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Dewatering performance of aerobic granular sludge

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ABSTRACT

Aerobic granular sludge (AGS) is promising biotechnology for wastewater treatment. With the rapid development of this technology, a large amount of surplus AGS will be generated. However, the characteristics and mechanisms of AGS dewatering are nearly unreported. In this study, the dewatering performances of AGS and activated flocculent sludge (AFS) were compared, and the mechanism of AGS dewatering was systematically investigated. The results showed that the specific resistance to the filtration of the raw AGS ((2.52 ± 0.02) × 10^9 m/kg) was much lower than that of the raw AFS ((1.23 ± 0.03) × 10^{14} m/kg). The addition of cationic polyacrylamide (CPAM) had no significant effect on the AGS dewatering, and without the CPAM, the moisture content reached 80.2% after the AGS dewatering with the pressure block. The pressure block triggered the dissolution of extracellular polymeric substances. The results of three-dimensional fluorescence spectroscopy showed that the AGS could rapidly release a large amount of protein-like matter.

Keywords: Aerobic granular sludge (AGS); Dewatering performance; Extracellular polymeric substances (EPS); Three-dimensional excitation-emission matrix (3D-EEM)

1. Introduction

In recent years, aerobic granular sludge (AGS) has been considered to be one of the most promising biological wastewater treatment technologies because of its good settling performance, high biomass retention and high pollutant removal performance [1,2]. Compared with activated flocculent sludge (AFS), the AGS structure is dense, with regular shapes, smooth surfaces, nearly spherical or elliptical shapes, and clear outlines. The particle size of AGS is usually between 0.2 and 5 mm [3,4].

Currently, AGS technology has been successfully applied in full-scale wastewater treatment plants (WWTPs), such as at the Epe, Dinxperlo and Vroomshoop WWTPs in the Netherlands [5], the Gansbaai sewage treatment plant and Wemmershoek WWTPs in South Africa [6], and the Yancang WWTP in China [7,8]. Therefore, with the development of AGS technology, the surplus AGS also needs to be handled effectively. Dewatering is a crucial process for sludge treatment, which can effectively reduce the sludge quantity and sludge transportation and disposal costs. However, studies on AGS dewatering have rarely been reported.

Extracellular polymeric substances (EPS) are a product of the microbial activity, which is beneficial to the polymerization and adhesion between cells and plays an important role in maintaining the stability of the whole microbial community structure [9]. The main components of EPS are polysaccharides (PS) and proteins (PN) [10]. In addition, EPS content has a significant effect on floc stability and dewatering performance [11]. Higgins and Novak [12] demonstrated that dewaterability was related to the ratio of PN to PS in EPS. Zhang et al. [13] reported that PN

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generally played a more important role in AFS dewatering than PS. EPS in AFS can bind a large volume of water and prevent its release [14].

There are four types of water in AFS: free water, interstitial water, surface water, and internal water [15]. Only free water is independent of solid particles and can be easily removed by precipitation and mechanical dewatering. Interstitial water and surface water are trapped in the crevices of the sludge flocs and remain on the surface of solid particles. Internal water is water in bacterial cells and chemically bound in the sludge. According to previous studies, the sum of the interstitial water, surface water, and internal water are called bound water [16]. Therefore, AGS or AFS has two types of water, including free water and bound water. In AGS, EPS also facilitates the polymerization and bonding between bacterial cells, and they contain a large amount of bound water. EPS plays an important role in the AGS dewatering process [17,18].

Traditional chemical flocculants, such as inorganic flocculants and polymers [19,20], can reduce the water content of AFS by charge neutralization and bridging and by accelerating the filtration rate [21]. Other technologies, such as ultrasonic pretreatment, acid-base thermochemistry, and advanced oxidation [22], can decompose the components of EPS and decompose sludge. During these processes, the bound water is released, and the water content of the sludge cake decreases. However, research on AGS has mainly focused on its cultivation and stable operation [23–25]. There are few studies on the dewatering performance of AGS. The effect of chemical flocculants on AGS dewatering must be understood. The mechanism of the EPS degradation and transformation of bound water from AGS remains unclear.

In this study, AGS collected from a sequencing batch reactor (SBR) was used for the dewatering experiment, and the excess AFS dewatering performance was also analyzed. To explore the AGS dewatering mechanism, the dissolution of the EPS, and the release of bound water in the AGS were analyzed. The morphology and particle size of the AGS were also examined. A three-dimensional excitation-emission matrix (3D-EEM) was introduced to measure the fluorescence intensity of the protein-like matter.

2. Materials and methods

2.1. Source of AGS and AFS

The AGS experiment was controlled and cultivated with synthetic wastewater [3,26] in a 4 L inorganic glass SBR (1,000 mm × Φ90 mm) using AFS taken from the secondary sedimentation tank of the Qige Municipal Wastewater Treatment Plant (Hangzhou, China) (Fig. 1). The composition of the synthetic wastewater used in the study was sodium acetate, KH_2PO_4, NH_4Cl, $CaCl_2$, $MgSO_4·7H_2O$, $FeSO_4·7H_2O$, and other necessary nutrients (Table 1). The chemical oxygen demand (COD), NH_4–N, and total phosphorous (TP) concentrations in the synthetic wastewater were 500–550 mg/L, 25–28 mg/L, and 5–8 mg/L, respectively. When the SBR was operated for 50 d, the concentrations of COD, NH_4–N, and TP in the effluent were below 35, 1.3, and 0.82 mg/L, and the removal rates were above 93%, 95%, and 83.5%, respectively. After 70 d of operation, the concentrations of COD and NH_4–N in the effluent were almost zero. In this experiment, the SBR reactor was operated for 6 cycles per day, which was controlled by a time-delay relay, and every operation cycle mode was set to 5 stages: water inlet (5 min), aeration (150 min), sedimentation (30 min), drainage (5 min) and idle (50 min). The temperature and pH value of the reactor was controlled at 20°C ± 5°C and 7.0–7.5, respectively. After operating for 70 d, the mature AGS particles were 2–2.5 mm in size, spherical or oval, orange or yellow, and the surfaces were smooth and dense

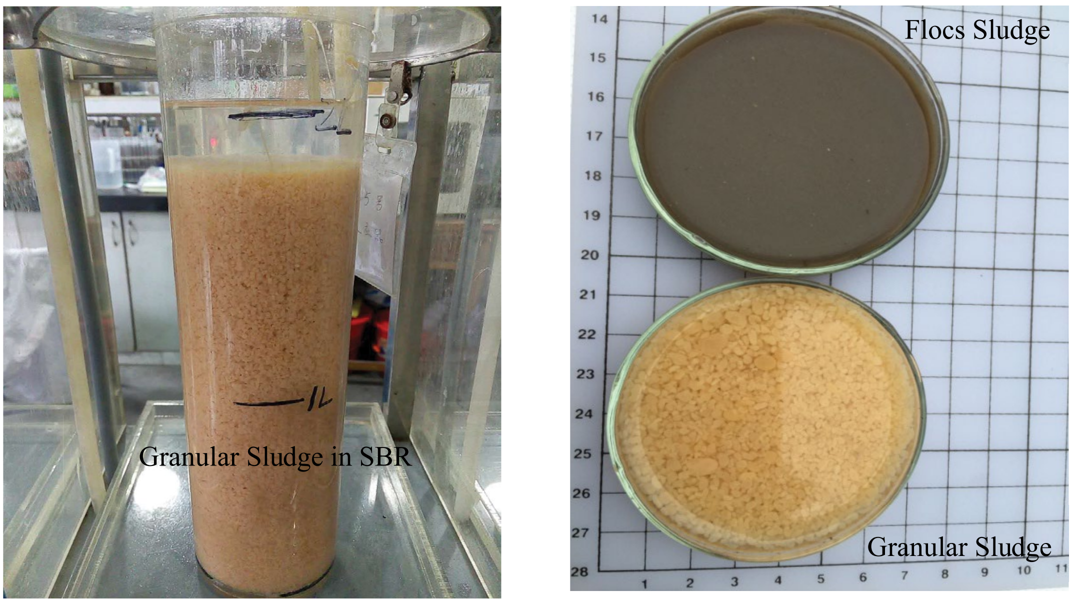

Fig. 1. Aerobic granular sludge from the lab-scale reactor and activated floc sludge from the secondary sedimentation tank.

with a volume index value between 45–47 mL/g. The solid concentration of the raw AGS was 18.21 g/L, which was mildly concentrated to 20.01 g/L for the experiments.

The AFS used in the experiments was also from the secondary sedimentation tank of the Qige WWTP and cultured by the ordinary SBR method, which had been in operation for a long time. The solid concentration of the raw AFS was 20.03 g/L for the experiments.

2.2. Centrifugal equipment

The centrifugal dewatering system includes a special centrifugal tube (Φ30 mm × 115 mm, made in our laboratory) and a conventional laboratory centrifuge (Anhui Zhongke Zhongjia low-speed centrifuge sc-03) (L × W × H = 350 mm × 370 mm × 295 mm). The self-designed centrifugal tube consists of three parts. The sludge samples were injected into Part I, and the dewatering sludge cakes were sampled from Part II. Finally, centrifugal water was obtained from Part III (Fig. 2). The pressure blocks were added when required.

2.3. Experimental design

Twelve beakers (1,000 mL) were divided into two groups (group A and group B). Group A was filled with 500 ml AGS, and group B was filled with 500 ml AFS. Three experimental conditions were conducted in each group, and six sets of experiments were performed in parallel (Table 2). For each sludge treated with the cationic polyacrylamides (CPAM), the mixture solutions were stirred quickly for 3 min and then stirred slowly for another 10 min. The amount of sludge for each experimental group was 20 g. The CPAM was provided by the WWTP. The moisture content was measured for all dewatered cakes, which were centrifuged for 20 min at 4,000 rpm. The fluorescence intensity of the protein-like matter was assessed for all centrifugal suspensions after centrifugation at 1,500 and 4,000 rpm for 20 min. The same size pressure block was added after the sample was prepared in a centrifugal tube before centrifugal dewatering occurred in set 2, set 4, and set 6.

Table 1
Detailed components of synthetic wastewater

Component	Concentration (mg/L)	Component	Concentration (mg/L)
NaAc	1,200	$CaCl_2$	21
NH_4Cl	115	$MgSO_4\cdot7H_2O$	102
KH_2PO_4	63	Microelement	0.5 mL/L
$FeSO_4\cdot7H_2O$	8	–	–

2.4. Analytical methods

2.4.1. Specific resistance to filtration

The specific resistance to filtration (SRF) is a comprehensive index that characterizes the sludge filtration performance and reflects the trend of blocking the solid–liquid separation. The SRF value was determined with the sludge of set 1 and set 5, and the main determination procedures were consistent with the literature [27].

2.4.2. Free water

According to Lee's research [28], free water is not associated with solid particles and includes void water that is not affected by capillary forces. To determine the percentage of free water in the AGS, a certain amount of samples were collected for dewatering by using a Büchner funnel (diameter = 12 cm) for 30 min at 0.05 Mpa. The moisture content of the filtered sludge cake was calculated according to the literature [27].

A separatory funnel and cylinder were used in the static concentrated filtration experiment. The liquid separatory funnels, with accurately measured A1 and B1 groups, were placed on the cylinder, and the amount of water in the cylinder was observed with the change time and recorded.

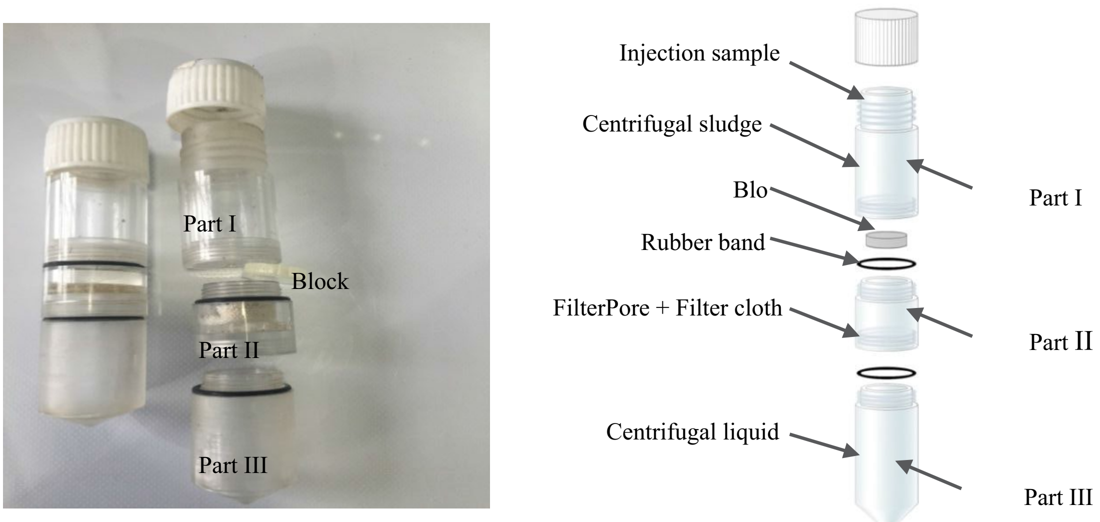

Fig. 2. Centrifugal experimental device.

Table 2
Six sets for experiments of AGS and AFS

	AGS (Group A)	AFS (Group B)
Set 1	Raw sludge (A1)	Raw sludge (B1)
Set 2	Raw sludge + block (A2)	Raw sludge + block (B2)
Set 3	Raw sludge + 20 mg/g SS CPAM (A3)	Raw sludge + 20 mg/g SS CPAM (B3)
Set 4	Raw sludge + 20 mg/g SS CPAM + block (A4)	Raw sludge + 20 mg/g SS CPAM + block (B4)
Set 5	Raw sludge + 40 mg/g SS CPAM (A5)	Raw sludge + 40 mg/g SS CPAM (B5)
Set 6	Raw sludge + 40 mg/g SS CPAM + block (A6)	Raw sludge + 40 mg/g SS CPAM + block (B6)

The experiment ended when there was no more filtrate exuded, and the moisture content of the filtered sludge cake was calculated.

2.4.3. EPS extraction and analysis

The AGS samples were centrifuged at 1,500; 3,000; and 4,000 rpm (20°C) for 20 min. The centrifuged supernatant was collected. Then, a modified methanol-NaOH extraction method was used to extract the EPS from the sludge [29]. PN and PS, which are the main components of biopolymers in the EPS, were measured using different methods. The PN concentrations were measured using the Kaumas Bradford method [30]. The PS concentrations were determined using the phenol sulfuric acid method [3].

2.4.4. Three-dimensional excitation-emission matrix

The 3D-EEM is a very useful spectral fingerprint technique to detect the different fluorescent peaks of proteins and humic acids [31]. The granular sludge can release biopolymers into the filtrate during the centrifugal dewatering process as intermediates between the inorganic particles in the AGS. The 3D-EEM spectra were measured on an F-4500 fluorescence spectrophotometer (Shanghai Lengguang Technology Co. Ltd., China) with an excitation range from 240 to 360 nm at 5 nm sampling intervals and an emission range from 280 to 360 nm at 2 nm sampling intervals. The scan speed was 2,000 nm/min with a 0.2 s response time, and the photomultiplier voltage was 500 V. The 3D-EEM data were analyzed by using Origin 8.0.

3. Results and discussion

3.1. Dewatering performance of AGS

In general, filtration is easier when the SRF value is less than 0.4×10^{13} m/kg. Filtration becomes difficult when the SRF value is between $(0.5–0.9) \times 10^{13}$ m/kg, and dewatering is more difficult when the SRF value is higher than 1×10^{13} m/kg [32]. In the experiment, the SRF value of the raw AGS was $2.52 \pm 0.02 \times 10^{9}$ m/kg, and that of the AFS was $1.23 \pm 0.03 \times 10^{14}$ m/kg. It was clear that the SRF value of the AGS was much smaller than that of the AFS. Compared with the reported value (0.4×10^{13} m/kg), the AGS is categorized as sludge with good dewatering performance. A5 was the AGS sample treated with 40 mg/g SS CPAM, and the SRF value was $1.98 \pm 0.05 \times 10^{9}$ m/kg. B5 was the AFS sample treated with 40 mg/g SS CPAM, and the SRF value was $1.69 \pm 0.08 \times 10^{13}$ m/kg (Table 3). The results show that the AGS had a smaller SRF value than that of the AFS regardless of the CPAM addition. This observation occurred due to the large specific gravity and the large grain size of the AGS. With the CPAM addition, the SRF value was smaller than that of the control group, which is possible because the CPAM can neutralize colloidal particles and bridge flocculation occurring between particles, which makes it easy to dehydrate the sludge.

3.2. Amount of free water

The AGS had a large proportion of free water content. Here, 20.03 g of the AGS sample was obtained through vacuuming; the wet weight of the granular sludge was 5.76 g, and the dry weight was 0.40 g after drying at 105°C for 2.5 h. Then, the amount of free water accounted for 72.68% of the total water. The results showed that the free water content accounted for a large proportion of the total water content in the AGS. The amount of free water in the AFS was 70.02%.

The filtration rate of the AGS (group A1) was significantly faster than that of the AFS (group B1), and the volume of the AGS filtered fluid rapidly increased in the cylinder filtrate (Table 4). After 15 min, the A1 average filtrate volume was 7.29 mL, and there was no free water exuded. However, the results were different in the AFS funnel. The substantial water was still absorbed in the sludge, and the dripping rate was very slow. There was only 3.5 mL of the filtered fluid. After 1 h of filtration, the average filtered fluid volume of the AGS was 7.75 mL, and that of the AFS was 4.79 mL. Therefore, the free water in the AGS could be easily removed by gravity, which helped to reduce the sludge volume.

3.3. Performance of centrifugal dewatering

The previous results showed that there is a strong bonding force between the sludge particles and the bound water [33]. The removal of this water required external forces, such as mechanical dewatering. Fig. 3 shows that the dewatering performance of the AGS was significantly better than that of the AFS. After centrifugation, the moisture contents of A1 and B1 were 87.1% and 94.2%, respectively. When adding the block during the centrifugal dewatering process, the moisture content of the AGS (A2) cake decreased from 87.1% to 82.1%, while the moisture content of the AFS (B2) cake was still higher than 90%. This result indicates that adding a block can enhance the AGS

Table 3
SRF values of AGS and AFS

	AGS	AGS	AFS	AFS
Sample ID	A1 (raw)	A5 (40 mg/g SS PAM)	B1 (raw)	B5 (40 mg/g SS PAM)
SRF value (m/kg)	$(2.52 \pm 0.02) \times 10^9$	$(1.98 \pm 0.05) \times 10^9$	$(1.23 \pm 0.03) \times 10^{14}$	$(1.69 \pm 0.08) \times 10^{13}$

Table 4
The filtered fluid volume of AGS and AFS variation with time during static concentrated filtrating

Time (min)	0	1	2	3	5	7	10	15	30	60
AGS fluid (ml)	0	1.41 ± 0.02	2.23 ± 0.04	3.34 ± 0.03	4.24 ± 0.06	5.65 ± 0.03	6.61 ± 0.03	7.29 ± 0.002	7.63 ± 0.05	7.75 ± 0.04
AFS fluid (ml)	0	0.56 ± 0.01	0.92 ± 0.02	1.31 ± 0.05	1.88 ± 0.02	2.42 ± 0.04	3.13 ± 0.05	3.49 ± 0.03	4.29 ± 0.04	4.79 ± 0.03

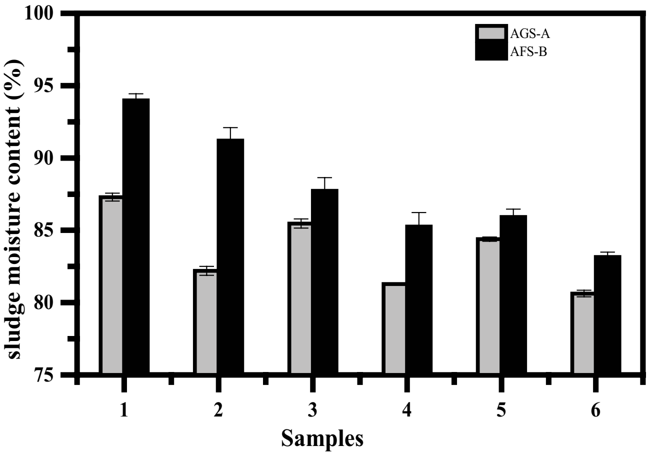

Fig. 3. The moisture content of the AGS and AFS cakes after centrifugal dewatering (S-1: raw sludge; S-2: raw sludge with block; S-3: raw sludge treated with 20 mg/g SS CPAM; S-4: raw sludge treated with 20 mg/g SS CPAM and block; S-5: raw sludge treated with 40 mg/g SS CPAM; and S-6: raw sludge treated with 40 mg/g SS CPAM and block).

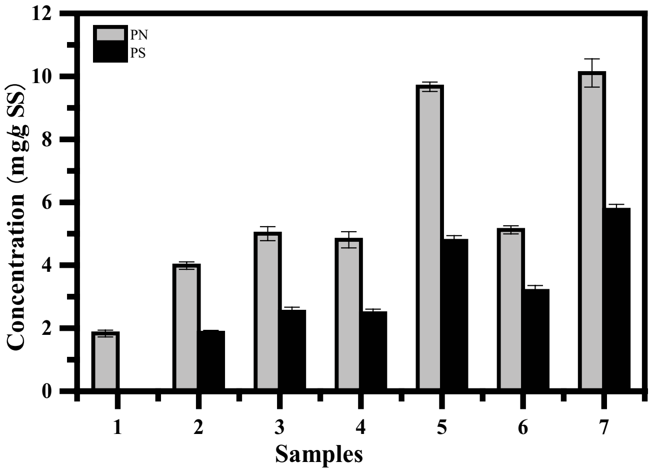

Fig. 4. Changes in the biopolymer concentrations in the AGS centrifugal fluids (S-1: AGS supernatant; S-2: AGS centrifuged for 20 min at 1,500 rpm; S-3: AGS centrifuged for 20 min at 1,500 rpm; S-4: AGS centrifuged for 20 min at 4,000 rpm; S-5: AGS centrifuged for 20 min at 4,000 rpm and block; S-6: AGS centrifuged for 20 min at 4,000 rpm and treated with 40 mg/g SS CPAM; and S-7: AGS centrifuged for 20 min at 4,000 rpm and treated with 40 mg/g SS CPAM and block).

dewatering due to the compressing synergism between the inorganic particles and the external force. After centrifugal dewatering, the moisture content of the AGS cake treated with 20 mg/g SS CPAM was 84.1% (A3), which was similar to that of the raw AGS (A1) cake. However, the moisture content of the AFS (B1 and B3) cakes treated by the CPAM was significantly decreased from 94.3% to 85.2%. The moisture content of the AGS (A4) cake was quite similar to that of group A2, which indicates that the CPAM played a very minor role in the AGS dewatering. In contrast, the CPAM played an important role in the AFS dewatering because the moisture content of group B4 was much lower than that of group B2. After centrifugal dewatering with the block, the moisture contents of the AGS (A6) and AFS (B6) cakes were 80.2% and 82.3%, respectively. The AGS moisture content of the sludge after treatment with 40 mg/g SS CPAM (A5) was slightly different from that of A3. The moisture content of the AFS (B3 and B5) cakes decreased significantly. The results showed that the CPAM had little effect on the dewatering performance of the AGS but could significantly decrease the moisture content of the AFS. One of the effects of the CPAM was to bridge the flocculation between particles, which was beneficial for allowing the water to pass through. The AGS has a large particle size of 2–2.5 mm, which was good for bridging the flocculation between particles. Then, during the centrifugal dewatering processes, the internal particles in the dense AGS structure compressed each other through the centrifugal force to promote the release of water. However, the composition of the AFS was highly dense fine solid particles and had strong hydrophilicity, so physical or chemical treatments, such as the addition of CPAM, are needed during centrifugal dewatering processes. When adding the external block pressure, the block pressure was larger between the particles, and more water was compressed out of the gap, which

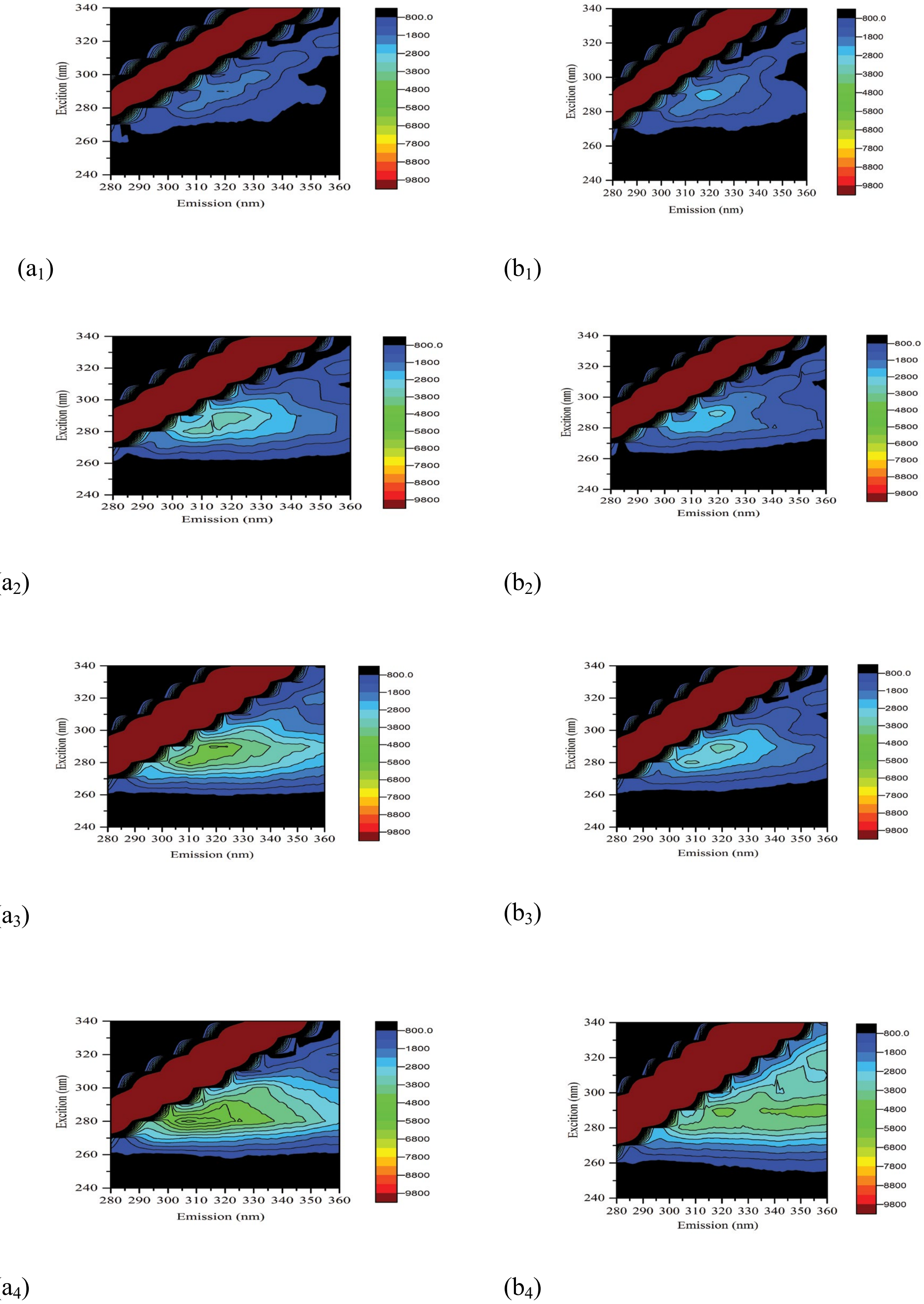

Fig. 5. Protein-like matter 3D-EEM fluorescence spectra of the supernatant and centrifugal fluid of AGS and AFS (the protein-like matter fluorescence peaks are located at excitation wavelengths/emission wavelengths = 275–300/310–330 nm). (a1) AGS supernatant, (a2) AGS centrifuged at 4,000 rpm, 20 min, (a3) AGS centrifuged at 4,000 rpm + block, 20 min, (a4) AGS at 4,000 rpm + 40 mg/g SS CPAM + block, 20 min, (b1) AFS supernatant, (b2) AFS centrifuged at 4,000 rpm, 20 min, (b3) AFS centrifuged at 4,000 rpm + block, 20 min, and (b4) AFS at 4,000 rpm + 40 mg/g SS CPAM + block, 20 min.

was beneficial for improving the dewatering performance. Therefore, the AGS can achieve a favorable dewatering performance without the CPAM treatment, which helps to save the costs associated with the CPAM dosages.

3.4. Release of biopolymers during the AGS dewatering process

EPS, which is a significant fraction of the sludge mass, has the ability to bind a large amount of water, especially bound water [22], and this has become a major obstacle for sludge dewatering. The moisture content of sludge is difficult to reduce by mechanical filtration [11]. Therefore, it is well accepted that the release of EPS and intracellular material could enhance sludge dewatering [34]. The dewatering performance of AGS can be evaluated by analyzing the biopolymer components in the centrifugal fluid. The changes in the main organic compounds in the biopolymers were measured. The results showed that mechanical centrifugation could promote the release of EPS from the AGS (Fig. 4). The supernatant of the raw AGS (without centrifugal dewatering) showed a small amount of PN content and no PS content. As the centrifugal speed was increased from 1,500 to 4,000 rpm, the PN content increased from 4.0 to 5.0 mg/g SS, and the PS content increased from 2.0 to 2.5 mg/g SS in the centrifugal solution. By analyzing the centrifugal fluid of the AGS samples treated with or without CPAM, the PN and PS concentrations in the centrifuged fluid were similar. These results indicated that CPAM had no obvious effect on the release of EPS into the AGS, which is consistent with the dewatering performance of AGS samples treated with CPAM. When the blocks were added, the PS and PN concentrations in the centrifuged fluid increased significantly, indicating that more PS and PN contents were released from the AGS. This increased release was attributed to the compression between the inorganic particles under the block pressure, which was beneficial for AGS dewatering.

3.5. 3D-EEM analysis

The 3D-EEM results of the AGS and AFS are shown in Fig. 5. There was almost no fluorescent value in the static supernatant, indicating that there was very little protein-like substance (Figs. 5a1 and b1). After centrifugation for 20 min at 4,000 rpm (Figs. 5a2 and b2), both sludge centrifugal liquids exhibited a small amount of fluorescence, but the fluorescence intensity in the AGS centrifugal liquid was higher than that in the AFS centrifugal liquid. This higher fluorescence intensity may be related to the mutual pressure effect of the AGS and partial protein material detachment. There was no change in the centrifuge conditions, but the fluorescent substances in the centrifuged liquid of the AGS increased significantly with the block addition for 20 min at 4,000 rpm centrifugation (Fig. 5a3). Based on the AGS particle shape after centrifugation, it could be concluded that the biological particles began to deform, and a small number of particles broke. These changes increased the protein-like substances in centrifugal solutions. The experimental results show that the internal inorganic particles with external stress could grind the sludge granules during the centrifugal dewatering process, which was good for separating permeable channels and polycrystalline reticular structures to help release the bound water. In the AFS centrifugal liquid, a fluorescent substance (Fig. 5b3) appeared but a very small amount, indicating that the effect of the block pressure on centrifugal dewatering of the AFS was not obvious. After adding 4 mg/g CPAM to the two kinds of sludge at 4,000 rpm for 20 min with block pressure centrifugation (Figs. 5a4 and b4), the small fluorescence intensity increased in the AGS centrifugal liquid; however, the fluorescence intensity in the AFS centrifugal liquid increased. Therefore, it can be inferred that the mineralized and inorganic substances of the AGS can help to break cells during centrifugation, so it would be faster and more beneficial for discarding these protein substances. Moreover, the mineralized and inorganic substances formed the dewatering passages, which were more conducive to compressing and releasing the intracellular extracellular bound water.

4. Conclusion

The AGS has a better dewatering performance than AFS. The SRF of the AGS was $(2.52 \pm 0.02) \times 10^9$ m/kg, which was much smaller than that of the AFS $((1.23 \pm 0.03) \times 10^{14}$ m/kg). Without physical and chemical treatments, the moisture content of the AGS dewatering can reach 80.2%, but external forces play a large role in the AGS dewatering. The mass dissolution of EPS is helpful for dewatering, and mechanical centrifugation could promote the release of EPS from AGS.

The AGS has a different dewatering mechanism than that of the AFS. According to the fluorescence intensity of the 3D-EEM results, the three-dimensional fluorescence spectroscopy in the AGS centrifugal liquid was higher than that in the AFS centrifugal liquid, so it is proposed that the AGS dewatering mechanism and method could be different from those of the AFS. Due to the large wet density, small specific surface area, and large inorganic particle size from 2–2.5 mm, the free water in AGS can be removed quickly under gravity. Then, under the action of pressure centrifugation, the other forms of water can be removed.

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