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Characteristics of sludge hydrolysis by ultrasound and thermal pretreatment at low temperature

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Abstract−Ultrasonic treatment and thermal treatment at low temperature were employed together to analyze and compare the effect of temperature on ultrasonic sludge hydrolysis. Waste activated sludge was more susceptible to ultrasound than anaerobic sludge and primary sludge. In ultrasonic treatment of waste activated sludge for 1 hour, ΔSCOD/(−ΔVSS) ratio decreased from 2.40 to 0.44, indicating that high COD components were solubilized faster than the low COD components. Ultrasonic treatment increased the temperature significantly and the heat effect on sludge hydrolysis was not negligible. Primary sludge was more susceptible to heat than waste activated sludge. A sequential treatment of heat and ultrasound of primary sludge showed that hydrolysis efficiency was more affected by the ultrasonic power than the temperature and the time duration. In case of waste activated sludge, the overall hydrolysis efficiency increased with the temperature up to 50 °C, and it remained almost constant at higher temperature. From the results the contribution of shear force by cavitation bubbles decreased at higher temperature. The effects of shear and heat in ultrasonic sludge treatment need to be analyzed separately for the optimum sludge pretreatment.

Key words: Pretreatment, Sludge Hydrolysis, Solubilization, Thermal Treatment, Ultrasonic Treatment

INTRODUCTION

Reduction of waste sludge in biological wastewater treatment plants is becoming one of the most serious challenges due to rising sludge management costs and stringent environmental legislation. Interest in renewable energy is also increasing due to high energy price and the global climate change problem, mainly caused by excessive use of fossil fuels. Recently, anaerobic sludge digestion has become a very important sludge management technology than ever before as renewable bioenergy (methane) is recovered from the sludge digestion. Anaerobic sludge digestion consists of series of reactions; sludge hydrolysis, acidogenic and acetogenic reactions followed by methanogenic reaction [1,2]. Primary sludge and waste activated sludge are the two main sludges of biological wastewater treatment plants. Particulate organic matters are the main component of primary sludge, while waste activated sludge is mostly comprised of excess bacterial biomass. Primary sludge and waste activated sludge have different hydrolysis characteristics as they have different compositions. Hydrolysis kinetic constant of a waste activated sludge during anaerobic digestion is almost an order of magnitude lower than that of a primary sludge due to the bacterial cell wall [2]. Therefore, slow hydrolysis of waste activated sludge is known as the rate-limiting step of the anaerobic sludge digestion, and it takes a hydraulic retention time of 20 days or more for conventional sludge digestion, which requires huge volume of anaerobic digester [1,2].

Pretreatment of wastewater sludge prior to anaerobic digestion has been intensively studied for sludge minimization and enhanced bioenergy recovery [1-5]. Many pretreatment technologies have been studied including chemical, thermal, and mechanical methods [4-13]. Ultrasound, a mechanical pretreatment technology, causes a pressure wave converted into cavitation bubbles which rise and collapse violently with the energy. During the ultrasonic sludge treatment, shear force generated by the cavitation bubbles disintegrates the bacterial cell envelope and the structure of sludge flocs to transform some portions of the particulate organic matter into a soluble form [14-20].

Meanwhile, the excess power input for the ultrasonic treatment could yield a marked temperature rise after a long-duration operation. The temperature of ultrasonic sludge hydrolysis reactor may easily reach 70 °C [16,20]. Thermal pretreatment is usually carried out at 160-180 °C for sludge hydrolysis [3,7], which is significantly higher than the temperature of ultrasonic sludge hydrolysis. Recently, Ferrer et al. (2008) showed that thermal sludge pretreatment at low temperature (70 °C) was also very effective to increase biogas production in anaerobic digestion [13]. In this aspect, the heat effect is unavoidable and should not be neglected during the ultrasonic sludge pretreatment for hydrolysis [13,15].

Therefore, ultrasonic treatment hydrolyzes sludge by the simultaneous actions of the cavitation bubbles and the heat released from the dissipation of the ultrasound. In general, heat allows cavitation to be achieved at lower acoustic power [16]. On the other hand, it is also claimed that the physical and chemical impacts from the ultrasonic cavitation bubbles may be reduced due to the increased vapor pressure [21-23]. In this aspect, temperature can exert a significant effect on the efficiency of sludge hydrolysis during the ultrasonic sludge treatment.

The objective of this study is to estimate the relative effects of ultrasonic cavitation and the heat released from the ultrasound irradiation on the degree of sludge hydrolysis during the ultrasonic sludge treatment with different types of sludge (primary sludge, waste activated sludge). The results will be helpful to find the optimal temper-

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ature condition for the ultrasonic sludge treatment. For the purpose, the sludges were ultrasonically and thermally treated separately and sequentially at different temperatures (30-90 °C), and the efficiencies of sludge hydrolysis were compared.

MATERIALS AND METHODS

1. Ultrasonic and Thermal Sludge Hydrolysis

Two different experimental methods were employed to analyze the effect of temperature for the ultrasonic sludge hydrolysis. In the first method, the sludge was directly treated with ultrasound only (the detailed experimental condition is described in section 2.1.1). In the second method, the sludge was sequentially treated with thermal and ultrasonic methods (see section 2.1.2). Waste sludge was collected from a municipal wastewater treatment plant in Chuncheon City, Korea. Sludges from the anaerobic digester (anaerobic sludge), the primary settling basin (primary sludge), and the secondary settling basin (waste activated sludge) were used for this experiment and their characteristics were summarized in Table 1. All the sludge experiments were carried out in duplicate and the average values were used for the analysis.

1-1. Ultrasonic Treatment

Ultrasonic treatment was conducted with Ultrasonic Homogenizer (Model VC-750, Sonics & Materials, USA) with the frequency of 20 kHz at the average power of 0.2 kW/L for 60 minutes with 0.5 L sludge (anaerobic, primary, and activated sludge) in a 1 L beaker for the treatment. The efficiency of sludge hydrolysis or solubilization was monitored by measuring total chemical oxygen demand (TCOD) and soluble chemical oxygen demand (SCOD), and total and soluble proteins before and after the ultrasonic treatment.

1-2. Sequential Thermal and Ultrasonic Treatment

For the sequential thermal and ultrasonic sludge treatments, primary sludge and activated sludge were initially exposed at different temperatures (30, 50, 70, 90 °C) in a heating bath for 0-3 hours before the ultrasonic treatment. Hours in thermal treatment time means the thermal treatment time after the sludge temperature reaches the set point from the ambient temperature. After the thermal treatment the sludge was directly transferred for ultrasonic treatment for 20 minutes at an average power of 0.4 kW/L. The TCOD and the SCOD were measured before and after the treatments to determine the sludge hydrolysis or solubilization efficiency.

Table 1.Characteristics of waste sludge used in this experiment

	Anaerobic sludge	Primary sludge	Waste activated sludge
TSS (mg/L)	21,150	14,500	8,040
VSS (mg/L)	11,150	6,840	5,240
VSS/TSS	0.527	0.472	0.652
TCOD (mg/L)	17,800	9,700	6,750
SCOD (mg/L)	370	280	500
TCOD/TSS	0.842	0.669	0.840
Total protein (mg/L)	9,670	3,490	3,790
Soluble protein (mg/L)	189	12	22
T−Protein/COD	0.543	0.360	0.561

2. Analytical Methods

In general, the efficiency of sludge pretreatments has been assessed by the increase of soluble organic matters (i.e., volatile dissolved solids (VDSs), SCOD or soluble proteins). Some studies also focus on anaerobic biodegradability and biogas production, mainly in mesophilic batch assays [4-7]. In this study sludge hydrolysis (or solubilization) efficiency by pretreatment was based on the solubilized COD and defined as follows:

$$\text{Sludge hydrolysis (\%)} = (\text{SCOD} - \text{SCOD}_o)/(\text{TCOD}_o - \text{SCOD}_o) \times 100\%$$

Where, SCOD is the soluble COD of the treated sludge, and SCOD_o and TCOD_o are the initial soluble and total COD before the treatment. For the estimation of total suspended solids (TSS), volatile suspended solids (VSS) and COD, 'Standard Methods' of analysis [24] were employed. TCOD and SCOD of the sludge were measured before and after the treatment with Hach kit (LR 150, USA) by spectrophotometer HS 3300 (Humas, Korea).

Sludge hydrolysis efficiency based on protein was defined by the same principle used in COD. Soluble and total protein concentrations were measured by the Lowry method with bovine serum albumin as the standard [25]. For the preparation of soluble components, the sludge samples were centrifuged for 10 minutes at 6,000 ×g (Mega 17R, Hanil Science Industrial, Korea) and filtered (GF/C, Whatman, USA) for the further measurement.

RESULTS AND DISCUSSION

Sludge from the anaerobic digester (anaerobic sludge), primary settling basin (primary sludge), and secondary settling basin (waste activated sludge) was analyzed before the treatment as shown in Table 1. From the TCOD/TSS and T-protein/TCOD ratios, the anaerobic sludge and the waste activated sludge had similar organic and protein contents, and the ratios were higher than those of the primary sludge. TCOD/TSS ratios of the anaerobic sludge and the waste activated sludge were 0.842 and 0.840, and T-protein/TCOD ratios were 0.543 and 0.561 while the primary sludge had TCOD/TSS and T-protein/TCOD ratios of 0.669 and 0.360, respectively. It is well known that protein occupies about 50% of the bacterial biomass [26]. The results are well in agreement with the fact that bacterial biomass is the major component of the anaerobic sludge and the waste activated sludge.

1. Sludge Hydrolysis with Ultrasonic Treatment

Sludge hydrolysis efficiency increased with the ultrasonic energy input. At the ultrasonic energy input of 360 kJ/L, the hydrolysis of waste activated sludge reached 31.5%, while the anaerobic sludge and the primary sludge were hydrolyzed 17.2 and 9.4%, respectively, based on COD analysis. At the final energy input of 720 kJ/L, the hydrolysis efficiencies of the anaerobic sludge, the primary sludge, and the waste activated sludge were 36.6, 16.9, and 40.2%, respectively (Fig. 1(a)). The anaerobic sludge and the waste activated sludge, which mainly consist of excess bacterial biomass, were more susceptible to ultrasonic treatment than the primary sludge. The waste activated sludge was more rapidly and highly hydrolyzed than the anaerobic sludge, especially at low energy level (360 kJ/L). During the ultrasonic treatment, the sludge temperature increased from 10 °C to 78 °C due to the heat released from the ultrasonic cavitation bubbles while the pH was relatively constant (data not shown).

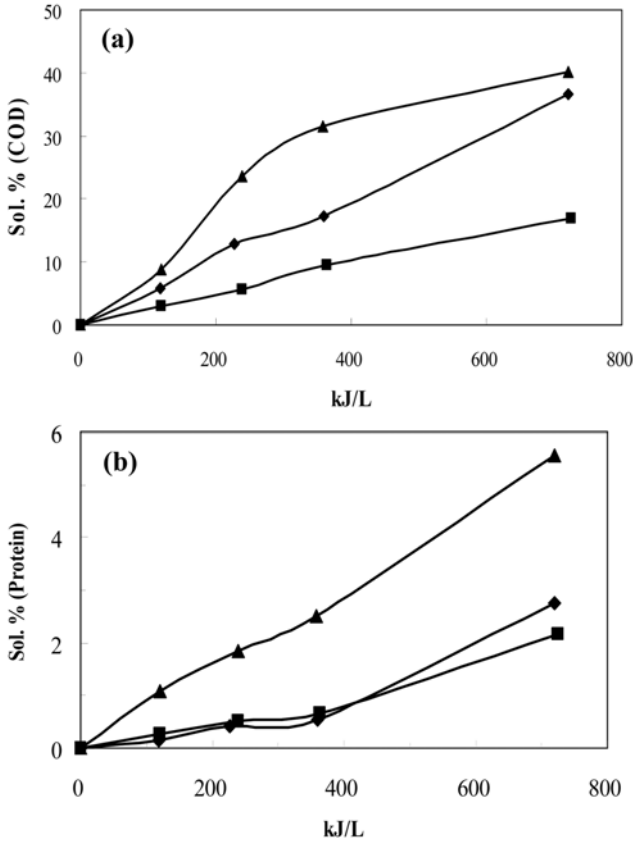

Fig. 1. Effect of volumetric energy input on hydrolysis (COD (a), Protein (b)) of anaerobic (◆), primary (■), and activated (▲) sludge during ultrasonic treatment (0.2 kW/L, 60 minutes).

Total proteins to total COD ratio (%) of the anaerobic sludge, the primary sludge, and the waste activated sludge before the ultrasound treatment were 54.3, 36.0, and 56.1%, respectively (Table 1). From the measurements of soluble proteins, ultrasound effectively solubilized the waste activated sludge than other sludge (Fig. 1(b)). The trend of sludge hydrolysis based on proteins was similar to that of COD, but the hydrolysis efficiencies of the anaerobic sludge, the primary sludge, and the waste activated sludge reached 2.7, 2.2, and 5.5%, respectively, which were significantly lower than the hydrolysis efficiencies based on COD. Lower sludge hydrolysis efficiency based on proteins seemed to be due to the fact that COD components are more easily solubilized than the proteins. Furthermore, some of the solubilized proteins can be transformed again into insoluble form by denaturation and coagulation at high temperature and ultrasonic condition [20]. Centrifugation and GF/C filtration of the sludge samples for the determination of the soluble components could also eliminate some of the colloidal protein particles from the supernatant.

The ratios of solubilized proteins to solubilized COD of the sludge were 4.1 (anaerobic), 4.7 (primary), and 8.3% (waste activated) after ultrasonic treatment and they were much lower than the ratios of the intact sludge. The result indicates that the COD components of sludge are more easily solubilized than the proteins. It can be also thought that proteins do not dissolve easily by the ultrasonic treatment because a significant amount of proteins are embedded in cell membranes or cell walls [26]. Wang et al. [19] also reported that only 25% of total proteins were solubilized from waste activated sludge at the ultrasonic power of 0.768 kW/L, which is almost four-times higher than this study. Protein solubilization efficiency seems to be proportional to the ultrasonic power input because about a quarter of protein solubilization efficiency (5.5%) was obtained in this study at a quarter of ultrasonic power (0.2 kW/L) to the result of Wang et al. (25% protein solubilization at 0.768 kW/L) [19]. They also obtained 31.6% of COD solubilization, which is higher than that of proteins. Comparing the above results, it shows that the soluble protein/SCOD ratio of the waste activated sludge used in this study is relatively lower than that of Wang et al. [19] because they used higher ultrasound power and different experimental conditions for the preparation and measurement of the soluble components as discussed above.

Table 2 shows the changes of TSS and VSS of the sludge during the ultrasonic treatment. TSS and VSS of the anaerobic sludge and the waste activated sludge decreased more rapidly than the primary sludge, suggesting that microbial (anaerobic, waste activated) sludge is more susceptible to ultrasound than primary sludge. After the ultrasonic treatment for 1 hour, TSS of the anaerobic, primary, and the waste activated sludge decreased 43.3, 20.7, and 46.5% while VSS decreased 52.2, 19.6, and 49.6%, respectively. The VSS/TSS ratio of the anaerobic sludge and the waste activated sludge changed from 0.54 and 0.65 to 0.46 and 0.61, respectively, before and after the treatment suggesting that VSS are more susceptible to ultrasound than TSS.

The dynamics of VSS hydrolysis of the sludge during the ultrasonic treatment was also investigated. The correlations of VSS reduction and SCOD increase are shown in Table 3. The average ΔSCOD/($-\Delta$VSS) ratio of the sludge (anaerobic, primary, waste activated) was 1.07, 1.19, and 0.97, respectively, during the ultrasonic treatment for 60 minutes. The primary sludge had a relatively high ΔSCOD/($-\Delta$VSS), which suggests that the solubilized VSS of the primary sludge had a higher COD content than other sludge even though its

Table 2. Concentration changes of the suspended solids (TSS and VSS) during the ultrasonic (US) sludge treatment

US time (min)	Anaerobic sludge (g/L)		Primary sludge (g/L)		Waste activated sludge (g/L)	
	TSS	VSS	TSS	VSS	TSS	VSS
0	21.15	11.50	14.50	6.84	8.04	5.24
10	19.75	10.30	13.85	6.54	7.77	5.01
20	17.85	8.80	13.60	6.45	7.07	4.27
30	17.00	8.00	12.10	5.75	6.42	3.87
60	12.00	5.50	11.50	5.50	4.32	2.64

Table 3. Changes of the soluble COD and the volatile suspended solid during the ultrasonic (US) treatment

US time (min)	Anaerobic sludge			Primary sludge			Waste activated sludge		
	ΔSCOD (g/L)	−ΔVSS (g/L)	ΔSCOD/ −ΔVSS	ΔSCOD (g/L)	−ΔVSS (g/L)	ΔSCOD/ −ΔVSS	ΔSCOD (g/L)	−ΔVSS (g/L)	ΔSCOD/ −ΔVSS
0-10	1.01	1.20	0.84	0.29	0.30	0.97	0.55	0.23	2.40
10-20	1.24	1.50	0.83	0.24	0.09	2.67	0.93	0.74	1.26
20-30	0.75	0.80	0.94	0.36	0.70	0.51	0.60	0.63	0.95
30-60	3.39	2.50	1.36	0.70	0.25	2.8	0.54	1.23	0.44
0-60	6.39	6.00	1.07	1.59	1.34	1.19	2.51	2.60	0.97

hydrolysis efficiency was low. In case of the waste activated sludge, the ΔSCOD/(−ΔVSS) was 2.40 (0-10 min) at the early stage of ultrasonic treatment and it gradually decreased to 1.26 (10-20 min), 0.95 (20-30 min), and 0.44 (30-60 min), indicating that high COD components were solubilized first, and low COD components were solubilized later.

Vlyssides [16] reported that the ΔSCOD/(−ΔVSS) ratio depends on the solubilized material. For carbohydrate the ratio is about 1.2, proteins are 2.0, and fats and lipids are 2.5. For thermal-alkaline treatment, it was reported that the solubilization of waste activated sludge started with carbohydrates followed by proteins, and finally by fats and lipids [11]. However, waste activated sludge disintegration by ultrasound showed that fats and lipids were solubilized first, followed by proteins and amino acids, then carbohydrates according to the above ΔSCOD/(−ΔVSS) result. The difference in the result may be due to the alkaline treatment which denatures proteins [11]. Wang et al. [19] also analyzed organic compounds released from the ultrasonic treatment of waste activated sludge and they reported that proteins were the main components in the liquid. Anaerobic sludge and primary sludge showed different behavior to the activated sludge, however, the reason is not clear yet.

2. Sequential Treatment of Heat and Ultrasonic Cavitation for Sludge Hydrolysis

The temperature rose significantly during the ultrasonic sludge treatment as the ultrasonic energy was finally dissipated as heat. Both the cavitation bubbles and the heat induced by the ultrasound contributed to sludge solubilization [13,15,20]. However, the relative contribution of cavitation bubbles and heat on sludge solubilization is unknown. Sequential thermal and ultrasonic treatment of the primary sludge and the waste activated sludge were carried out to analyze the effect of heat (temperature) during the ultrasonic treatment.

Fig. 2 shows the percentage of SCOD produced by the initial thermal treatment (white and hatched bars) and the following ultrasonic treatment (black bars), respectively. Sequential treatment of the primary sludge (Fig. 2(a)) showed that the total hydrolysis percent was almost constant at 10-13% regardless of thermal treatment temperature (30-90 °C) and time (0-3 hours) in the range of this experiment. The results suggest that the total amount of soluble substance in the primary sludge susceptible to combined thermal and ultrasonic treatment is fixed. Sludge solubilization by thermal treatment at 30 °C was negligible due to its low temperature. However, it reached a maximum of 13% at 90 °C with sequential treatment of thermal and ultrasound. The higher temperature and the longer thermal treatment time increased sludge solubilization. The relative contribution of the ultrasonic treatment on primary sludge

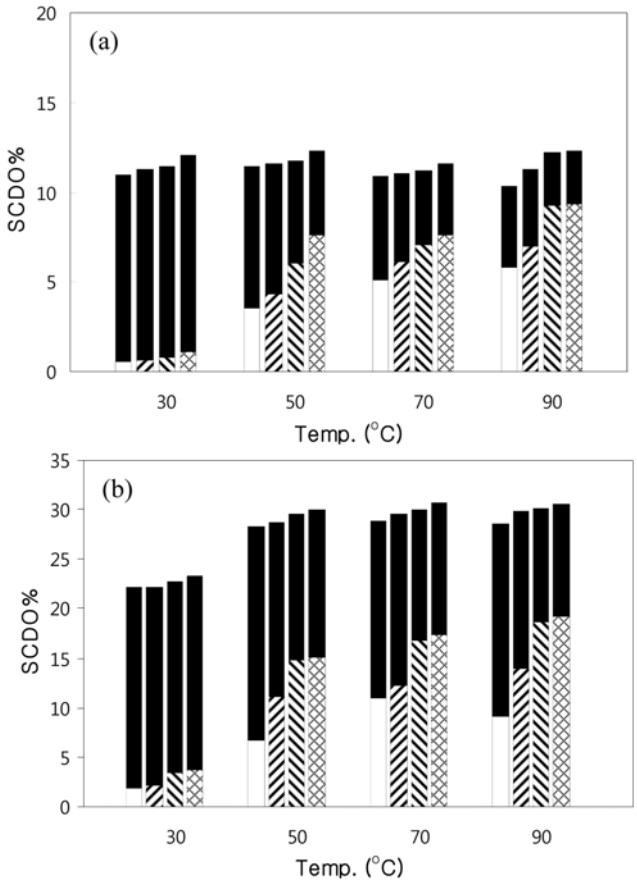

Fig. 2. Hydrolysis of the primary sludge (a) and the activated sludge (b) by sequential heat treatment (□: 0 hour, ▨: 1 hour, ▧: 2 hours, ▩: 3 hours) and ultrasound treatment (■: 0.4 kW/L, 20 minutes).

hydrolysis declined as the temperature and the thermal treatment time increased (Fig. 2(a)). Even the thermal treatment for 0 hour at 90 °C contributed sludge hydrolysis more than the ultrasonic treatment.

For the waste activated sludge (Fig. 2(b)), sequential thermal and ultrasonic treatment increased sludge hydrolysis up to 30%, and which is much higher than that of the primary sludge. The total SCOD percentage increased from 22-24% at 30 °C to 28-30% at 50 °C and remained almost constant at higher temperatures at 70 and 90 °C regardless of thermal treatment time. At 30 °C solubilization of the primary sludge and the waste activated sludge by heat was very

low and ultrasound provided a major contribution to the sludge solubilization. However, heat played a significant role in sludge solubilization at higher temperature and the contribution of ultrasound decreased as the treatment temperature and time increased. Grönroos et al. [18] also reported that sludge solubilization increased linearly in the temperature range of 20-60 °C. Up to 70% of the total hydrolysis was induced by heat in the case of the primary sludge at 90 °C, while less than 60% was induced by heat with the waste activated sludge. The difference seems to be the result of the differences of the materials and structure of the sludge.

It is well known that sludge hydrolysis increases with the reaction temperature, and cavitation can also be achieved at lower acoustic power if the reaction temperature is high. On the other hand, the physical and chemical effects resulting from the collapse of cavitation bubbles under sonication are reduced due to the increase in vapor pressure associated with the liquid [22,27-29]. Entezari and Kruus [30] noted that rate of KI oxidation decreases linearly with increasing temperature with 20 kHz ultrasound. Therefore, it is considered that the sequential treatment of heat and ultrasonic cavitation for sludge hydrolysis under higher temperature conditions gives rise to reduction in the physical and chemical effects produced by cavitational collapse under ultrasonication.

From the above results, primary sludge and waste activated sludge were highly susceptible to heat. The contribution of heat treatment to total hydrolysis was greater for the primary sludge compared to the waste activated sludge. Higher temperature and longer thermal sludge treatment time enhanced sludge hydrolysis before the ultrasonic treatment. Sludge hydrolysis by ultrasonic cavitation decreased at higher temperature due to the increase in vapor pressure. It can also be possible that the thermally pre-treated sludge had less soluble components than the intact sludge so that the subsequent ultrasonic treatment produced less soluble products than the thermal treatment because the easily soluble components are already solubilized during the thermal treatment. It is also possible that the sludge may become more resistant to ultrasonic hydrolysis at higher temperature like protein denaturalization.

In summary, the components with high COD per mass, like fats and lipids, were more easily solubilized than other components during the ultrasonic treatment with waste activated sludge. The hydrolysis efficiency of primary sludge with the combined sequential thermal and ultrasonic treatment was independent of the treatment temperature and time duration. Similarly, the sequential thermal and ultrasonic treatment of the activated sludge showed almost constant hydrolysis efficiency at 50 °C or higher. However, the efficiency increased with the thermal treatment time. Sequential thermal and ultrasonic treatment achieved higher hydrolysis than ultrasonic treatment alone. Therefore, the temperature and time for sludge preheating before ultrasonic treatment need to be optimized. Ultrasonic sludge hydrolysis is the result of both shear stress generated by the collapse of cavitation bubbles and temperature rise due to the energy dissipation. And their contribution of sludge hydrolysis needs to be separated and quantified for the optimum design and operation of the sludge hydrolysis facility.

CONCLUSIONS

1. Activated sludge was highly susceptible to ultrasonic treatment for hydrolysis as compared to anaerobic sludge and primary sludge. Sludge hydrolysis efficiency was higher when it was based on COD rather than proteins.

2. In case of activated sludge hydrolysis by ultrasound, the $\Delta SCOD/(-\Delta VSS)$ decreased from 2.40 to 0.44 indicating that high COD components were solubilized first, and low COD components were solubilized later.

3. Total hydrolysis efficiency of activated sludge by the sequential thermal and ultrasonic treatment increased with reaction time and temperature up to 50 °C and it remained almost constant at higher than 50 °C. The contribution of heat effect became more significant as temperature and heat exposure time increased, while the effect of ultrasound on sludge hydrolysis decreased.

4. Heat generated during the ultrasonic treatment played a key role in sludge solubilization. For the optimum design and operation of the sludge hydrolysis facility, the contribution of shear force by the collapsed cavitation bubbles and the heat released during ultrasonic treatment for the sludge hydrolysis needs to be analyzed separately.

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