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Cocoamidopropyl Betaine Dosage Dependence of Short-Time Aerobic Digestion for Waste-Activated Sludge Reduction

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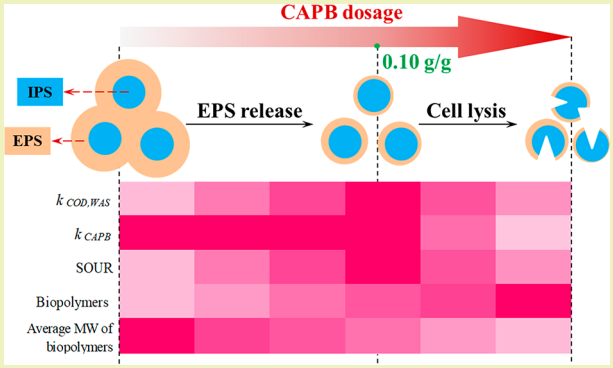
ABSTRACT: Cocoamidopropyl betaine (CAPB) has newly been found to improve the reduction of waste-activated sludge (WAS) by a short-time aerobic digestion (STAD) process. This work systematically discloses the influences of CAPB dosage on the reduction of WAS by the STAD process. Results showed that CAPB lower than 0.10 g/g of TSS (total suspended solids) mainly increase the organic substances release and low-molecular-weight (MW) fractions proportion via its surfactant action, leading to the increased concentration of soluble biopolymers. The concentration of soluble PO_4^{3-}-P and NH_4^+-N increased during the biopolymers release, but gradually decreased in the later stage. Moreover, the specific oxygen uptake rate (SOUR) of digested sludge (denoted as aerobic microorganisms activity) was increased, resulting in the increased biodegradation rate of WAS chemical oxygen demand ($k_{COD,WAS}$). A higher amount of CAPB (>0.10 g/g of TSS) led to cell lysis and intracellular polymeric substances (IPS) release, which led to an additional increase of PO_4^{3-}-P, NH_4^+-N, soluble organic matters, and its low MW fractions; all these fractions gradually decreased later. Nevertheless, cell lysis reduces both pH value and SOUR, which caused the decrease of CAPB biodegradation rate and $k_{COD,WAS}$. This study lays the foundation for improving the reduction efficiency of WAS in the STAD system by optimizing surfactant dose.



KEYWORDS: *Sludge reduction, Short-time aerobic digestion, Cocoamidopropyl betaine, Biopolymers, Cell lysis*

INTRODUCTION

Short-time aerobic digestion (STAD) was used as a unique method for waste-activated sludge (WAS) reduction, which also attracted a great deal of attention because of the convenience in operation and low capital investment.[1−3] For example, with control of the mixture's dissolved oxygen concentration at 2−3 mg/L, the volatile suspended solids (VSS) removal rate of sludge was about 18.5% after a 24-h treatment with the STAD process,[2] which is higher than the previously published results by 10−16%.[4,5] Thus, the WAS treatment cost could be decreased by reducing the dissolved oxygen demand and shortening the sludge reduction time. Especially, aerobic digestion is more suitable for sludge reduction in medium-sized or small-sized wastewater treatment plants (WWTPs),[6] meaning STAD should be suitable in China for the stabilization and reduction of WAS as over 70% of the WWTPs have wastewater processing capacities lower than 10 000 tons.[7] Thus, further shortening of the stabilization process of WAS is important and valuable for reducing the cost and achieving practical application of the STAD system in WAS reduction.

As a typical amphoteric surfactant, cocoamidopropyl betaine (CAPB) has been widely used in skin and hair care cosmetics, dishwashing agents, and technical cleaners because of their mildness to the skin and eyes combined with low toxicity and biodegradable potential.[8,9] CAPB contains a carboxylic acid anion, quaternary ammonium cation, and long alkyl chain,[8] which can also form micelles and improve the release of extracellular polymeric substances (EPS) from WAS.[10] Zhou et al.[11] found that cetyltrimethylammonium bromide could significantly improve the production of biopolymers containing proteins (PN), polysaccharides (PS), and nucleic acids (NA) from WAS. Additionally, a high dose of CAPB can produce more micelles and even led to cell lysis and the release of IPS.[2,11] Because of the special physicochemical properties, CAPB has been reported to shorten the stabilization process of WAS. Xia et al.[3] found that CAPB could significantly improve the solubilization of WAS in the STAD system by enhancing the solubilization of soluble proteins, polysaccharides, nucleic acids, humiclike substances, and the proportion of low MW fractions (<20 kDa) in the STAD system. Normally, low-molecular-weight compounds in soluble organic matters could

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Table 1. Parameters of the Linear Relationship between the CAPB Dosage and the Concentrations of Biopolymer and Its Fractions at the Noted Aerobic Digestion Times[a]

	proteins		polysaccharides		nucleic acids		biopolymers	
time (h)	K (mg/g)	R^2	K (mg/g)	R^2	K (mg/g)	R^2	K (mg/g)	R^2
0	198	0.978	51.5	0.961	34.9	0.930	285	0.9583
2	226	0.964	58.4	0.982	45.5	0.947	330	0.9616
4	228	0.975	60.7	0.970	50.3	0.985	337	0.9825
8	197	0.958	73.6	0.956	49.4	0.994	320	0.9767
12	180	0.961	68.9	0.951	49.6	0.960	299	0.955
24	147	0.952	54.7	0.950	56.7	0.969	247	0.9572

[a]K is the slope of the linear relationship between the CAPB dosage and the concentrations of biopolymer and its fractions. $y = Kx + m$, where y is the concentrations of biopolymers and its fractions, x is the dosage of CAPB, and m is the intercept.

be easily biodegraded by heterotrophic bacteria during the aerobic digestion process.[12] Meanwhile, the specific oxygen uptake rates (SOUR), dehydrogenase enzyme activity, and the proportion of some functional microorganisms including *Proteobacteria*, *Planctomycetales*, *Acinetobacter*, *Pseudomonas*, and *Aeromonas* in the WAS microbial community were also dramatically increased after the addition of 0.08 g/g of CAPB, and then the reduction efficiency of WAS was improved. CAPB could be used to improve the reduction of WAS, but high doses of CAPB could damage the cell membrane and cause intracellular polymeric substances (IPS) release, which could reduce the activity and proportion of functional bacteria and then further affect the STAD efficiency of WAS. Optimizing surfactant dose for WAS stabilization and reduction by STAD should also be valuable for reducing the treatment cost of WAS. Thus, it is important and valuable to improve the knowledge of how CAPB dosage affects the reduction of WAS by the STAD process.

In this work, we evaluated in detail how CAPB dosage impacts the reduction of WAS in the STAD system by determining the changes in key parameters, including the soluble total organic carbon (STOC), ultraviolet light intensity at 254 nm wavelength (UV_{254}), soluble PO_4^{3-}-P, NH_4^+-N, and WAS chemical oxygen demand (COD_{WAS}). On the basis of the release of organic matters and the molecular weight (MW) distribution, the variation of WAS specific oxygen uptake rate (SOUR), and the biodegradation rate of COD_{WAS} ($k_{COD,WAS}$) and CAPB (k_{CAPB}), we systematically disclosed the mechanisms on how CAPB dosage affects the STAD of WAS.

MATERIALS AND METHODS

Chemicals and WAS Samples. Cocoamidopropyl betaine ($C_{19}H_{38}N_2O_3$, CAPB, 342.52 g/mol) was purchased from Shanghai Chem. Co. Ltd., China. Sludge samples were obtained from the secondary clarifier of a municipal WWTP with the sludge retention time of 20 days in Shanghai, China. The grit in sludge was first removed by screening through a 1.2 mm sieve. Sludge samples were concentrated by sedimentation at 4 °C for 2 h. The main parameters of concentrated sludge were as follows: pH 6.8 ± 0.1; total suspended solids (TSS), 8.65 ± 0.13 g/L; VSS, 6.24 ± 0.25 g/L; total chemical oxygen demand, 12038 ± 26 mg/L; and total residual CAPB, 4.29 ± 0.4 mg/L. Concentrated sludge was stored at 4 °C and used within 48 h.

STAD of WAS and the Collection of Digested Sludge. STAD experiments were carried out in six reactors and the CAPB concentrations were controlled at 0.05, 0.08. 0.10, 0.13, and 0.15 g/g TSS (hereafter g/g), respectively, and the reactor without addition of CAPB was the control. The configuration of the reactor was described in detail in our previous studies.[1−3] The work volume of the reactor was 5.0 L, and the solution pH, temperature, and dissolved oxygen (DO) were controlled at 6.8 ± 0.1, 24 ± 2 °C, and 2−3 mg/L, respectively. We took samples that were subjected to immediate characterization without storage. Part of the sample was centrifuged at 4 °C with centrifuge rotation speed of 4000*g* for 20 min in a high-speed freezing centrifuge (Heraeus Multifuge X1R, Thermo Scientific, Germany). The supernatant was collected and centrifuged again at 12000*g* and 4 °C for 10 min to further remove particles. The collected supernatant was stored at 4 °C for the analysis of soluble biopolymers, STOC, UV_{254}, supernatant PO_4^{3-}-P and NH_4^+-N, and MW distribution. Part of the digested sludge was used to determine the pH value, SOUR, COD for the mixed culture (COD_{Total}), and total residual CAPB.

Chemical Analyses. The DO and pH of the mixed culture were measured by a DO and pH meter (HQ40d, Hatch, USA). TSS and COD_{Total} of the mixture, supernatant NH_4^+-N and PO_4^{3-}-P, and supernatant UV_{254} were analyzed using the standard methods.[13] STOC concentration was measured by an automatic TOC analyzer (TOC-V_{CSH}, Shimadzu, Japan), with potassium hydrophthalate (Shimadzu) solution as the calibrator. The MW distributions of the soluble organic matters were analyzed by a high-performance liquid chromatograph (HPLC) (LC-10AD, Shimadzu, Japan) equipped with a size exclusion chromatograph (SEC) column, and an evaporative light-scattering detector (ELSD, PL-ELS 2100, Polymer Laboratories, Sophonsire, UK) was employed to analyze the MW distribution of soluble organic matters. Soluble PN was determined by the Bradford method using bovine serum albumin as the standard.[14] soluble PS was measured by the phenol-sulfuric acid method using glucose as the standard.[14] Diphenylamine colorimetric method was used to determine the soluble NA, and calf thymus DNA was the standard.[14] A 100-mL sealed glass bottle equipped with aerator and DO probe was used to determine the oxygen uptake rate (OUR) of WAS. Sludge samples were aerated for 5 min to saturate the oxygen, and DO concentration of the mixture was continuously measured and the data recorded by a computer. The SOUR of digested sludge was calculated by the combination of linear regression analysis and sludge VSS.[15] The determination of total residual CAPB was based on the acid orange II method, which was described in detail in previous studies.[3,16]

As the residual CAPB could be part of the solution COD_{Total}, COD of WAS (COD_{WAS}) could be calculated using eq 1,

$$COD_{WAS} = COD_{Total} - COD_{CAPB} \quad (1)$$

where COD_{CAPB} is the residual concentration of CAPB present in COD. COD_{WAS} is the WAS concentration present in COD, and COD_{Total} is the total concentration of mixture containing WAS and CAPB present in COD. The conversion factor of CAPB equivalents to COD is 2.38 mg of COD/mg of CAPB.[2]

The first-order biochemical degradation kinetics was used to simulate the removal of organic matters containing CAPB and COD_{WAS},[2]

$$dC/dt = -kC \quad (2)$$

where C (mg/L) is organic matters concentration, t (h) is the reaction time, and k (h^{-1}) is the biodegradation rate constant.

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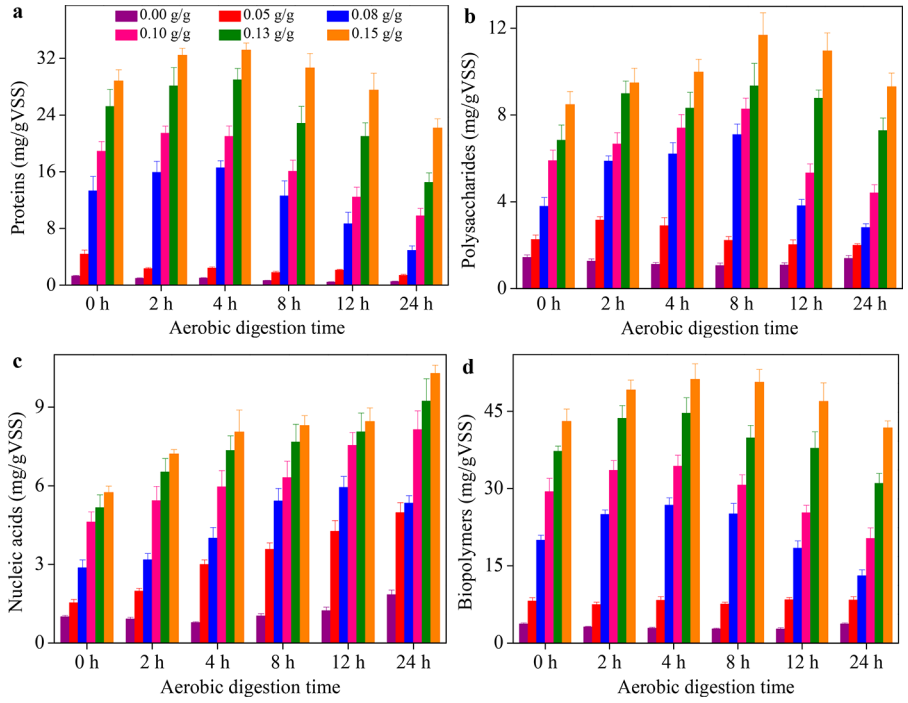

Figure 1. Effects of CAPB dosage on the production of (a) proteins, (b) polysaccharides, (c) nucleic acids, and (d) biopolymers from WAS at the noted aerobic digestion times.

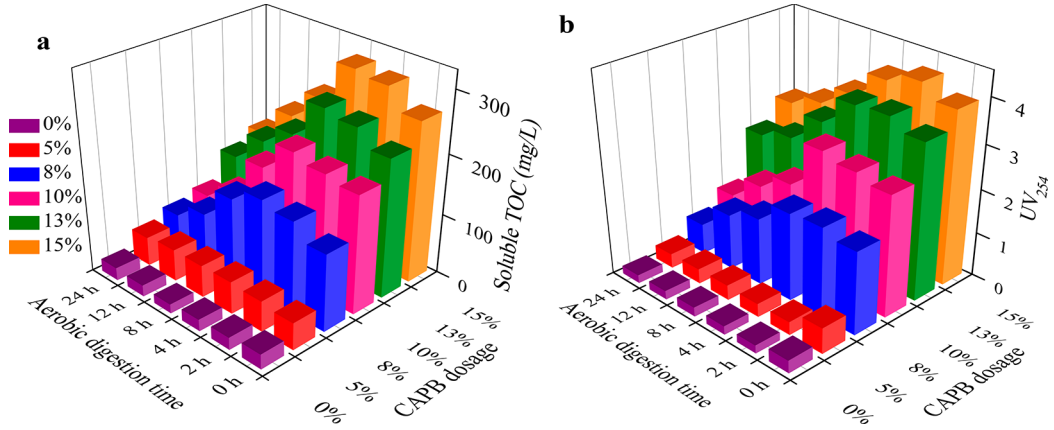

Figure 2. Effects of CAPB dosage on the (a) soluble TOC and (b) UV_{254} in the supernatant of WAS at the noted digestion times.

Statistical Analysis. We took three samples from the aerobic digestion reactor and analyzed them one time for PN, PS, NA, STOC, UV_{254}, pH, SOUR, soluble PO_4^{3-}-P and NH_4^+-N, COD, TSS, and residual CAPB for each sample. Results are shown as the mean and standard deviation (mean ± SD). One representative result was shown when presenting the MW distribution of soluble organic matters. Software of Origin 8.1.5 (Origin Lab Inc., USA) and Pearson's correlation coefficient (R^2) were used to identify or estimate the correlation between two parameters. Correlations were considered statistically significant when the confidence interval is higher than 95% ($P < 0.05$).

RESULTS AND DISCUSSION

Release of Soluble Biopolymers during the STAD of WAS. Solubilization and hydrolysis of macromolecular organic matters (MOM) from WAS led to increased soluble biopolymers, which is another key step for WAS reduction.[5,6] The contents of PN, PS, NA, and biopolymers increased dramatically by increasing CAPB dosage, which also had a strong and nearly linear relationship with the dosages of biopolymers as the value for all R^2 were higher than 0.93 (Table 1). Throughout the digestion process, the release constant (K) of organic matters dramatically increased from 0 to 4 h for PN and biopolymers, within 8 h for PS, and even higher for NA. Meanwhile, results showed that CAPB could improve the release of biopolymers from WAS. The concentration of NA kept increasing but PN and PS gradually decreased later, suggesting that PN and PS are more readily biodegraded compared to NA during the STAD process.[2,3,17]

Supernatant STOC and UV_{254} During the STAD of WAS. Soluble TOC represents total organic matters and UV_{254} reflects the humic acid-like macromolecular matters and aromatic compounds containing C═C and C═O, both of which could be used as the substrate and electron donor for heterotrophs.[18,19] Figure 2 presents the UV_{254} and STOC of soluble organic matters after addition of various dosages of CAPB at different digestion times. Both STOC and UV_{254}

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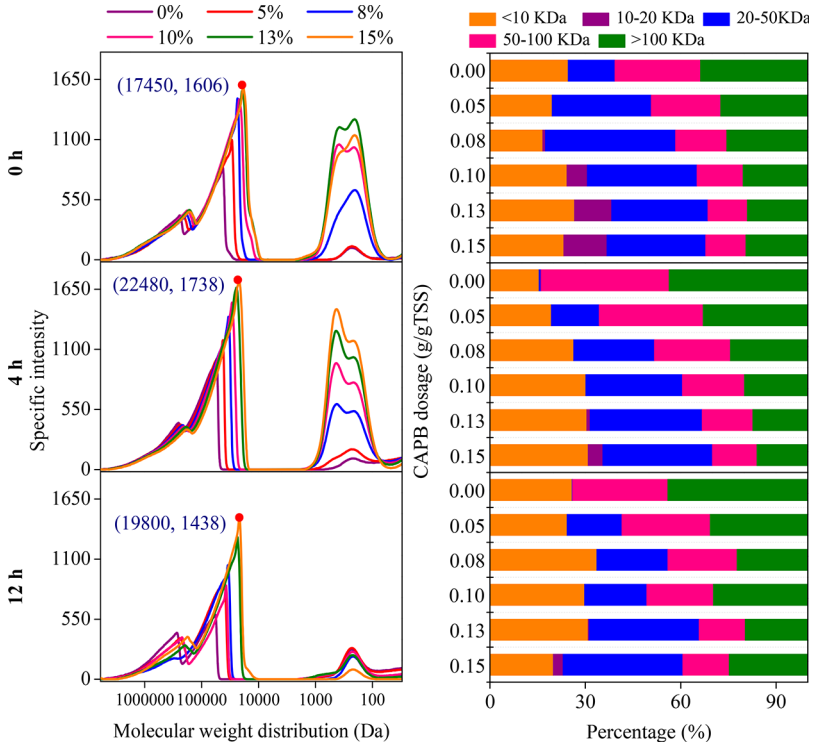

Figure 3. Effects of CAPB dosage on the MW distributions of the soluble biopolymers in supernatant of WAS at the aerobic digestion times of 0, 4, and 12 h, respectively.

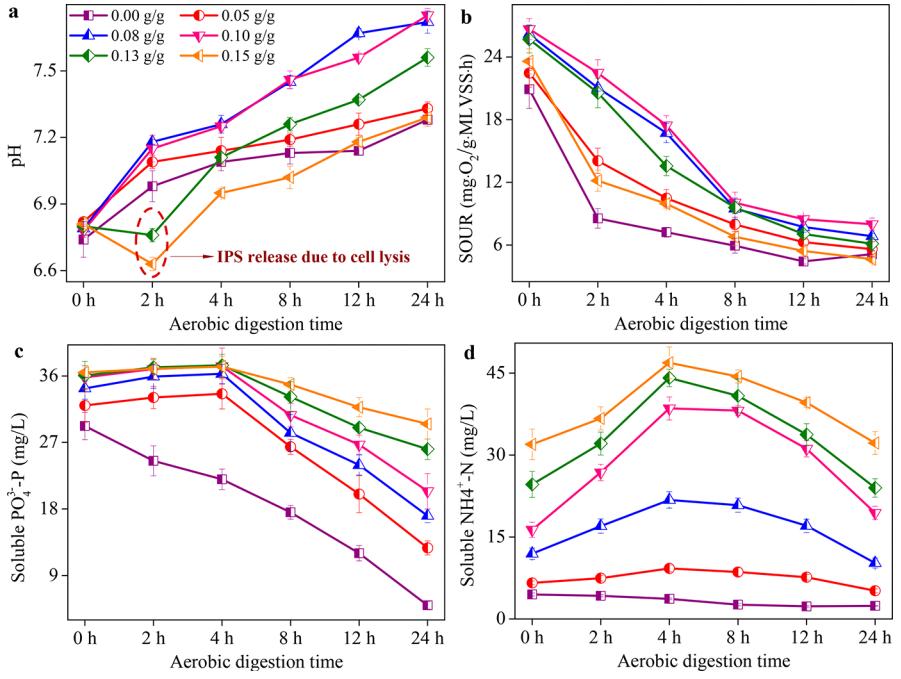

Figure 4. Effects of CAPB dosage on (a) system pH value, (b) SOUR, and the soluble (c) PO_4^{3-}-P and (d) NH_4^+-N of WAS at the noted digestion times. IPS is intracellular polymeric substances.

dramatically increased with the increased CAPB dosage, which was consistent with the biopolymers release after addition of CAPB in Figure 1. Throughout the digestion process without addition of CAPB, both UV_{254} and STOC gradually decreased from 0 to 8 h, which should be attributed to the fact that soluble biopolymers could be the electron-donor substrates and excellent carbon sources for heterotrophs.[20] In the later stage, depletion of biodegradable carbon sources could cause the EPS solubilization or even cell lysis,[4,5] and the subsequent increase in STOC and UV_{254}. After addition of CAPB, UV_{254} and STOC kept increasing in the first 8 h because the nonpolar linear hydrocarbon groups in CAPB can form micelles and improve the solubilization of biopolymers.[21] Thereafter, the released soluble organic matters could be gradually biodegraded by heterotrophs.[20]

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MW Distribution of Soluble Organic Matters. The MW distribution of soluble matters could influence their biodegradation by heterotrophs.[12] Figure 3 presents the MW distributions of soluble organic matters after addition of various dosages of CAPB and aerobic digestion for 0, 4, and 12 h. The area and specific intensity for all peaks dramatically increased with the increasing of CAPB dosage, and the peaks with MW between 10 and 100 kDa gradually moved to the right at the noted times, meaning the release of low MW biopolymers. Without addition of CAPB, the peak's area continuously decreased, and the proportion of low MW fractions also reduced throughout the digestion process, suggesting the continuous biodegradation of low MW fractions in biopolymers during the STAD process.[3,6] After addition of CAPB, both the proportion and concentration of low MW fractions kept increasing in the initial 4 h, but decreased later. Thus, CAPB could improve biopolymers solubilization and increase the low MW fractions proportion at the beginning of the digestion process; however, it gradually decreased with the increased digestion time, indicating soluble organic matter could be degraded by heterotrophs.[22] Thus, CAPB should be an excellent chemical agent to improve the reduction performance of WAS during the STAD process.

pH and SOUR of the Mixed Culture. Figure 4a presents the system pH value after addition of various dosages of CAPB at the noted digestion times. With addition of CAPB from 0 to 0.10 g/g, the system pH value gradually increased and tended to be alkalescent throughout the STAD process, which was consistent with previous studies.[2,5] Additionally, a high dose of CAPB caused a higher system pH value but then remained stable at about 7.8 when CAPB dosage increased from 0.08 to 0.10 g/g. With a higher CAPB dosage (>0.10 g/g), the system pH value decreased in the initial 2 h and a high CAPB dosage led to a lower pH value, which might result from the cell lysis and the release of intracellular NA after addition of a high dose of surfactant.[11,23] Thereafter, system pH value kept increasing and showed a similar trend when a low dose of CAPB was added to the system.

As fundamental indices of microbial growth, SOUR is commonly used to evaluate aerobic microorganism activities in WAS.[24] Figure 4b shows the SOUR of WAS after various doses of CAPB were added at the noted digestion times. SOUR kept increasing as CAPB dosage increased from 0 to 0.10 g/g, but it reduced for a higher CAPB dosage at all the noted digestion times. During the STAD process, the SOUR of WAS for all of the CAPB dosages gradually decreased. Results showed that CAPB dosage from 0 to 0.10 g/g could significantly improve the activities of aerobic microorganisms and then improve the biodegradation of organic matters. However, high dosage (>0.10 g/g) of CAPB caused the cell lysis and reduced the aerobic microorganism activities, which further reduced the reduction efficiency of WAS.

Supernatant PO_4^{3-}-P and NH_4^+-N. Figure 4c presents the concentration of soluble PO_4^{3-}-P after various doses of CAPB were added at various digestion times. Throughout the digestion process without addition of CAPB, the concentration of soluble PO_4^{3-}-P decreased gradually, which should result from the accumulation of phosphorus inside the cell and EPS adsorption due to the functional groups of quaternary ammonium ($-NH_3^+$).[19] After addition of CAPB, soluble PO_4^{3-}-P concentration slightly increased in the first 4 h because of the release of adsorbed PO_4^{3-}-P in EPS during the biopolymers solubilization process.[2] In the later stage, soluble PO_4^{3-}-P gradually decreased, resulting from the uptake and storage of PO_4^{3-}-P by EPS and phosphorus-accumulating organisms (PAOs), respectively.[2,25]

Figure 4d presents the soluble NH_4^+-N concentration after a serial dosage of CAPB was added in the STAD process. Without addition of CAPB, soluble NH_4^+-N gradually decreased, which should be due to the oxidization of NH_4^+-N by ammonia-oxidizing bacteria.[26] After addition of CAPB, supernatant NH_4^+-N increased initially, which was caused by the biopolymers solubilization and the release of stored NH_4^+-N that could be taken up by the negatively charged functional groups in biopolymers.[27,28] In the later stage, soluble NH_4^+-N gradually decreased because of the reuse of ammonia by heterotrophs.[29] Additionally, the system value continued to increase and tended to be alkalescent in this stage, leading to the continuous stripping of NH_4^+-N in the aeration process,[4] which also caused the decrease of soluble NH_4^+-N.

Biodegradation of COD_{WAS} and CAPB in the STAD System. Figure 5 presents the biodegradation of CAPB and

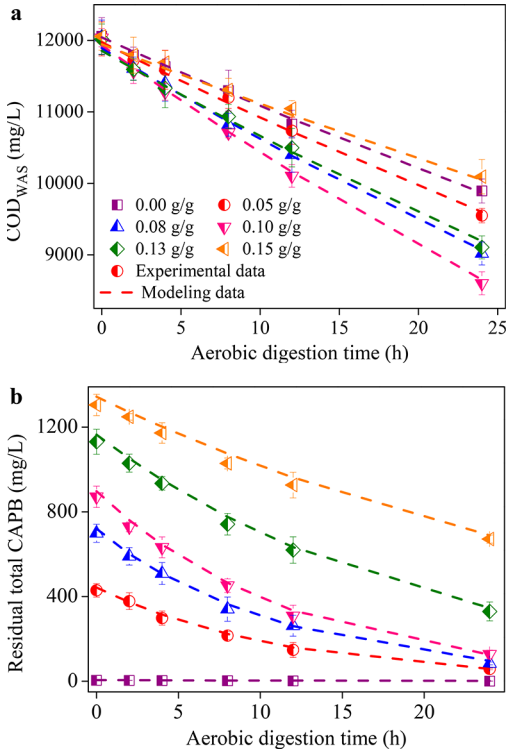

Figure 5. (a) Biodegradation of COD_{WAS} and (b) residual total CAPB under various CAPB dosages at the noted digestion times.

COD_{WAS} after various dosages of CAPB were added during the STAD of WAS; Table 2 shows the biodegradation rate constants of COD_{WAS} and CAPB. COD_{WAS} kept decreasing at all conditions, which could be simulated well by the degradation kinetics as the correlation coefficients (R^2) for all experiments were higher than 0.95. Moreover, the biodegradation rate of COD_{WAS} ($k_{COD,WAS}$) kept increasing from 0.0083 to 0.0134 h^{-1} when CAPB dose increased from 0 to 0.10 g/g, but which continuously declined with higher CAPB dose. After addition of 0.10 g/g CAPB, the COD_{WAS} removal efficiency of sludge was about 28.5% after treatment for 24 h with the STAD process, which is higher than the previously published results that were lower than 18%.[5,30−33] Results showed that a low dose of CAPB could improve the

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Table 2. Removal Constants of COD_{WAS} and CAPB during the STAD of WAS Using the First-Order Biochemical Degradation Kinetics at Various CAPB Dosages

	COD_{WAS}		CAPB	
CAPB dosage (g/g)	$k_{COD,WAS}$ (h^{-1})	R^2	k_{CAPB} (h^{-1})	R^2
0	0.0083	0.968	0.0866	0.981
0.05	0.0092	0.972	0.0858	0.966
0.08	0.0113	0.978	0.0841	0.972
0.10	0.0134	0.968	0.0827	0.976
0.13	0.0106	0.954	0.0508	0.985
0.15	0.0072	0.971	0.0279	0.980

aerobic microorganisms activity (denoted as SOUR), leading to a high biodegradation rate of COD_{WAS} ($k_{COD,WAS}$). However, a high dose of CAPB could damage the cell membrane; the reduced amount of active bacteria also led to the decreased $k_{COD,WAS}$.

For the biodegradation of CAPB (Figure 5b), all of the residual CAPB continued to decrease and meanwhile CAPB removal could be simulated well by the degradation kinetics because all R^2 values were higher than 0.96 (Table 2). Moreover, CAPB biodegradation rate (k_{CAPB}) showed a slight decrease as CAPB dosage gradually increased from 0 to 0.10 g/g, but dramatically decreased at a higher CAPB dosage. Results showed that a low dose of CAPB has only a minimal effect on the CAPB biodegradation bacterial activity, but cell lysis occurred at a higher dosage of CAPB, leading to the decrease of CAPB biodegradation bacteria and k_{CAPB}.

Influential Mechanisms of CAPB on the Reduction of WAS. Figure 6 synthesizes the mechanisms of how CAPB dosage affected the STAD of WAS. At low doses (≤0.10 g/g), CAPB mainly lead to the release of macromolecular organic matters from WAS via its surfactant action, which further resulted in the increased concentration of STOC and UV_{254} (Figure 2) and soluble biopolymers and its low MW fractions (Figure 1 and 3). Meanwhile, biopolymers release also caused the increased concentration of soluble PO_4^{3-}-P and NH_4^+-N within the first 4 h (Figure 4), but both of them continuously decreased later. Moreover, SOUR of WAS also increased after CAPB was added (Figure 4), leading to the increased $k_{COD,WAS}$ (Table 2). Higher CAPB (>0.10 g/g) caused cell lysis, leading to further increased concentration of PO_4^{3-}-P, NH_4^+-N, UV_{254}, STOC, and the fractions of soluble organic matter with low MW. However, cell lysis also led to the decrease of the system pH value (Figure 4) and SOUR (Figure 5), which caused the decrease of the biodegradation rate for both COD_{WAS} ($k_{COD,WAS}$) and CAPB (k_{CAPB}) (Table 2).

Significance. For environmental sustainability, wastewater after the stabilization and reduction of WAS could contain surfactant, which will seriously threaten the environment and human beings when discharged arbitrarily. A biodegradable cationic surfactant, CAPB, was selected in this work,[2] and 85.4% of CAPB could be removed after 1 day of STAD; in addition, the quaternary ammonium compound could be continuously biodegraded by an oxygen-based membrane biofilm reactor (O_2-MBfR).[34] Thus, biodegradation of CAPB should prevent its discharge into aquatic environments and avoid secondary pollution.

CONCLUSIONS

STAD combined with CAPB could be a unique method for the reduction of WAS. CAPB doses lower than 0.10 g/g could significantly improve the biopolymers solubilization and the low MW fractions, and increase PO_4^{3-}-P, NH_4^+-N, STOC, and UV_{254} concentrations; all of them could be further biodegraded or reused by functional microorganisms during the aerobic digestion process. Moreover, the activity of aerobic microorganisms (denoted as SOUR) also increased, leading to an increased COD_{WAS} biodegradation rate ($k_{COD,WAS}$). However, higher CAPB (>0.10 g/g) led to cell lysis, resulting in the decrease of system pH value and SOUR of WAS, and reduction of the biodegradation rate for both COD_{WAS} and CAPB. Without obvious cell lysis, a modest dose of CAPB (0.10 g/g) could achieve the maximum WAS reduction and low residual CAPB.

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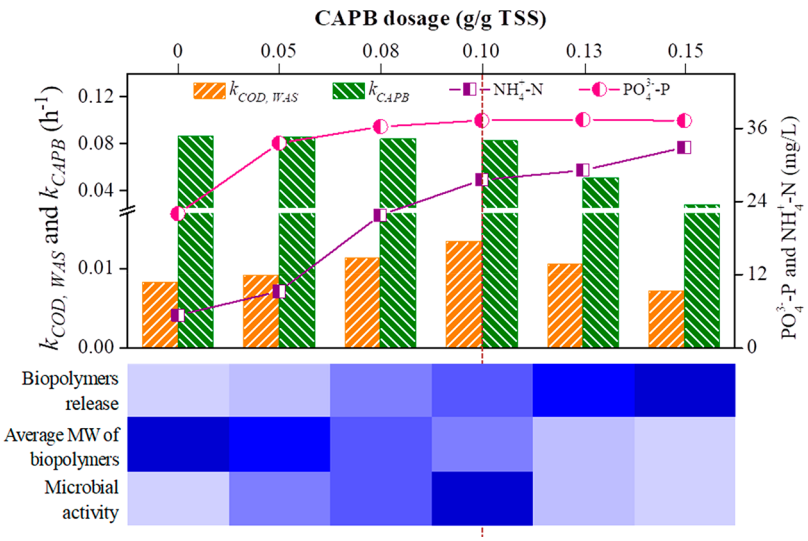

Figure 6. Synthesis of mechanisms about how CAPB affects the short-time aerobic digestion (STAD) of waste-activated sludge (WAS). The deeper color and white color in a bar signify the high and low value of the parameters, respectively.

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Notes

The authors declare no competing financial interest.

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■ ABBREVIATIONS

CAPB, cocoamidopropyl betaine; CTAB, cetyl trimethylammonium bromide; DO, dissolved oxygen; EPS, extracellular polymeric substances; IPS, intracellular polymeric substances; $k_{COD,WAS}$, biodegradation rate of waste-activated sludge present in COD; k_{CAPB}, biodegradation rate of CAPB; MOM, macromolecular organic matters; MW, molecular weight; NA, nucleic acids; PAOs, phosphorus-accumulating organisms; PN, proteins; PS, polysaccharides; SOUR, specific oxygen uptake rate; STAD, short-time aerobic digestion; STOC, soluble total organic carbon; COD, chemical oxygen demand; TSS, total suspended solids; WAS, waste activated sludge; WWTPs, wastewater treatment plants

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