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Enhancement of methane production in anaerobic digestion of sewage sludge by thermal hydrolysis pretreatment

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ABSTRACT

This study was performed to optimize thermal hydrolysis pretreatment (THP) of sewage sludge for enhanced anaerobic digestion (AD). Using the response surface methodology (RSM), the optimal conditions were found 180 °C of reaction temperature and 76 min of reaction time. Through THP under optimal conditions, high molecular substances in sewage sludge such as soluble microbial by-products (SMPs) and extracellular polymeric substances (EPSs) were hydrolyzed into low molecular ones without the generation of refractory compounds. The microbial community analysis revealed that relative abundances of *Methanomicrobia* such as *Methanosarcina*, *Methanosaeta* (acetoclastic methanogens), and *Methanoculleus* (hydrogenotrophic methanogens) in AD with THP were higher than those in conventional AD.

1. Introduction

Anaerobic digestion (AD) is one of the most cost-effective technologies due to the high energy and resource recovery from sewage sludge (Ding et al., 2017). However, AD needs a large digester volume and long retention time because it is a slow process (Han et al., 2017a). AD mainly involves the following stages: 1) hydrolysis, 2) acidogenesis, 3) acetogenesis, and 4) methanogenesis. It is well known fact that hydrolysis of organic solid waste, such as sludge, is the rate-limiting step of the AD. Therefore, the pretreatment has been applied for accelerating the hydrolysis step. This enhances the rate of AD and methane production (Ding et al., 2017; Neumann et al., 2017; Veluchamy and Kalamdhad, 2017). Sludge pretreatment methods include chemical (Li et al., 2012; Shao et al., 2012), thermal (Chen et al., 2017; Han et al., 2017a,b; Neumann et al., 2017), mechanical (Żubrowska-Sudoł et al., 2017), and biological (Ding et al., 2017; Kavitha et al., 2014) disintegration. Among these pretreatment methods, thermal hydrolysis pretreatment (THP) has been proven to improve sludge disintegration and biogas production (Li and Noike, 1992; Donoso-Bravo et al., 2011; Abelleria-Pereira et al., 2015). Additionally, THP is the most profitable and reliable sludge pretreatment method (Donoso-Bravo et al., 2011; Abelleria-Pereira et al., 2015; Cano et al., 2015).

Temperature and reaction time are the major operational factors that affect the efficiency of THP. Bougrier et al. (2008) reported that chemical oxygen demand (COD) solubilization and biogas production were enhanced at temperatures lower than 190 °C (for a reaction time of 30 min). Furthermore, Higgins et al. (2017) reported that the total biogas production decreased with increasing temperature (120–180 °C for 30 min). Li and Noike (1992) evaluated the impacts of different reaction times (15, 30, 60 and 120 min; temperature of 150 °C) on the biogas production. They found that biogas production increased with increasing reaction time. Xue et al. (2015) was found that although COD solubilization increased significantly at reaction times of 90–120 min (temperature in the range of 120–180 °C), no significant increase was observed when the reaction time was longer than 120 min. Previous studies have mainly focused on the efficiency of the subsequent fermentation step (e.g., AD) that follows the THP step, which is generally carried out at temperatures of 100–180 °C for reaction times of 7–180 min (Carrère et al., 2010; Xue et al., 2015; Suárez-Iglesias et al., 2017).

The objective of this study is to determine the optimum conditions for THP of sewage sludge using response surface methodology (RSM) based on the central composite design (CCD). Furthermore, the impact of THP under the optimum conditions on characteristics of organic matter in sewage sludge is investigated using molecular weight distribution and excitation-emission matrix. Finally, continuous tests are conducted to evaluate the performance of THP + AD, and the effects of THP on the microbial community in anaerobic digestion is investigated using 454 pyrosequencing.

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Table 1
Characteristics of sewage sludge before and after thermal hydrolysis pretreatment.

Item	Unit	Untreated sewage sludge	Pretreated sewage sludge (under optimal conditions)
TCOD	g/L	169.0 ± 1.9	159.1 ± 0.3
SCOD	g/L	1.7 ± 0.1	47.3 ± 0.1
TVFAs	g/L	0.8 ± 0.0	8.2 ± 0.1
T-N	g/L	1.1 ± 0.1	1.0 ± 0.0
S-N	g/L	0.1 ± 0.0	0.2 ± 0.0
T-P	g/L	0.8 ± 0.0	0.8 ± 8.6
S-P	g/L	0.1 ± 0.0	0.3 ± 0.0
T-protein	mg/L	536.7 ± 11.4	483.7 ± 8.4
S-protein	mg/L	232.4 ± 12.7	324.4 ± 5.3
T-carbohydrate	mg/L	2,347.1 ± 81.4	2,147.1 ± 34.3
S-carbohydrate	mg/L	102.7 ± 4.6	343.8 ± 12.6

(*Note:* mean ± standard deviation).

2. Materials and methods

2.1. Seeding and sewage sludge

The seeding sludge for batch and continuous tests was obtained from the effluent of the digester located in a wastewater treatment facility in H City. The pH, alkalinity and volatile solids (VS) concentration of the seed sludge were 6.9, 2.2 g/L as $CaCO_3$, and 19.3 g/L, respectively. The sewage sludge for THP was collected from a wastewater treatment plant in D city. The characteristics of sewage sludge before and after THP, are shown in Table 1.

2.2. Thermal hydrolysis pretreatment

The sewage sludge was thermally pretreated at the thermal hydrolysis process pilot plant (Capacity: 1 ton/cycle, COWT, BKT, Korea). To achieve a total solids (TS) concentration of 14.3 ± 1.0%, the sewage sludge was subjected to dewater before THP. The thermal hydrolysis process was operated at a temperature of 75–225 °C and reaction time of 15–105 min (Table 2). Then, the thermal hydrolysis reactor was cooled down to a room temperature.

2.3. Experimental design

A central composite design (CCD) was employed to find the optimum conditions. Reaction temperature (X_1) and reaction time (X_2) were chosen as two independent factors. CH_4 yield was selected as the response variable. The experimental variables of the independent factors were transformed to coded variables by the following equation:

$$x_i = (X_i - X_i')/\Delta X_i \quad (1)$$

where x_i is the coded value of the ith experimental value, X_i is the actual value of the ith experimental value, X_i' is the center point among experimental values, and ΔX_i is the distance between X_i and X_i'. The response was fitted using a polynomial quadratic equation to correlate with independent variables. The general form of the predictive polynomial quadratic equation is described as:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_i x_i^2 + \sum_{i<j}^{k} \beta_{ij} x_i x_j \quad (2)$$

where y is the predicted response, β_0 is a constant, β_i is a linear coefficient, β_{ii} is a quadratic coefficient, and β_{ij} is an interactive coefficient.

Table 2
Design matrix of RSM with CCD in the each thermal hydrolysis pretreatment.

Run	Reaction temp. (°C)	Reaction time (min)	CH_4 yield (mL CH_4/g COD)
1	100	90	246.2 ± 11.2
2	200	30	221.7 ± 8.9
3	150	60	271.6 ± 10.2
4	200	90	243.2 ± 11.1
5	75	60	232.7 ± 10.7
6	150	60	273.2 ± 5.6
7	150	15	228.4 ± 7.1
8	225	60	231.7 ± 10.7
9	150	105	263.2 ± 6.8
10	100	30	234.2 ± 7.7
11	150	60	268.4 ± 11.3

(*Note:* mean ± standard deviation).

2.4. Batch and continuous tests

Batch tests for optimization of THP were performed using serum bottles with a working volume of 200 mL. Seeding sludge was input at 10% of the working volume and the remaining volume was filled with a mixture of substrate and distilled water. 5.0 M NaOH and 3.0 M HCl were used to fix the initial pH at 7.0 ± 0.2. Then, nitrogen gas was used to purge the headspace of each bottle. The biogas was measured using a glass syringe. Each condition was tested in triplicate.

The continuous tests were performed in continuous stirred tank reactors with a working volume of 3 L. The feed of reactor 1 (R1) and reactor 2 (R2) were untreated sludge and pretreated sludge (under optimum conditions based on batch tests), respectively. All reactors were filled with seeding sludge and distilled water (50:50), then operated on batch mode. Continuous operation started when biogas production and methane content reached 0.1 L/L·d and 40% in batch mode, respectively. Continuous reactors were operated for about 150 days. Continuous reactors were initially loaded with 1.4 kg COD/m^3·d, and then they increased from 1.4 kg/m^3·d to 4.4 kg COD/m^3·d. All samples were collected daily and measured three or more times.

2.5. Analytical and assay methods

The concentrations of COD, TS, VS, total nitrogen (T-N) and total phosphorus (T-P) were measured according to Standard Methods for the Examination of Water and Wastewater (APHA et al., 2005). VFAs were analyzed by a high-performance liquid chromatograph (HPLC, YL9100, Young-Lin Instrument Co., Korea) equipped with an ultraviolet (210 nm) detector and a 100 mm × 7.8 mm fast acid analysis column (Bio-Rad Laboratories, USA) using 0.005 M H_2SO_4 for the mobile phase. The liquid samples were pre-treated with a 0.45 μm membrane filter before injection to HPLC. The gas composition was analyzed using a gas chromatograph (GC, Gow Mac series 580, Gow-Mac Instrument Co., USA) equipped with a thermal conductivity detector (TCD) and 6 ft × 2 mm (i.d.) stainless steel Porapak Q (80/100 mesh) column. The operating temperatures of the injector, detector and column were maintained at 90, 80, and 50 °C, respectively. In addition, high purity (99.999%) helium gas was used as a carrier gas. The produced gas was adjusted to the standard temperature and pressure (STP).

Cumulative methane production curves in batch tests curves were described by the following modified Gompertz Equation:

$$M = P \cdot exp\left[-\exp\left\{\frac{R_m \cdot e}{P}(\lambda - t) + 1\right\}\right] \quad (3)$$

where M is the cumulative methane production, P is the maximum methane production, R_m is the maximum methane production rate, λ is the lag phase, t is time, and e is the exp(1) = 2.71828.

The dissolved organic compounds were fractionated using ultrafiltration. Fractionation of samples was performed using a 200 mL stirred cell (Amicon 8200; Millipore Corp. USA). Three membranes made of regenerated cellulose with different molecular weight cut-offs were used as follows: (1) 10 kDa and (2) 1 kDa. The initial volume was 100 mL. The samples were filtered through the membranes in series from (1) to (2). Each time a measured volume of 25 mL was retained for analysis, while the volume that permeated the membrane was passed