Evaluating the excess sludge reduction in activated sludge system with ultrasonic treatment

Shuang Wu, Min Zheng, Qian Dong, Yanchen Liu and Chengwen Wang

ABSTRACT

Ultrasonic treatment for enhancing biological processes has recently attracted considerable attention in wastewater treatment. In this study, we systematically investigated the mixed liquor properties of activated sludge under ultrasonic treatment. The sludge samples were collected from the aerobic tank of a full-scale membrane bioreactor (MBR) treating municipal wastewater, and the volatile suspended solids (VSS) concentration was approximately 6.0 g/L. The results showed that ultrasonic treatment induced floc disintegration, organics release, temperature increase, microbial activity and pH variation. The maximum mg soluble chemical oxygen demand (COD) per mg VSS released was estimated to be 0.147 using the Monod equation. The exponential increase in the concentration of dissolved organic matter is related to the loss of relative heterotrophic bacterial activity. A sonolysis-cryptic growth model was demonstrated to be capable of describing ultrasonic sludge reduction, which would support the further development of ultrasonic treatment technology in activated sludge systems.

Key words | excess sludge reduction, floc disintegration, organics release, ultrasonic treatment

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INTRODUCTION

Ultrasound is a commonly used oscillating sound pressure wave with a frequency above 20 kHz. Sonobioreactors, which are bioreactors that employ ultrasound for sludge treatment, have been considered a 'green' technology owing to their high efficiency, low instrumental requirements, significantly reduced process time compared with other conventional techniques, and economically viable performance (Rokhina *et al.* 2009; Zhang *et al.* 2012).

Ultrasound is commonly recognized as a nonthermal sterilization technique. Ultrasonic treatment of activated sludge incurs floc disintegration, cell lysis and inactivation (Zhang *et al.* 2007), and ultrasonic treatment of waste sludge enhances methane production during the anaerobic digestion process (Pilli *et al.* 2011). Low-intensity ultrasound has been found to enhance net biofilm growth because the low intensity increases the rate of transport of oxygen and nutrients to the cells as well as the rate of transport of waste products away from the cells (Pitt & Ross 2003). Therefore, the use of low-intensity ultrasound could improve the overall reactor performance in activated sludge systems, including nitrogen removal (Zhang *et al.* 2011; Zheng *et al.* 2013a, 2016) and excess sludge reduction (Li *et al.* 2008; Zhang *et al.* 2009). The sonolysis-cryptic growth process, that is, when the contents and nutrients stored in the activated sludge are released and enter into the metabolism cycle of other microorganisms, has been considered a key process to achieving ultrasonic sludge reduction (Mohammadi *et al.* 2011). However, no information evaluating this process has been reported thus far.

This study aims to systematically investigate the mixed liquor properties (floc disintegration, organics release and microbial activity loss) of activated sludge under ultrasonic treatment. Activated sludge samples were collected from a full-scale membrane bioreactor (MBR) treating municipal wastewater. Groups of batch experiments with different ultrasonic intensities were performed on the collected activated sludge. The particle size distribution, release and aerobic degradation of dissolved and colloidal organic matter, activity of heterotrophic bacteria, temperature and pH were analyzed. Based on the results, a sonolysis-cryptic growth model was proposed to investigate the effects of ultrasonic treatment on excess sludge reduction. The model was demonstrated to be capable of describing ultrasonic sludge reduction, which would support the further development of ultrasonic treatment technology in activated sludge systems.

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MATERIALS AND METHODS

Collection of sludge samples

Mixed liquor samples were collected from the aerobic tank of a full-scale membrane bioreactor treating municipal wastewater, located on the campus of Tsinghua University, Beijing, China. The sludge retention time was approximately 25–40 days. Long-term operation of the MBR process showed good performance of organics and nitrogen removal, comparable with the results in our previous reports (Zheng *et al.* 2013b, 2014). The mixed liquor suspended solids (MLSS) concentration of the collected sludge was in the range of 10.0–12.0 g/L, and the concentration of volatile suspended solids (VSS) was approximately 6.0 g/L.

Ultrasonic treatment experiment

Ultrasonic treatment experiments were conducted at ambient temperature (24–26 °C) in reactors made from Plexiglas cylinders. The collected sludge samples were added to the reactors, stirred and then treated using an ultrasound generator (SCIENTZ–IID, Ningbo Xinzhi Co., Ltd, variable parameters in the ranges of 9.5–950 W, 0–999 min, 20–25 kHz). The ultrasonic treatment was performed at various power levels ranging from 40 W to 300 W. Particle size distribution, concentration of released soluble organic matter, activity of heterotrophic bacteria, temperature and pH were analyzed periodically.

The ultrasonic intensity, expressed as energy density (E_S, kJ/mL), was calculated in Equation (1):

$$E_s = \frac{P \cdot t}{1000V} \tag{1}$$

where P is the ultrasonic active power (W); t is the sonication time (s); and V is the effective volume of the ultrasonic treatment unit (mL).

Analytical methods

Chemical analysis and calculation

Mixed liquor samples were centrifuged at 10,000 r/min for 10 min. The supernatant was collected and filtered through a 0.45 μm filter. Measurements of chemical oxygen demand (COD) total nitrogen (TN), and MLSS and VSS concentrations were performed in accordance with standard methods (Ministry of Environmental Protection 2006). Temperature, dissolved oxygen (DO) and pH were autorecorded using a pH probe and meter (WTW, pH/Oxi340i). Mixed liquor samples were pretreated to determine total (TOC), soluble (DOC), and colloidal (COC) organic carbon concentrations in the supernatant. The pretreatment procedure to remove colloidal organic matter was as follows: 250 mg/L $Al_2(SO_4)_3$ solution was added to the supernatant, stirred for 10 min for flocculation, and then centrifuged for 10 min at 5,000 r/min. The organic carbon concentration was measured using a total organic carbon analyzer (Shimadzu TOC-5000A, Japan). The COC concentration is calculated as the measured TOC concentration minus the DOC concentration.

Particle size distribution of flocs

The particle size distribution of sludge flocs was determined by a particle size analyzer (Beckman LS13320/ULM2). The D_{10}, D_{50} and D_{90} parameters represent the corresponding particle size when the percentage of the cumulative particle size distribution reached 10%, 50% and 90%, respectively.

Fluorescence measurements

Fluorescence spectroscopic analysis was carried out on a Cary Eclipse fluorescence spectrophotometer (Hitachi F-7000). The sample pretreatment procedure consisted of 0.45 μm and 0.22 μm membrane filtration. Filtration with a 0.22 μm membrane is used to remove colloidal material and reduce interference of colloidal material in spectral analysis.

Heterotrophic activity assay

The endogenous respiration activity of heterotrophic bacteria was characterized using the oxygen uptake rate. The sludge samples (10–200 mL) were centrifuged at 1,000 r/min for 1 min, and the concentrated sludge was transferred to an iodine bottle with an effective volume of 300 mL. Then, oxygen-rich water was added into the iodine bottle until full. The temperature was controlled at 25 ± 1 °C. The DO concentration was autorecorded every 10 s. The slope of the measured DO versus time, calculated by linear fitting, represents the activity of heterotrophic bacteria under endogenous respiration conditions. The relative activity r_{XH}, represented by the ratio of activity with ultrasound energy density to initial activity (without ultrasound), was normalized by the activity in the presence of ultrasonic treatment versus that without ultrasound.

RESULTS AND DISCUSSION

Effect of ultrasonic energy density on floc disintegration

Flocculation structure plays an important role in sludge settleability in activated sludge systems. Table 1 summarizes the particle size distribution parameters of ultrasonically treated activated sludge. The average particle size, D_{10}, D_{50} and D_{90} parameters of the initial sludge were 54.92 μm, 13.00 μm, 40.26 μm, and 97.04 μm, respectively, indicating a good distribution. These parameters all significantly decreased when ultrasonic energy density increased to 0.6 kJ/mL. Further increasing the ultrasonic intensity caused different effects on these parameters. For example, the D_{90} increased, up to 69.59 μm, while the D_{10} continued to decrease, down to 1.28 μm, under the condition of $E_S =$ 2.0 kJ/mL. The particle size distribution width, represented by the ratio of (D_{90} − D_{10}) to D_{50}, gradually increased with increased ultrasonic intensity. The width reached a maximum value of approximately 6.0 at a set E_S of 2.0 kJ/mL and remained the same regardless of further increase in the ultrasonic intensity (Figure 1). The results indicated the ultrasonic destruction of sludge floc structure, consistent with the results reported in the literature (Zhang *et al.* 2007; Pilli *et al.* 2011).

Effect of ultrasonic energy density on organics release

Figure 2 shows organics release in the ultrasonic treatment experiments with different energy densities. The ultrasonic power and sonication time were set in the range of 50–300 W and 0–30 min, respectively. The results showed that organic matter was released into the supernatant under the treatment. The released soluble COD (mg) per VSS (mg) gradually increased with increasing ultrasonic intensity. Nonlinear regression plotting results showed that the increase in SCOD versus E_S could be well described by the Monod model (R-squared value of 0.97) in the following:

$$\frac{\Delta \text{SCOD}}{\text{VSS}} = \frac{0.147 \cdot E_S}{2.35 + E_S} \tag{2}$$

The maximum increase in SCOD was estimated to be 0.147 mg COD/mg VSS. The half-saturation constant was estimated as 2.35 kJ/mL, indicating that the increase in ultrasonic intensity would significantly affect soluble organic

Table 1 | Particle size distribution parameters of the collected sludge with ultrasonic treatment

E_S (kJ/mL)	Average particle size (μm)	D_{10} (μm)	D_{50} (μm)	D_{90} (μm)
0.00	54.92	13.00	40.26	97.04
0.01	42.64	11.14	34.94	81.42
0.02	43.55	10.06	32.09	80.51
0.10	24.85	6.06	18.83	45.49
0.20	20.48	4.41	14.28	37.65
0.40	15.79	2.72	10.48	33.25
0.60	15.01	2.19	9.27	33.20
1.20	21.40	2.59	10.65	58.81
2.00	24.06	1.28	11.50	69.59
4.00	13.81	1.14	6.50	40.88

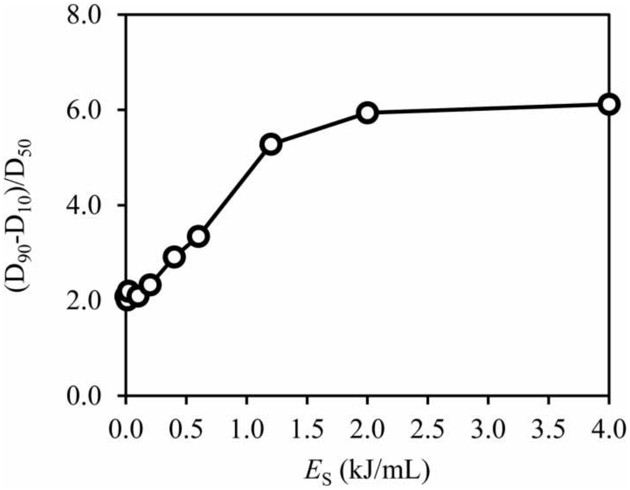


Figure 1 | Particle size distribution width at different ultrasonic energy density values (E_S).

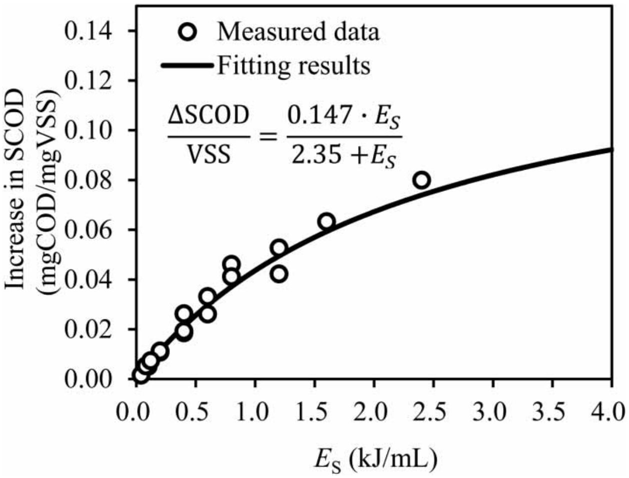


Figure 2 | Soluble organic matter release under ultrasonic treatment at different ultrasonic energy densities (E_S). The Monod equation was used to fit the measured data.

matter release. The ultrasonic energy efficiency was also calculated in terms of g COD per kJ energy input based on the results above. The results showed that the energy efficiency decreased from 358.7 to 201.2 g COD/kJ when the power increased from 50 to 300 W. Therefore, to achieve more organics release, ultrasonic treatment with relatively low power and long sonication time should be more efficient than a treatment with the same energy input at higher power and shorter time. This is consistent with the results reported by Li *et al.* (2009). The energy efficiency significantly increased as the initial VSS concentration of the treated sludge increased. The SCOD/TN ratio of released organic matter was as high as 5.5–8.2. This indicated that the released carbon source should be favourable for enhancing biological nitrogen removal in wastewater treatment.

Under ultrasonic conditions with E_S in the range of 0.2–0.9 kJ/mL, dissolved organic matter released from the treated sludge showed two characteristic fluorescence peaks. The maximum peak of Ex/Em was 270/340 nm, which represents proteins. The second peak Ex/Em wavelength was 360/440 nm, representing humic acids. The main peak was very close to the peak of microbial extracellular polymer (EPS), which has a maximum wavelength of 280/340 nm (Ni *et al.* 2009). These results indicated that the soluble organic matter released during ultrasonic treatment might be related to the released EPS via floc disintegration.

Relationship between heterotrophic activity loss and organics release

The concentration of released soluble organic matter increased with a decrease in heterotrophic bacterial activity (Figure 3). The increase in SCOD was as low as 0.03 mg COD per mg VSS when the loss of heterotrophic bacterial activity reached 50%. Further decreasing the relative activity of the heterotrophic bacteria down to 30% robustly improved the organics release.

The relationship can be described by a power function equation as follows:

$$\frac{\Delta \text{SCOD}}{\text{VSS}} = 189.5 \cdot \text{r}_{\text{XH}}{}^{-2.284} \quad (3)$$

Statistical analysis showed that the deviation between the measured data and fitting results was acceptable ($p <$ 0.05); this is a good indication that the released organics were from the lysed cells of heterotrophic bacteria.

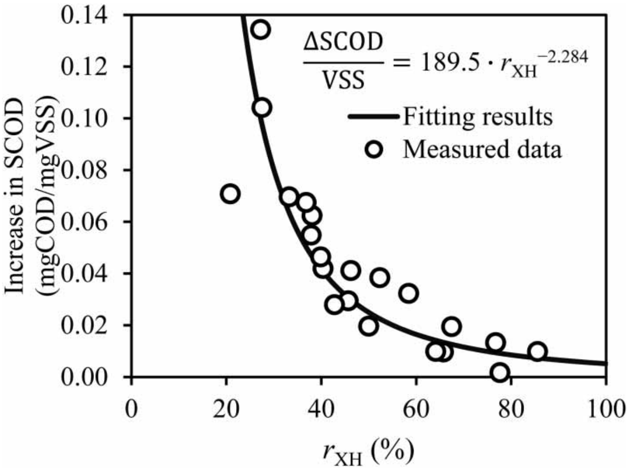


Figure 3 | Relationship between relative heterotrophic activity loss and organics release.

Previous studies showed that relatively low-intensity ultrasonic treatment enhanced microbial activity, whereas treatment with relatively high intensity inactivated microbes. In this study, we found that ultrasonic treatment with energy densities above 0.17–0.50 kJ/mL began to inactivate heterotrophic bacteria. These ultrasonic energy densities are comparable with the values of 0.18–0.48 kJ/mL corresponding to sludge disintegration of approximately 35% in the literature (Li *et al.* 2009).

Changes in released organic matter with aeration

The aerobic biodegradability and flocculation of the released organic matter in ultrasonically treated sludge samples were analyzed, as shown in Figure 4.

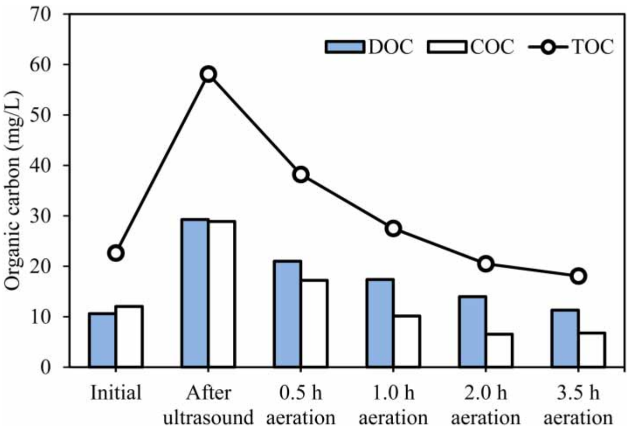


Figure 4 | Variations of supernatant total, dissolved and colloidal organic carbon concentrations of ultrasonically treated activated sludge with aeration. Ultrasonic conditions: $E_S = 0.91$ kJ/mL, $P = 38$ W and $t = 2$ h.

Initially, DOC and COC concentrations in the supernatant of the collected samples were 10.6 mg/L and 12.0 mg/L, respectively. After ultrasonic treatment, the concentrations were both increased by approximately 30.0 mg/L. With aeration of 3.5 h, the DOC concentration decreased to the initial level. The aerobic degradation rate of the released dissolved organic matter was 0.37–0.41 mg C/(g MLSS·h). This is likely because organic species with molecular weights of less than 2 kDa were the most abundant in the ultrasonically treated sludge samples (Wang *et al.* 2010). The concentration of COC was restored to the initial level within aeration times less than 1.0 h. The time was much shorter than that for the DOC. In general, colloidal flocculation is faster than the biological degradation of organic matter (Huang & Wu 2008). The results indicated that the floc structure of activated sludge with ultrasonic treatment could be aerobically restored.

Thermal effect and pH variation of ultrasonically treated activated sludge

Ultrasonic transmission in aqueous solutions has a thermal effect. The thermal effect can convert part of the energy into heat and cause an increase in liquid temperature. High temperatures significantly affect microbial activity. When the temperature exceeds 40 °C, microbial enzymes can be permanently inactivated, leading to cell lysis. In this work, the conversion ratio of ultrasonic energy to heat was analyzed using Equation (4):

$$Q = \mathrm{c} \cdot m \cdot \Delta T \tag{4}$$

where Q (kJ) is the calorific value; m is the mass of aqueous solution (kg); ΔT (°C) is the increase in temperature; and c is the specific heat capacity of an aqueous solution, with a constant value of 4.2 kJ/(kg·°C).

The initial temperature of the sludge was ambient temperature. The temperature of the treated sludge increased with increasing sonication time. According to the evaluation in Equation (4), the Q values of the mixed liquor were only in the range of 0.16–9.1 kJ in all ultrasonic treatment experiments, with total ultrasonic energy input around 1.9–342.0 kJ. Therefore, thermal energy conversion was stabilized at a low level, below 10%, during the ultrasonic treatment of sludge.

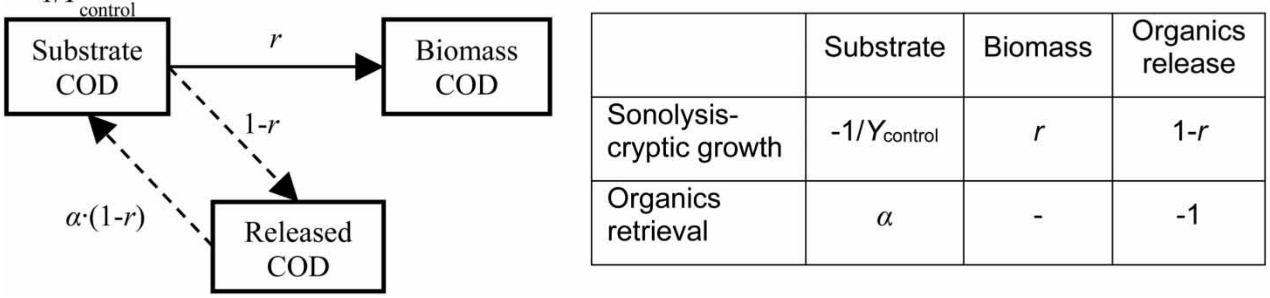
Due to the organics release, the pH of the mixed liquor gradually decreased as the ultrasonic energy density increased. However, the pH was still above 7.0 in all experiments at E_S less than 2.5 kJ/mL. Therefore, the pH variation of the treated sludge should be negligible in activated sludge systems treating municipal wastewater.

Modeling the excess sludge reduction with ultrasonic treatment

The results above clearly demonstrate that ultrasonic treatment would cause floc disintegration, cell lysis, and biodegradable organic matter release, but floc structure could be restored with aeration. Due to the degradation of released organic matter, ultrasonic treatment should induce excess sludge reduction, that is, the contents and nutrients stored in activated sludge are released and enter the metabolic cycles of other microorganisms. Hereby, we proposed a steady-state sonolysis-cryptic growth process that can be used to investigate the effects of ultrasonic treatment on excess sludge reduction, as shown in Figure 5. The sludge reduction yield is calculated as follows:

$$Y_{\mathrm{ultrasound}} = \frac{r}{(1/Y_{\mathrm{control}}) + \alpha \cdot (1 - r)} \tag{5}$$

where $Y_{\mathrm{ultrasound}}$ (g VSS/g COD) is the bioyield in a bioreactor with ultrasonic treatment; Y_{control} (g VSS/g COD)



	Substrate	Biomass	Organics release
Sonolysis-cryptic growth	$-1/Y_{\mathrm{control}}$	r	1-r
Organics retrieval	α	-	-1

Figure 5 | Steady-state sonolysis-cryptic growth model.

is the bioyield in the reactor without treatment; r is the relative activity (equal to r_{XH}); and α is the conversion coefficient of biodegradable portion to total released organics (nondimensional).

Therefore, the biomass reduction percentage can be calculated in Equation (6).

$$\text{Biomass reduction} = \frac{Y_{\text{control}} - Y_{\text{ultrasound}}}{Y_{\text{control}}} \cdot 100\% \qquad (6)$$

Using Equations (2)–(6), we showed that ultrasonic sludge reduction gradually increased with increasing energy density (Figure 6). The biomass reduction could reach 60–70% at $E_S = 1.0–1.5$ kJ/mL. The result is comparable with the sludge reduction ratios of approximately 67.6%–78% obtained by optimization of ultrasonic conditions in an activated sludge system (He *et al.* 2011; Mohammadi *et al.* 2011). Sensitivity analysis showed that the conversion coefficient α was a non-sensitive parameter in the steady-state sonolysis-cryptic growth model. The α value varied in the range from 0.4 to 1.0 and had almost no effect on the model output of sludge reduction. The model output results also agreed well with the excess sludge reduction results collected from long-term bioreactor operational data (Li *et al.* 2008; Zhang *et al.* 2009; Zheng *et al.* 2013a). Therefore, the proposed model could be useful for describing ultrasonic sludge reduction and supporting the further development of ultrasonic treatment technology in activated sludge systems.

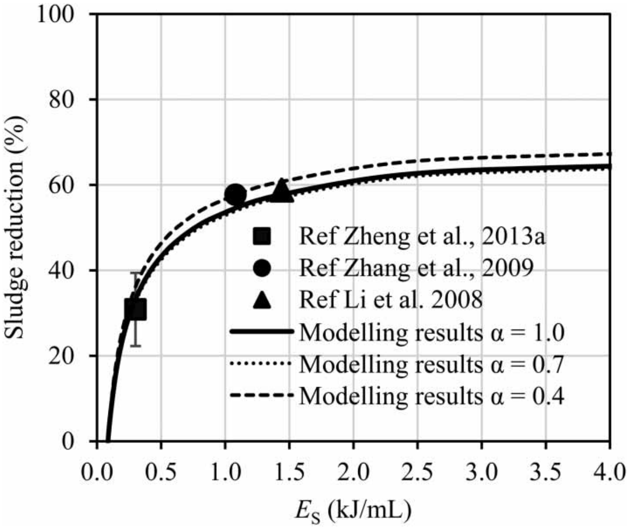


Figure 6 | Comparison between model output and sludge reduction data reported in the literature ($Y_{\text{control}} = 0.26$ g VSS/g COD).

CONCLUSIONS

Ultrasonic treatment experiments were performed on activated sludge samples to investigate the effects of ultrasonic intensity on mixed liquor properties, including floc disintegration, organics release, activity of heterotrophic bacteria, temperature and pH. The particle size distribution width and the concentration of released soluble organic matter increased with increasing ultrasonic intensity up to 2.0 kJ/mL. Further increasing the ultrasonic intensity only increased the concentration of released organic matter. The released dissolved organic matter was easily biodegradable, and the floc structure of the treated sludge could be aerobically restored. The thermal conversion of ultrasonic energy to heat was below 10%. The pH was still above 7.0 after ultrasonic treatment with intensity up to 2.5 kJ/mL. A sonolysis-cryptic growth model was demonstrated to be capable of describing ultrasonic-induced sludge reduction, which would support the further development of ultrasonic treatment technology for activated sludge systems.

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