Influence of thermal hydrolysis pretreatment on physicochemical properties and anaerobic biodegradability of waste activated sludge with different solids content

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Abstract

The influence of thermal hydrolysis pretreatment (THP) on physicochemical properties (pH, total solids, volatile solids, chemical oxygen demand, total nitrogen, ammonium nitrogen, volatile fatty acids, viscosity, and cell morphology) and anaerobic biodegradability of highly concentrated waste activated sludge (WAS) with TS content ranging from 1-7% was evaluated at different temperatures ranging from 100-220 °C. The biomethane potential (BMP) of the WAS was systematically analyzed and evaluated. Images of its cellular structure were also analyzed. The results indicated that THP is a useful method for solubilizing volatile solids and enhancing CH_4 production regardless of the TS content of the WAS feed. The ultimate CH_4 production determined from the BMP analysis was 313-348 L CH_4/kg VS (72.6-74.1% CH_4) at the optimum THP temperature of 180 °C. The results showed that THP could improve both the capacity and efficiency of anaerobic digestion, even at a high TS content, and could achieve the dual purpose of sludge reduction and higher energy recovery.

Keywords: thermal pretreatment; sludge properties; wastewater activated sludge; anaerobic digestion; biomethane potential

1. Introduction

A large amount of excess sludge is produced daily at wastewater treatment plants (WWTPs) worldwide (Zhang et al., 2017). In South Korea , the average generation of sewage sludge (as wetted solids) in 2013 was about 3.6 million t/y, thereby accounting for 19.65% of the municipal solid waste stream by volume (Korea Ministry of Environment, 2016). Since sewage sludge contains an array of pathogens, nutrients, degradable organics, and possibly some hazardous substances and heavy metals, it needs to be handled carefully and its mass must be reduced appropriately before disposal or reuse (Raheem et al., 2017; Semblante et al., 2014).

Standards and policies for sewage sludge management vary by country (Villar et al., 2016). In general, they aim to promote cost-effective and sustainable sludge management through best practice as well as research and development. The management of sewage sludge presents many technical challenges, and the associated capital and operating costs can exceed 50% of the total wastewater treatment costs (Serrano et al., 2015). Additionally, because sewage sludge contains mostly degradable organics, it is a valuable source of sustainable biomass energy (Leng et al., 2018). Consequently, it is expected to become a mainstream energy source for environmentally and socially sustainable development (Nguyen et al., 2017a; Tyagi and Lo, 2013).

There are several technologies for sewage sludge treatment, including alkaline stabilization, aerobic digestion, anaerobic digestion (AD), composting, landfilling, and incineration (Bougrier et al., 2007; Chiu et al., 2015; Zhang et al., 2017). Among them, AD has likely been the most widely used process for treating sewage sludge at WWTPs because of its many advantages, such as high renewable energy production, low environmental impacts, and low solid residue with fewer pathogens (Chiu et al., 2015; Nizami et al., 2017). However, further studies and development are necessary to optimize the performance of AD in a retrofit design or new project. These limitations include i) slow and incomplete decomposition of organic matter; ii) long digestion time and large digester volume causing high capital and operating costs; (iii) sensitivity to temperature, pH, loading changes, and inhibitors; and iv) low biogas production and CH_4 content (Appels et al., 2008; Bougrier et al., 2007; Koch and Drewes, 2014; Ushani et al., 2018). To overcome the aforementioned issues and to improve the efficiency of AD, various pretreatments can be used, including biological (e.g., enzyme and ensilage) (Liu et al., 2017), mechanical (e.g., ultrasonic, agitator, high pressure homogenization, and jetting) (Le et al., 2016; Nguyen et al., 2017c), chemical (e.g., alkali or acid) (Li et al., 2012; Rajesh Banu et al., 2017), thermal (Appels et al., 2010; Eftaxias et al., 2018; Li et al., 2018), and combined (Peng et al., 2018) processes.

Thermal hydrolysis pretreatment (THP) has been proposed as an effective technology for accelerating the hydrolysis of organic matter (Jin et al., 2016; Li et al., 2016; Urrea et al., 2018). During THP, complex organic substances can be broken down into smaller and soluble molecules suitable for biodegradation (Yang et al., 2017) and cell walls rupture, releasing their contents into the homogenized solution of which the largest are lysed microbial cells that subsequently become available for metabolic activity and microbial growth. In addition, THP provides a homogenized sludge solution with stable conditions, thereby preventing shock loading during AD, which consequently leads to shortened digestion time, decreased sludge mass, enhanced biodegradation, and CH_4 production (Ariunbaatar et al., 2014; Bougrier et al., 2007; Kim et al., 2015; Liao et al., 2016). The general advantages are outlined above, but the specific physicochemical and biological properties (organic/insoluble fraction, solubilization, viscosity, biodegradability, cell morphology, etc.) of thermally pretreated waste activated sludge (WAS) in different experimental conditions (solid load and temperature) and their relationships have not been systematically elucidated (Liu et al., 2012). In addition, in Korea, there are still very few investigations on the use of the thermal pretreatment method to enhance sludge hydrolysis, anaerobic biodegradability, and biogas production to meet local conditions (environment, organic components, waste sources, etc.) (Yang et al., 2017).

The major goal of this study was to investigate and evaluate the performance of THP for highly concentrated WAS. The results will be helpful for integrating THP processes into existing and new WWTPs in South Korea, with the goal of improving sludge digester performance and biogas yields. The physicochemical properties of sludge, namely volatile solids (VS), chemical oxygen demand (COD) solubilization, ammonium nitrogen (NH_4^+-N), volatile fatty acids (VFA), viscosity, and cellular structure, were measured. Biomethane potential (BMP) was also tested. Furthermore, the optimum temperature of THP for various solids concentrations was evaluated in order to obtain the most effective economic and environmental benefits.

2. Materials and Methods

2.1. Preparation of substrate and inoculation

The raw WAS and inoculum sludge used in this study were sourced from a WWTP with a capacity of 85 000 m^3/d and a mesophilic AD plant, respectively. Both plants were located in "N" city, South Korea. Large debris and inert materials were eliminated from the samples using a No. 18 mesh sieve (0.85 mm). Samples were then stored in a plastic bottle at 4 °C and in an incubator at 35 °C, respectively, before experimental use. WAS of high total solids (TS) concentration (69.6-72.4 g TS/L) was utilized by diluting it with deionized water to desired solids concentrations of 9.9 g TS/L, 29.91 g TS/L, 49.90 g TS/L, and 70.06 g TS/L, which corresponded to $WAS_{1\%}$, $WAS_{3\%}$, $WAS_{5\%}$, and $WAS_{7\%}$, respectively. Characteristics of the WAS samples are summarized in Table 1.

Table 1: Characteristics of raw waste activated sludge (WAS) used for the experiment.

[Insert Table 1]

2.2. Thermal pretreatment and evaluation of chemical oxygen demand solubilization

A batch-type processor was used for the high-pressure THP. The processor consisted of a control unit, pressurized vessel, pressure-reducing valve, pressure gauge, heating element, hydraulic cooling system, and thermometer (Fig. 1); these were designed by our lab members for laboratory testing only. The stainless steel reactor had a total capacity of 1 L. The automatic temperature control apparatus was designed to maintain a constant temperature with an uncertainty of ± 1 °C by activating the heating or cooling system as needed. Thermal pretreatment of WAS was conducted at temperatures ranging from 100-220 °C with the sludge sample volume held at a constant value of 700 mL for each experiment.

[Insert Figure 1]

Fig. 1. Thermal pretreatment apparatus a) schematic diagram and b-c) photographs.

In order to create an anaerobic environment and avoid oxidation of the samples during hydrolysis, the headspace of the THP reactor was initially flushed using N_2 at a rate of approximately 0.5 L/min for 3 to 5 min before performing the thermal hydrolysis. The pretreatment temperature was increased to the set point and maintained for 30 min. During the pretreatment experiment, the pressure inside the pressurized vessel varied from 0.8 to 2.6 MPa depending on the setpoint pretreatment temperature. The agitation velocity was set at 180 rpm and maintained until sampling. After each reaction, the temperature of the sludge was allowed to cool to less than 60 °C, and saturated vapor was relieved to reduce pressure for sample collection.

COD solubilization served as an index indicating the efficiency of the pretreatment process by way of Eq. 1.

$$\text{COD solubilization (\%)} = \frac{C_s - C_{So}}{C_o - C_{So}} \times 100 \qquad \text{Eq. 1}$$

where Co = total chemical oxygen demand (TCOD) concentration before pretreatment in g/L; Cso = soluble chemical oxygen demand (SCOD) concentration before pretreatment in g/L; and Cs = SCOD concentration after pretreatment in g/L.

2.3. Biochemical CH_4 potential tests

In order to evaluate enhancements in the biodegradability of substrates and ultimate biogas yields due to THP, BMP tests were performed (Dwyer et al., 2008; Holliger et al., 2016; Nguyen et al., 2017a).

Tests were conducted in 630 mL serum bottles (effective volume of 330 mL) with coiled butyl rubber stoppers at 35 °C. Inoculum (10%, v/v) and substrate (2 g $VS_{substrate}$/L) were placed in a growth medium containing 1.8 g/L NH_4Cl, 0.7 g/L KH_2PO_4, 0.4 g/L $MgCl_2·6H_2O$, 0.2 g/L $CaCl_2·2H_2O$, 20 mg/L $FeCl_2·4H_2O$, 5 mg/L $CoCl_2·6H_2O$, 1 mg/L $MnCl_2·4H_2O$, 1 mg/L $NiCl_2·6H_2O$, 0.5 mg/L $ZnCl_2$, 0.5 mg/L H_3BO_3, 0.5 mg/L Na_2SeO_3, 0.4 mg/L $CuCl_2·2H_2O$, and 0.1 mg/L $Na_2MoO_4·2H_2O$ (Del Río et al., 2011). If necessary, the pH was adjusted to 7.1 ± 0.1 using either 2 M HCl or 2 M NaOH solution. To prevent a sudden decrease in pH caused by acid fermentation during the early stages of AD, 2.6 g/L of $NaHCO_3$ was included in the serum bottle as a buffer. Pure N_2 was used to purge the serum bottles to create anaerobic conditions (Nguyen et al., 2017b).

The biomethane production was expressed as the volume of CH_4 produced per gram of VS of substrate (mL CH_4/g VS_{add}). CH_4 production was measured periodically using a 50 mL glass syringe to extract the gas. In order to take volume expansion and saturated steam pressure into account, the CH_4 volume was converted to standard state (STP: 0 °C, 1 bar) volume (V) using the ideal gas law and the 35 °C saturated vapor pressure of 42.2 mmHg.

2.4. Analytical methods

TS, VS, TCOD, SCOD, TN, and NH_4^+-N analyses were performed using standard methods (APHA-AWWA-WEF, 2005). Soluble fractions of $SCOD_{Cr}$, NH_4^+-N, and VFA were analyzed using filtrate obtained from filtering the supernatant with a 1.2 µm GF/C microfiber filter (Whatman, UK) after centrifugation at 7000 rpm for 20 min. pH was measured using a pH meter (Hanna HI223, USA). VFA were analyzed by gas chromatography (GC) with a flame ion detector (Agilent 7890A, Agilent Technologies, Inc., USA) after solvent extraction (1:1 with ether) and adjustment of filtrate pH to 2.2 ± 0.2 using 3% HCl (Nguyen et al., 2016). CH_4 content in the generated biogas was measured by employing the BMP test and was examined using gas chromatography with a thermal conductivity detector (Agilent 7890A, Agilent Technologies, Inc., USA). The GC operation conditions used for VFA and CH_4 analyses are summarized in Table 2.

Table 2: Gas chromatography analysis conditions for volatile fatty acids and CH_4.

[Insert Table 2]

The viscosities of the untreated and pretreated sludge samples were determined at 20 °C using a viscometer (Brookfield Viscometer DV2T, USA). The torque percentage was adjusted through a range of 20% to 800% by adjusting the rotational speed of the spindle (Xue et al., 2015). In order to observe changes in the microorganism cell structure, images were taken using a transmission electron microscope (80 Kv, JEM1010, JEOL, Japan).

3. Results and Discussion

3.1. Effects of thermal hydrolysis pretreatment on waste activated sludge characteristics

THP was undertaken in the range of 100 to 220 °C using four kinds of sludge ($WAS_{1\%}$, $WAS_{3\%}$, $WAS_{5\%}$, and $WAS_{7\%}$) with solid concentrations of 9.90 g TS/L, 29.91 g TS/L, 49.90

g TS/L, and 70.06 g TS/L, respectively. The changes in sludge physicochemical properties were analyzed and the results are shown in Figs. 2 and 3.

The TS, TCOD, and TN concentrations of the thermally pretreated sludge decreased marginally due to the formation of a scale layer on the inner wall of the thermal reactor (Fig 2). On the other hand, SCOD, NH_4^+-N, and VFA in the soluble phase gradually increased directly proportional to the increase in pretreatment temperature from 100 to 220 °C in all four samples (Fig. 2). It is of particular note that THP resulted in significant COD solubilization. The most notable increase in SCOD was observed with $WAS_{7\%}$ (sample with the highest solid content). In addition, there was a notable increase in the SCOD fraction of TCOD of this sample from 49.63 to 55.11% at the pretreatment temperature of 220 °C. Figure 2 also shows a decrease in the pretreated sludge pH as the reaction temperature increased. This coincided with the increase in VFA concentration (Fig. 2), thereby confirming that thermal pretreatment was effective at promoting hydrolysis and solubilization. Additionally, during WAS thermal hydrolysis, the VFA concentrations in all WAS samples consistently increased with increased thermal hydrolysis temperature (Fig. 2). An increase in VFA concentration could possibly be ascribed to the hydrolysis of unsaturated lipids (Liao et al., 2018; Wilson and Novak, 2009).

[Insert Figure 2]

Fig. 2. Influence of thermal hydrolysis pretreatment temperature on physicochemical properties of treated sludge: a) $WAS_{1\%}$, b) $WAS_{3\%}$, c) $WAS_{5\%}$, and d) $WAS_{7\%}$.

At the pretreatment temperature of 220 °C, the NH_4^+-N concentration was 0.213, 0.596, 1.120, and 1.804 g/L for $WAS_{1\%}$, $WAS_{3\%}$, $WAS_{5\%}$, and $WAS_{7\%}$, respectively, and increased by 600-1675% depending on the TS concentration. Total ammonia nitrogen (TAN) exists in the form of free ammonia nitrogen and ionized ammonium nitrogen (NH_4^+-N). TAN disrupts anaerobic digestive reactions, thereby decreasing digestive efficiency and biogas generation (Nakakubo et al., 2008; Rajagopal et al., 2013).

Wilson and Novak (2009) demonstrated that proteins existing as particulate matter can be converted into NH_4^+-N through thermal pretreatment. Ammonia is an inhibitory intermediate during AD (Xie et al., 2016). AD performance starts to decline when the concentration of TAN in the digester reaches 1.7 to 2.7 g/L (Yenigün and Demirel, 2013). In this study, the concentration of NH_4^+-N reached 1.804 g/L for $WAS_{7\%}$ at the pretreatment temperature of 220 °C. Thus, there could be a relationship between effective hydrolysis and excessive NH_4^+-N concentration in the reactor. Since both of these were promoted by thermal pretreatment, an optimal pretreatment temperature was expected, as discussed in a later section.

[Insert Figure 3]

Fig. 3. Variations in volatile fatty acids composition and pH at different temperatures for thermally pretreated a) $WAS_{1\%}$, b) $WAS_{3\%}$, c) $WAS_{5\%}$, and d) $WAS_{7\%}$.

Figure 3 shows the variations in pH and VFA composition of each sample at different THP temperatures. Acetic acid, propionic acid, butyric acid, and valeric acid accounted for roughly 73.6 to 85.9% of the VFA, with the percentage of acetic acid being the highest. More specifically, the acetic acid accounted for a significantly higher proportion (62.9-100%) in the group of four acids obtained above. Acetic acid can be readily converted into CH_4 compared to other VFA. Thus, given the high concentration of acetic acid shown in Fig. 3, an enhanced AD performance was expected, as discussed in Section 3.5.

3.2. Organic matter fraction and solubilization

The VS over TS ratio of these sludge samples varied from 0.773 to 0.793, thereby indicating that the sludge consisted mainly of organic substances. The highest COD was associated with the solid fraction rather than the soluble fraction, as evidenced by the rather low SCOD to TCOD ratio in the range of 0.022 to 0.046.

Solubilization of all WAS samples (1%, 3%, 5%, and 7%) increased as pretreatment temperature increased (Fig. 4). The change was marginal between 100 and 120 °C, but then suddenly increased above 140 °C. All the WAS (1%, 3%, 5%, and 7%) showed maximum solubilization (51.2%, 47.3%, 45.9%, and 44.0%, respectively) at 220 °C.

Del Río et al. (2011) and Seviour et al. (2009) reported that aerobic granular sludge exists as a gel structure at temperatures around 115 °C. Moreover, the sludge formed a gel structure due to the high content of extracellular polymeric substances (EPS) with gel-forming properties at or below temperatures of 115 °C. They reported that the microorganisms release small amounts of EPS at moderate temperatures, which then act as a bond to maintain the gel structure. However, EPS lose these gel-forming properties at high temperatures. These results were confirmed in the current study, which showed low solubilization at temperatures around 100-120 °C.

Evaluation of solubilization for each TS concentration of sludge revealed that as TS concentration increased, the maximum solubilization decreased. This is perhaps understandable that for WAS with high solids concentration, the heat transfer coefficient was lower than that in WAS with low solids concentration. Therefore, when applying THP to WAS with high solids concentration, higher thermal energy input or longer residence time might be required.

[Insert Figure 4]

Fig. 4. Impact of pretreatment temperature on solubilization of waste activated sludge: (a) $WAS_{1\%}$, b) $WAS_{3\%}$, c) $WAS_{5\%}$, and d) $WAS_{7\%}$.

3.3. Sludge viscosity

Viscosity is an important parameter that significantly affects the solubilization and biodegradability of sludge. It reflects the degree of interaction between the particles and sludge flocs within the mixed sludge (Markis et al., 2016). The variations in sludge viscosity by different initial solids concentrations at an initial temperature of 20 °C and after THP are shown in Fig. 5.

At 20 °C, the viscosity of raw concentrated WAS increased proportionally to the solids concentration (Fig. 5a). The sludge viscosity at a TS concentration of 9.9 g/L was 40 MPa·s, and increased to 3102 MPa·s for sludge with a TS concentration of 70.63 g/L. As the solubilizing temperature increased, the viscosity of each sludge decreased gradually (Fig. 5b). At 140 °C, viscosity suddenly decreased, and at 220 °C, it had decreased to 3 MPa·s, 12 MPa·s, 28 MPa·s, and 250 MPa·s for WAS of 1%, 3%, 5%, and 7%, respectively. As sludge concentration increased, viscosity also increased, which meant that more pumping energy would need to be supplied for pipe transfer and reactor agitation could be increased. Therefore, it is likely that as sludge viscosity decreases from thermal pretreatment, the energy required for facility operation (i.e., pumping and mixing) could be reduced (Brar et al., 2005; Xue et al., 2015) and particulate fraction biodegradability could be improved (Bougrier et al., 2008; Brar et al., 2005).

[Insert Figure 5]

Fig. 5. Change in sludge viscosity at (a) different initial solids concentrations and (b) after thermal pretreatment at various temperatures.

3.4. Transmission electron microscope analysis

Transmission electron microscope analysis was used to study the effects of THP on cell morphology (Fig. S1). The microorganism cell walls were destroyed by thermal pretreatment, thereby resulting in the release of cell contents (EPS and intracellular organic substances). At 120 °C, the outer cell wall was partially destroyed (Del Río et al., 2011). As the pretreatment temperature increased, internal cellular substances were eluted through the outer cell wall. However, at 180 °C, the outer cell wall contracted. Because the cell walls of microorganisms comprising WAS exist as a solid structure, the hydrolysis process was delayed during the AD stage. Cell walls of microorganisms comprising WAS are destroyed by THP, and the subsequent elution of the internal cellular solution causes the amount of dissolved substances to increase (Seviour et al., 2009). Increases in dissolved substances facilitate conversion to CH_4 later in the process, and thus increase the biodegradability and biogas output of AD (Nguyen et al., 2017c).

3.5. Effects of thermal hydrolysis pretreatment on ultimate biomethane potential

BMP is an important parameter to evaluate the biogas production potential of an organic substrate by AD because it can be used to assess anaerobic biodegradability, CH_4 production rate, and the ultimate biomethane yields of substrates. All of these variables affect the economics of the AD process. BMP test results obtained from samples both before and after THP are presented in Table 3.

Increasing the THP temperature from the ambient value to 220 °C resulted in CH_4 production increasing for all WAS samples (1%, 3%, 5%, and 7%), reaching peak values of 348, 345, 320, and 313 L CH_4/kg VS_{add} (or 244, 254, 235, and 232 L CH_4/kg COD_{add}), respectively, at 180 °C THP, which corresponded to CH_4 production increasing by 86.1%, 84.3%, 76.2%, and 76.0%, respectively, compared to those without THP. Above 180 °C for THP, CH_4 production decreased gradually. This was probably caused by increases in NH_4^+-N at high temperatures acting as an inhibiting factor in AD processes. It should also be noted that non-biodegradable substances generated by carbohydrates and proteins at a high enough temperature (optimal temperature) could cause decreases in CH_4 production, which has been reported in the literature. For example, Bougrier et al. (2008) undertook WAS solubilization at 95-210 °C, and reported that solubilization of carbohydrates and proteins in the sludge increased as temperature increased. However, in the case of carbohydrates, solubilization above 170 °C gradually decreased, which was attributed to a decrease in biodegradable substances through the Maillard reaction (i.e., a bonding between sugar and amino acids at high temperatures).

Furthermore, Dwyer et al. (2008) confirmed that thermal hydrolysis produces colored, recalcitrant compounds and melanoidins that can affect anaerobic biodegradability and CH_4 production. Del Río et al. (2011) also confirmed that at a higher sludge pretreatment temperature, dissolved carbohydrates and proteins decrease, which causes decreasing CH_4 production. Results from this study also indicated that at the same THP temperature, despite the increase in solids concentration from 1% to 7%, CH_4 production gradually decreased. This decline in CH_4 production could have been due to the higher TS concentration causing mass transfer limitations and reduced sludge fraction and solubility.

The results of this study showed that THP is an effective method for transforming WAS into easily biodegradable substrates with higher solids reduction regardless of the change in TS concentration of the sludge. Nevertheless, there exists an optimal pretreatment temperature, and the highest CH_4 production in this study was achieved at 180 °C (Table 3).

Compared with the biomethane production values from previous studies (e.g., 0.314 m^3 CH_4/kg VS_{add} or 0.217 m^3 CH_4/kg COD_{add} for 1.45% TS (Bougrier et al., 2007), 0.337 m^3 CH_4/kg VS_{add} or 0.219 m^3 CH_4/kg COD_{add} at 2.19% TS (Del Río et al., 2011), 0.261 m^3 CH_4/kg COD_{add} (Yang et al., 2010), 0.215 m^3 CH_4/kg COD_{add} at 4.6% TS (Mottet et al., 2009), and 0.286 m^3 CH_4/kg VS_{add} at 7.68% TS (Donoso-Bravo et al., 2011)), this study showed much higher CH_4 yields (Table 3). Higher CH_4 yields consequently led to high potential energy recovery, thereby allowing more widespread application in treating WAS both in facility retrofitting and in new facility designs.

[Insert Table 3]

Table 3: Biomethane yield during the biomethane potential test.

4. Conclusions

THP was performed by targeting WAS with TS contents of 1%, 3%, 5%, and 7%. Physicochemical features for pretreated sludge and CH_4 production efficiency were analyzed. In all samples, dissolved substances increased by increasing pretreatment temperature. The CH_4 production was significantly increased in the samples with THP and reached 1.76 to 1.86-fold compared with that of un-pretreated WAS samples. The optimum temperature for THP was 180 °C for achieving the highest CH_4 production. THP is an effective method for solubilizing WAS regardless of high sludge concentration. Therefore, THP is expected to greatly improve both the capacity and efficiency of the commercial AD process. Although THP offers numerous technological advantages for AD, it is still necessary to assess its economic feasibility at larger scales before practical application.

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