. The flocculation ability of PAM-A was governed mainly by bridging. Addition of PAM-A could not get a higher flocculating efficiency in two addition way. When PAC was added into AEO-9-SDBS pretreated BC suspension solution, the electrostatic attraction and the charge neutralization between PAC and BC particles were enhanced significantly.

**Keywords** flocculants; flocculating efficiency; surfactant mixture; zeta potential

## INTRODUCTION

Soot in black smoke (BS), as one of the major air pollutants, is derived from incomplete combustion of carbon-based fuels. The main components of soot are black carbon (BC) and organic carbon (OC) (1,2). Black carbon, which is the main component of soot, can lead to global warming, carry carcinogenic compounds, and cause serious health risks (2–4). China is the largest coal producer and consumer country in the world; therefore, pollution by black carbon is even more serious (5–8). Nevertheless, due to the special

physical and chemical characteristics of BC, conventional dedusting technologies such as cyclone separator, electric precipitation, and bag-type dedusting, etc., could not get a high removal efficiency of BC particles (8,9). Thus, developing an effective method to remove carbon black particles from BS is urgent and significant.

In terms of removing black carbon by wet dust removal, the biggest problem is the hydrophobic surface of black carbon particles which makes it not easy to be trapped by liquids (9). In order to decrease the interfacial tension of black carbons, surfactants are used to modify their hydrophobic surface (10,11) and hence BC will be easier to be removed from black smoke. However, in this method, a large amount of black carbon is retained in water which inhibits the reuse of water to a large extent. Therefore, for the recycling of water and surfactants, the particles must precipitate to the bottom of the solution quickly. Flocculation, as a process widely used in industry to remove finely divided suspended solids (12,13) by adding flocculants to the mixture solutions, can meet this requirement.

Recently, for the numerous applications and potentially superior properties of surfactant mixture compared to a single surfactant, its behavior and performance has been extensively studied (14,15). Moreover, nonionic surfactants are relatively insensitive to ionic strength and pH, which provided a robust means for sterically stabilizing colloidal particles through adsorption (10). Thus, in our study, nonionic surfactant fatty alcohol polyoxyethylene ether (AEO-9) blended with anionic surfactant sodium dodecyl benzene sulfonate (SDBS) was investigated. We chose these surfactants as they have shown excellent solubilization performance and are fairly cost-effective (9). Hence, in what follows, the nonionic surfactant AEO-9 was mixed with anionic surfactant SDBS in different mole ratios. Meanwhile, the addition of flocculants was studied in two modes: (1) the surfactant mixture was used as a pretreatment agent. After the addition of the surfactant mixture, anionic polyacrylamide (PAM-A) or polyaluminium chloride

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(PAC) were added in different concentration; (2) PAM-A or PAC was added simultaneously with the addition of surfactant mixture in different concentrations, respectively.

## EXPERIMENTAL

### Materials

AEO-9 (AR Shanghai, China), SDBS (AR Tianjin, China), PMA-A (AR Tianjin, China), and PAC (AR Shanghai, China) were used as received. The critical micelle concentration (CMC) of AEO-9 and SDBS are 0.12 mM and 1.0 mM, respectively. Stock solutions of AEO-9, SDBS, PMA-A, and PAC were made up to 1 mM, 0.01 M, 0.1 g/L, and 10 g/L, respectively. All the solutions were prepared using deionized water.

The BC samples were derived from a coal-fired ceramics factory in Liling, China. Median particle diameter ( $D_{50}$ ) of the black carbon which was measured using a laser particle analyzer (JL-1155, Sichuan, China) was  $5.16\mu\text{m}$ , and the Brunauer-Emmett-Teller (BET) specific surface area of black carbon was  $14.589\text{ m}^2/\text{g}$ . Samples were dried at  $105^\circ\text{C}$  for 3 hours and then kept in desiccators at room temperature.

### Experimental Procedures

#### Surface Tension Measurements

A Model JZ-200A surface tensiometer (Chengde Precision Instrument Co, China) was used to measure the surface tensions of the solutions with surfactant mixture in different mole ratios at  $25^\circ\text{C}$ . Moreover, the mutual interaction between flocculants and surfactants could also be characterized through measurement of surface tension measurement (16). For comparison purposes, different concentrations of PMA-A (0.5 mg/L, 1 mg/L, 3 mg/L, 5 mg/L, and 7 mg/L) or PAC (100 mg/L, 200 mg/L, 300 mg/L, 500 mg/L, and 700 mg/L) were added into the AEO-9 solution, SDBS solution, and AEO-9-SDBS mixed solution in CMC, respectively. All experiments were performed in triplicates for the average calculation.

#### Flocculation Experiments

In mode (1), AEO-9-SDBS mixtures were prepared in CMC. Subsequently, 0.1 g BC was added to the solutions, respectively. After each addition, the mixture was vigorously stirred for 30 s, and then PMA-A or PAC was added in different concentration, followed by slow mixing for 1 min. In mode (2), the solutions were prepared by PMA-A or PAC blended with AEO-9-SDBS mixture. After the addition of 0.1 g BC, they were also vigorously stirred for 30 s and followed by slow mixing for 1 min. After that, both mode (1) and mode (2) were sustained for 5 min of gravitational segregation procedure (17).

To investigate the flocculating efficiency (FE), optical density (OD) of the settled solutions were measured with a visible spectrophotometer (722, Shanghai, China) at

550 nm. The control experiment was prepared by adding 0.1 g BC to the surfactant mixture solution. Samples were drawn from 2 cm below water surface and measured for  $\text{OD}_{550}$ . The FE of different flocculants in different concentrations was calculated according to Eq. (1) (17–19). All experiments were performed in triplicates for the average calculation.

$$FE(\%) = \left( \frac{b-a}{b} \right) \times 100 \quad (1)$$

where  $a$  and  $b$  are the optical density (OD) of the sample and control experiment at 550 nm, respectively. While re-suspended in the supernatant/sediment bed phase, they were sampled for later measurement of zeta potential (20).

#### Zeta Potential Analysis

The flocculation mechanisms of different flocculants were investigated by analyzing the zeta potentials of the flocculation systems. In the two modes, the variations of zeta potentials in different concentration of PMA-A and PAC were all measured using a Malvern ZS Nano S analyzer (UK). Besides, the zeta potential of BC suspension solution without additives and BC suspended solution with AEO-9-SDBS mixture in CMC were also measured.

## RESULTS AND DISCUSSION

### Surfactant Mixture Properties

The surface tension variations of the surfactant mixture AEO-9-SDBS in different mole ratios were investigated. The concentrations of AEO-9 solutions and SDBS solutions were varying from 0.01 mM to 0.12 mM and from 0.1 mM to 1.2 mM, respectively. The results were shown in Fig. 1.

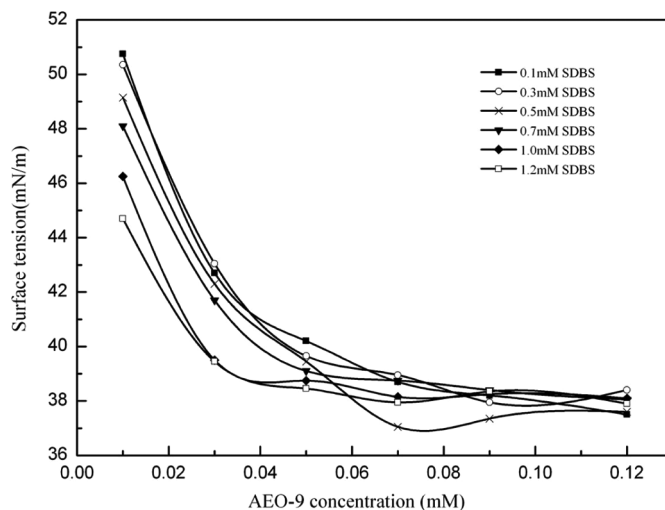


FIG. 1. Surface tension of different mixture solutions.

As shown in Fig. 1, when the mole ratio of AEO-9: SDBS was 1:7 and the solution concentration was 0.57 mM, the mixture solution had the lowest surface tension. When the solution is treated as the ideal solution, the CMC of the mixture surfactants solutions can be estimated by the following equation (15,21,22):

$$1/\text{CMC} = X_1/\text{CMC}_1 + X_2/\text{CMC}_2 + \dots + X_i/\text{CMC}_i \quad (2)$$

where  $X_i$  is the mole fraction of surfactant  $i$  in the mixed solution, thus the theoretical value of CMC is 0.53 mM. It is close to the solution concentration at which the solution had the lowest surface tension. Xie (22) found that, in general, the difference between the theoretical value and the experimental value of CMC was 0.01 mM-0.06 mM in the AEO-9-SDBS system. Therefore, the CMC of AEO-9-SDBS mixture could be regarded as 0.57 mM in the permitted range.

In order to analyze the nature and strength of the interaction between two surfactant molecules in the mixed micelle, the value of  $\beta$  which is the molecular interaction parameter, can be used. As the solution concentration is CMC, the  $\beta$  value is  $\beta^M$  for micelle, which can be calculated using Eq. (3) (23,24):

$$\beta^M = \frac{\ln \frac{\alpha c_1^M}{x_1^M c_1^M}}{(1 - x_1^M)^2} \quad (3)$$

where  $\alpha$  is the mole ratio of surfactant 1 in the mixed aqueous solution, and  $x_1^M$  is the mole ratio of surfactant 1 in the mixed monolayer of the mixed solution surface.  $c_1^M$  and  $c_2^M$  are the CMC of surfactant 1 solution and the mixed solution, respectively.  $x_1^M$  can be calculated by the following equation (23,24):

$$\frac{(x_1^M)^2 \ln \frac{\alpha c_2^M}{x_1^M c_1^M}}{(1 - x_1^M)^2 \ln \frac{(1-\alpha) c_1^M}{(1-x_1^M) c_2^M}} = 1 \quad (4)$$

where  $(1 - \alpha)$  is the mole ratio of surfactant 2 in the mixed aqueous solution.  $1 - x_1^M$  is the mole ratio of surfactant 2 in the mixed monolayer of mixed solution surface.  $c_2^M$  is the CMC of surfactant 2 solution.

According to the calculation, the value of  $\beta^M$  was 0.32 which closed to zero, indicating that there was little or no change in interactions upon mixing (24). To prove this conclusion, the surface tension variations of solutions with AEO-9(0.07 mM)-SDBS (0.5 mM), AEO-9(0.07 mM), and SDBS (0.5 mM) in different temperatures were investigated, respectively. The results were shown in Fig. 2. It was clear that the surface tensions of AEO-9(0.07 mM)-SDBS (0.5 mM) solutions were always close to that of AEO-9(0.07 mM) solutions when the temperature of the solutions was varying from 10 to 70°C. Thus the interaction between AEO-9 and SDBS was negligible in the given concentration.

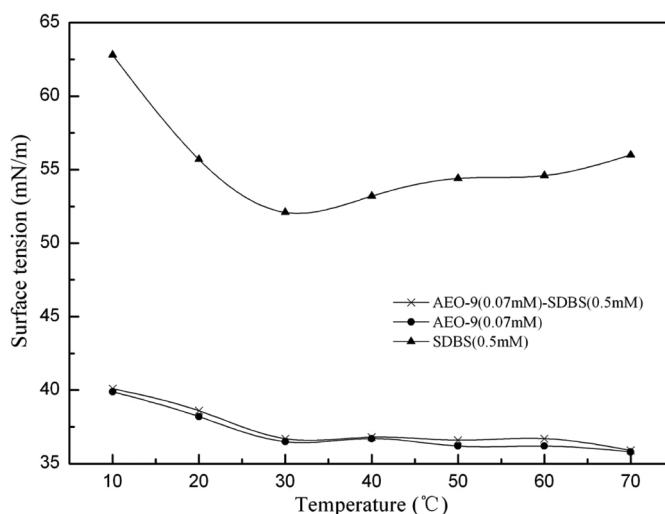


FIG. 2. Influence of temperature on solutions' surface tension.

Besides,  $x_1^M$  was 0.55 at this time, and this calculation suggested that the content of AEO-9 in micelle was slightly higher than SDBS. While AEO-9 had a much better quality than SDBS in terms of lowering the solution's surface tension (9), and the surface tension of AEO-9(0.07 mM)-SDBS (0.5 mM) solution was much lower than that of SDBS (0.5 mM) solution when the temperature of the solutions were varying from 10 to 70°C. Meanwhile, according to the analysis of Fig. 2, it could be found that the effect of temperature on the solution of AEO-9 was slighter than that of the solution of SDBS. Therefore, the presence of AEO-9 in the surfactant mixture reduces the influence of temperature on the solutions' surface tension.

### Surfactant-Flocculant Interaction

After adding different kinds of flocculants with different concentrations, the interactions between flocculants and surfactants (surfactant mixture) were investigated based on the changes of the solutions' surface tension. To elucidate the behaviors of AEO-9 and SDBS in the mixture, the surface tensions of AEO-9(0.07 mM) mixed with different concentrations of PAM-A or PAC was measured, and so was SDBS (0.5 mM).

The effects of the flocculants in terms of the solutions' surface tension are presented in Fig. 3. As shown in Figs. 3(a) and 3(b), the surface tension of AEO-9(0.07 mM) mixed with different concentrations of PAM-A or PAC were all changed in a small range. The surface tension of AEO-9(0.07 mM) solution was 36.9 mN/m, which was a little lower than that of the mixtures. Nevertheless, the surface tension of SDBS (0.5 mM) solution was 54.3 mN/m, the surface tension of SDBS (0.5 mM) mixed with PAM-A or PAC were changed from 61.8 mN/m to 70.2 mN/m, and from 60.3 mN/m to 69.0 mN/m, respectively. Thus, the

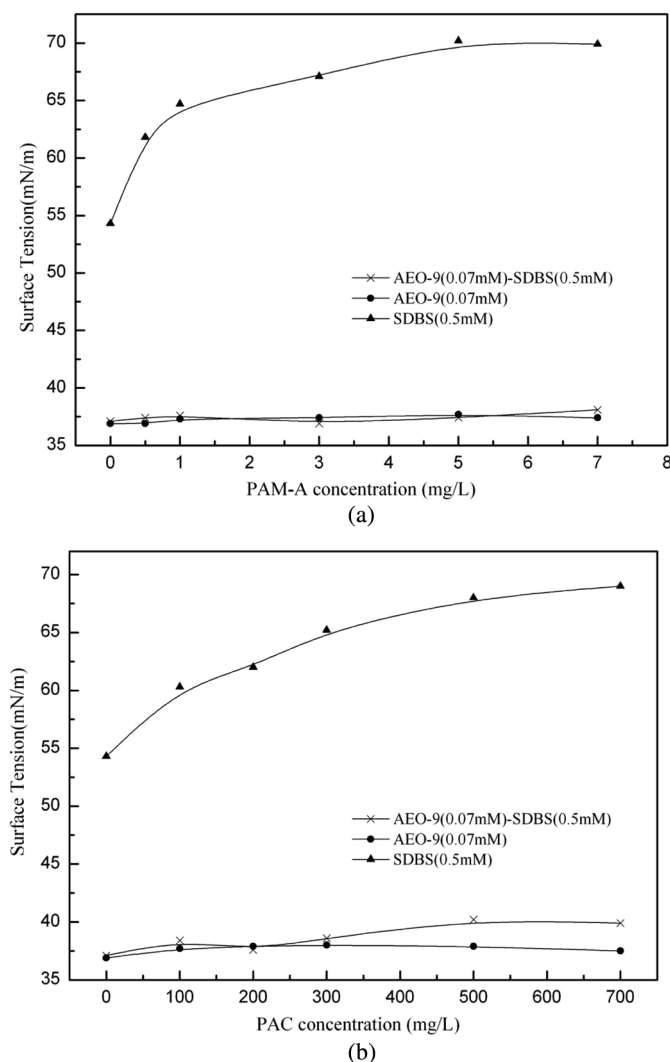


FIG. 3. The interaction between surfactant and flocculants.

addition of flocculants made the SDBS solutions' surface tension increase larger. Consequently, it could be concluded that the mutual interaction between PAM-A (PAC) and AEO-9 was negligible, while the interaction between PAM-A (PAC) and SDBS was immense.

However, from Fig. 3(a), when SDBS (0.5 mM) was mixed with AEO-9(0.07 mM) and PAM-A in different concentrations, the surface tension change was just close to AEO-9(0.07 mM) alone. Nevertheless, from Fig. 3(b), when SDBS (0.5 mM) was mixed with AEO-9(0.07 mM) and PAC in different concentrations, the surface tension changed from 37.6 mN/m to 40.2 mN/m. While the surface tension of AEO-9-SDBS solution was 37.1 mN/m. Hence, the addition of PAM-A almost had no impact on the surfactant mixture solutions' surface tension, but the addition of PAC made the surface tension of the solution increase slightly. It seemed to be that the existence of AEO-9 lowered the impact of PAM-A or PAC on the surfactant mixture, even if the

content of AEO-9 in the surfactant mixture was extraordinary little. Moreover, as the dissociation of SDBS and the hydrolysis of PAC produced ions with different charges, the combination of the two kinds of ions led to the slight increase of the surface tension of the solutions. Thus, a small quantity of AEO-9 could change the properties of the surfactant mixture enormously.

## Flocculation Behavior

### Effect of Different Addition Ways of Flocculants

Both the flocculating efficiency (FE) of BC particles and zeta potential in the flocculation system were used to investigate the influence of different addition ways of flocculants on the flocculation. The results were shown in Figs. 4 and 5, respectively.

Figure 4(a) showed that when PAM-A was added into AEO-9-SDBS pretreated BC suspended solution, along

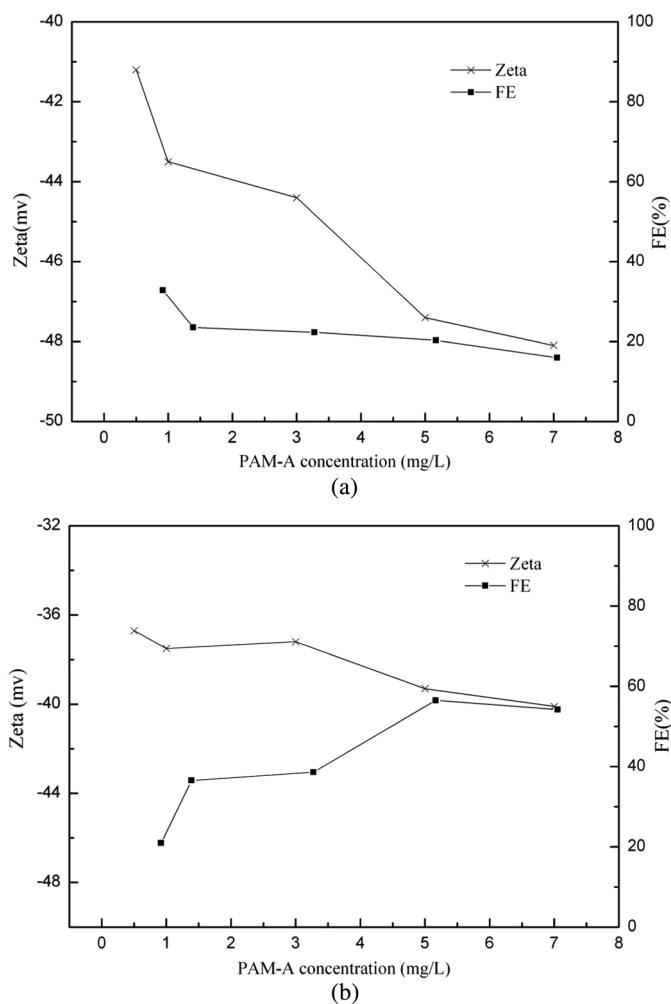


FIG. 4. Effect of PAM-A concentration on flocculating efficiency and zeta potential of BC suspended solution in different addition way: (a) PAM-A added to AEO-9-SDBS pretreated BC; (b) PAM-A : AEO-9-SDBS added to BC.

with the increase of PAM-A concentration, FE of BC particles was continuously falling and the peak efficiency was 32.8%, which was too low. However, from Fig. 4(b), when PAM-A and AEO-9-SDBS were added into the BC suspended solution simultaneously, along with the increase of PAM-A concentration, FE of BC particles increased at the beginning and reduced when the concentration of PAM-A exceeded 5 mg/L, and the maximum flocculating efficiency was 56.5%. Thus, the second addition way of PAM-A could get a higher flocculating efficiency. From Figs. 4(a) and 4(b), the two addition ways of PAM-A both made the systems' zeta potential lower than that of the BC suspended solution. It was likely that the hydrolysis of PAM-A and the dissociation of SDBS produced negative ions, when they adsorbed to the surface of BC particles, there would be more negative charges on the surface of BC particles. Besides, the values of zeta potential in mode (1) were lower than that of mode (2) in the whole range of PAM-A concentration.

As the hydrolysis of PAM-A produced negative ions which owned the same charges as the surface of BC particles, the main flocculation mechanism of PAM-A was bridging. Bridging occurred when PAM-A extended from the particles' surface into the solution. In certain situations, negative charges on the particles' surface could promote the extension of PAM-A. However, when they were too many, the repulsion forces between them would reject the flocs' aggregation. In mode (1), the pretreatment of AEO-9-SDBS made the surface of BC particles adsorb many negative ions due to the dissociation of SDBS. Thus, from Fig. 4(a), when PAM-A was added in, the values of zeta potential in the system reduced with the increase of PAM-A concentration. When the surface of BC particles owned too many negative charges, the flocs' aggregation was rejected and the FE of BC particles dropped. However, from Fig. 4(b), when PAM-A and AEO-9-SDBS were added simultaneously into the BC suspended solution, though zeta potential of the system fell, the FE of BC particles increased at the beginning. It was likely that the simultaneous addition of PAM-A and AEO-9-SDBS into the BC suspended solution made PAM-A and SDBS own the equal chance to adsorb on the BC particles' surface. Thus, when the concentration of PAM-A was equal, different addition ways of PAM-A might lead to different zeta potential. However, both the addition ways of PAM-A could not get a higher flocculating efficiency of BC.

On the contrary, in the two addition ways of PAC, mode (1) could reach higher flocculating efficiency than mode (2). As shown in Fig. 5(a), in mode (1), along with the increase of PAC concentration, the FE of PAC increased rapidly at the beginning, and when the concentration of PAC reached 300 mg/L, the FE of BC reached the peak value at 94.5%. After that, the FE of BC reduced rapidly as the concentration of PAC kept increasing. However, from Fig. 5(b), in mode (2), when PAC and AEO-9-SDBS were added

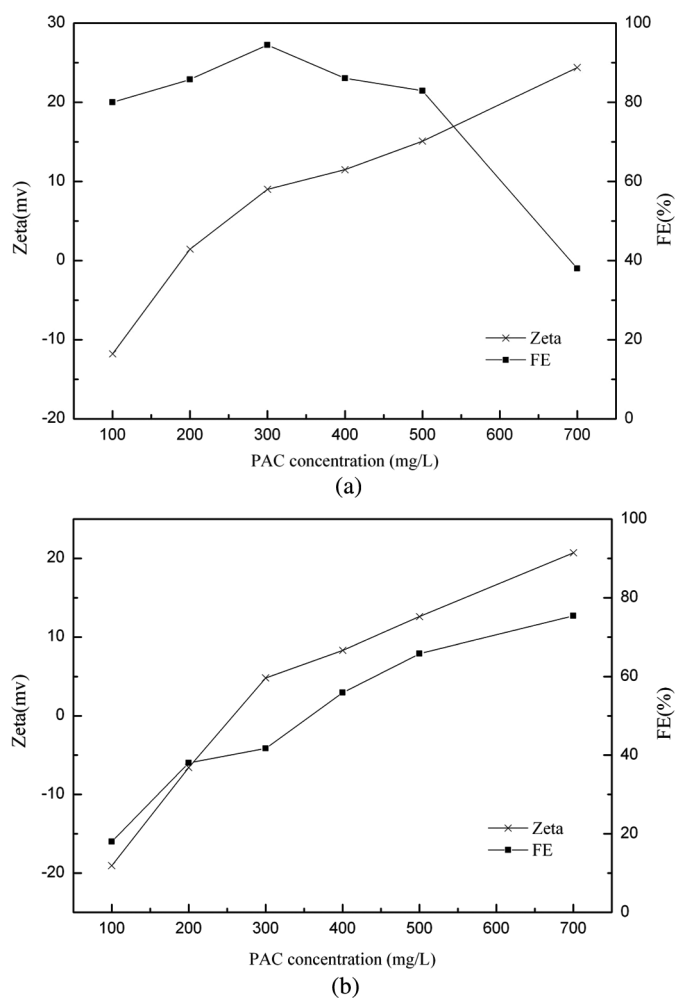


FIG. 5. Effect of PAC concentration on flocculating efficiency and zeta potential of BC suspended solution in different addition way: (a) PAC added to AEO-9-SDBS pretreated BC; (b) PAC:AEO-9-SDBS added to BC.

simultaneously, the FE of BC particles also increased with the increasing concentration of PAC. But the FE of BC was still far below 94.5%, and even the concentration of PAC was 700 mg/L.

Comparing Fig. 5(a) with Fig. 5(b), the influence of different addition ways of PAC on the zeta potential of the flocculation system could be found. In these two modes, it could be clearly seen that the zeta potential of these two addition ways of PAC followed a similar trend. That means when the concentration of PAC increased, the values of zeta potential in the two systems were all increased rapidly, and the values of zeta potential in mode (2) were always lower than that of mode (1) in the same concentration of PAC. It was known that the hydrolysis of PAC produced positive ions. In the two modes, followed by the increasing concentration of PAC, certain positive ions were released from the hydrolysis of PAC. Therefore, the values of zeta potential kept increasing and the charges of the particles' surface were altered.

It was likely that the highest flocculating efficiency occurred when the zeta potential of the flocs was close to zero (13). However, as shown in Fig. 5(a), it could be found that when the PAC concentration was 300 mg/L, the system had the maximum flocculating efficiency, but the value of the zeta potential in the system was 9.01 mV, which was a little higher than that of the addition of PAC at 200 mg/L whose zeta potential was 1.44 mV. To explain this, the flocculation process should be considered. It is well known that the flocculation can be divided into two steps. The first step of flocculation is the aggregation of suspended solids into larger particles and the second step involves the coalescing of aggregates into large flocs (13). Therefore, when the concentration of PAC was 200 mg/L the positive ions were almost used to neutralize the negative charges of BC particles. In order to get a higher setting rate, the concentration of the flocculant should be a little higher than the concentration at which zeta potential of the particles was close to zero. In this way, the bridging and enmeshment of PAC could give full play to the flocculation process. Hence, when the concentration of PAC was 300 mg/L, the second step of flocculation could complete successfully and PAC could obtain a good performance. Therefore, in the flocculation system, zeta potential could reflect the condition of charge neutralization of PAC. However, we could not confirm the optimal dosage of flocculant by using the zeta potential only. After that, the increase of PAC concentration would make the FE of BC particles decrease rapidly. Because the surface of BC particles owned too many positive charges, there was a strong repulsion force between them, and they would inhibit small flocs from growing into big ones (19). Then the solutions became re-stabilized.

From Fig. 5(b), in mode (2), when PAC and AEO-9-SDBS were added into the BC suspended solutions simultaneously, the charge neutralization between PAC and SDBS would produce micelles which would impair the charge neutralization between PAC and BC particles but avail to the bridging between PAC and BC particles. Hence, in mode (2), even though the value of zeta potential in the system reached 23.3 mV, the flocculating efficiency still increased, but this method could not have a higher flocculating efficiency even if the concentration of PAC was 700 mg/L.

In view of the above, in terms of BC particles, addition of PAC could have better flocculation performance than PAM-A, especially in mode (1). The optimal concentration of PAC was 300 mg/L and the FE of BC particles could reach 94.5% in 5 min.

#### *Roles of Surfactants in the Flocculation System*

In our previous work (8), the adsorption mechanism of SDBS was investigated. It was reported that the main interactions involved in SDBS adsorption on carbon blacks were hydrophobic interactions between the alkyl chains of SDBS

and the carbon black surface. The adsorption of SDBS made the black carbon surface own more negative charges. Thus, when the BC suspension solution was pretreated with SDBS, the charge neutralization of PAC in flocculation could be enhanced. Besides, when the concentration of SDBS was 0.46 mM, zeta potential was  $-45.3$  mV. While the BC suspended solution had a zeta potential of  $-25.7$  mV, and the zeta potential of BC suspended solution which was pretreated by the surfactant mixture (AEO-9 (0.07 mM)-SDBS (0.5 mM)) was  $-37.7$  mV. That means the adsorption of AEO-9 prevented the adsorption of SDBS from the black carbon surface slightly. Therefore, in the flocculation system, when the flocculant was PAC, SDBS was mainly used to enhance the flocculating efficiency.

#### *Setting Performance*

To understand the setting velocity of BC particles in mode (1), different concentrations of PAC were added into BC suspended solutions with surfactant mixture pretreated, and the  $OD_{550}$  changed with time were measured (Fig. 6). The time-points for sampling were 0, 1, 3, 5, 10, and 20 min. Figure 6 showed that most of the curves declined sharply in the first 5 min; however, when the concentration of PAC was 300 mg/L, the curve declined sharply in the first 3 min. After that, the  $OD_{550}$  almost remained unchanged. It means that when the concentration of PAC was 300 mg/L, the flocculating process could be finished in the first 3 min.

In order to investigate the removal efficiency of BC particles with the solution, particle size distribution of black carbon in supernatant was measured with a laser particle analyzer (JL-1155, Sichuan, China). After adding PAC (300 mg/L) into the surfactant mixture pretreated BC suspended, the mixed solution was sustained for 3 min gravity settlement procedure. Then the particle size distribution of black carbon in the supernatant was carried out to elucidate

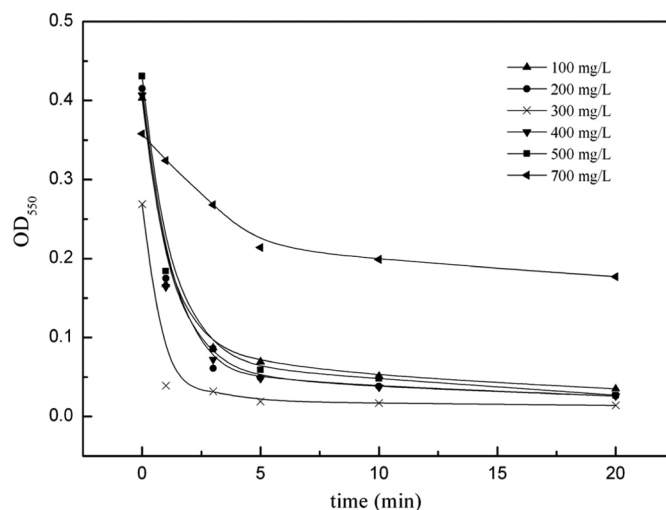


FIG. 6.  $OD_{550}$  varied with time at different concentration of PAC.

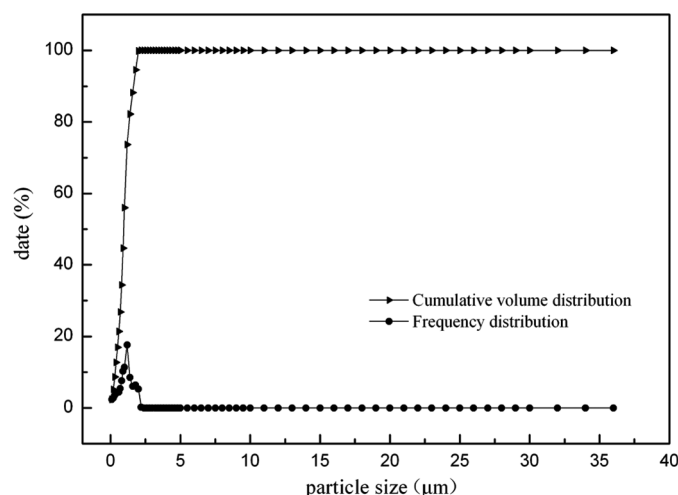


FIG. 7. Particle size distribution of black carbon in supernatant solution. Conditions: mixed solution was sustained for 3 min of gravity settlement procedure.

the flocculating performance of PAC (300 mg/L) with surfactant mixture (AEO-9-SDBS) pretreated. Figure 7 showed the distribution of particles size. It was clear that the particles size mainly distributed from 0.8  $\mu\text{m}$  to 1.4  $\mu\text{m}$ . While the average size of particles in the supernatant solution was 0.97  $\mu\text{m}$  and  $D_{50}$  was 0.95  $\mu\text{m}$ , compared with the  $D_{50}$  of BC sample (5.16  $\mu\text{m}$ ), the content of black carbon in the solution was reduced enormously.

## CONCLUSION

In this paper, the properties of the mixture surfactants, interactions between surfactants and flocculants, the mode of flocculants addition, and the optimal concentration of flocculants were investigated. The results showed that the optimal mole ratio of AEO-9: SDBS was 1:7, in this way the interaction between AEO-9 and SDBS was negligible. The presence of AEO-9 in the micelles reduced the influence of temperature on the solution and the interaction between the surfactant mixture and flocculants.

In the flocculation process, our results indicated that the surface of BC particles had negative charges, and the AEO-9-SDBS mixture pretreatment made the surface of BC particles itself more negative charges, which would be easier to combine with positive ions. As the hydrolysis of PAM-A produced negative ions which were identical with the charges on the surface of BC particles, the two addition ways of PAM-A could not get a higher flocculating efficiency. However, as the hydrolysis of PAC produced positive ions, which could neutralize the negative charges, zeta potentials of the systems in the two modes all increased along with the increase of the concentration of PAC. Especially, when PAC was added into the AEO-9-SDBS pretreated BC suspended solution, the flocculating efficiency

could reach to 94.5% in 5 min when the concentration of PAC was 300 mg/L. And the setting process could be completed in 3 min. The average size of particles in the supernatant could be controlled at 0.97  $\mu\text{m}$ .

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## SUPPLEMENTAL MATERIAL

Supplemental data for this article can be accessed on the publisher's website.

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