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Effect of hydrothermal pretreatment on anaerobic co-digestion of tofu residue and rice straw for biogas production

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Abstract

The effect of hydrothermal pretreatment (HTP) on the biogas production through the anaerobic co-digestion have been studied around the world. HTP of rice straw (100-180°C) has an impact on the biogas production and methane yield on anaerobic co-digestion and the optimal HTP condition was at a temperature of 160°C for rice straw. The maximum biogas production and methane yield was observed with HTP at 140°C for tofu residue, reaching 306 mL biogas/g VS and 200.1 mL CH₄/g VS respectively. These results can be used to design an effect anaerobic co-digestion process on management, effectively utilizing rice straw and tofu residue for renewable bio-energy production.

Keywords: Hydrothermal pretreatment (HTP), tofu residue, lignocellulosic waste, anaerobic co-digestion, biogas production

1. Introduction

Tofu residue (soybean curd residue, okara) is a byproduct of soybean processing products that is generating annually with an increasing in the world. (Guimarães *et al.*, 2018; Li *et al.*, 2012) [14, 19]. Tofu residue is a byproduct of soybean processing and the yield of tofu residue is increasing every year. Annually, tofu residue is generating around 2.8 million tones in China, while about 800 000 tons of tofu residue was produced as byproducts during soybean curd production in Japan. (Li *et al.*, 2013) [22]. In China, the annual yield of tofu residue is around 2.8 million tons, while in Japan 800 000 tons of tofu residue is produced in the processing of soybean curd.

Furthermore, tofu residue, as a byproduct of tofu production, could be used as the substrate for producing bioenergy and in many case it is simply buried down the earth and could be changed into waste. In some pig farms they use tofu residue for the feed of pigs by adding it to their feed directly. But it is impossible to use as the human food because of presence of trypsin inhibitor in it (Anderson and Wolf, 1995) [1]. Therefore, the treatment of tofu residue is a major challenge around the world to reduce the impacts on the environment and improve bioenergy. So it is important to solve the problem of how to treat tofu residue more economically and effectively.

Pretreatment of biomass is an effective method for improving substrate characteristics and hydrolysis in order to increase higher biogas production and decrease overall duration of anaerobic digestion (AD). Some researchers had focused on several pretreatment methods for increasing biogas production such as chemical, mechanical, ultrasonic, electrical and biological and hydrothermal pretreatment, which could be useful for enhancing hydrolysis of biomass during AD. (Talebnia *et al.*, 2010, Zheng *et al.*, 2014) [28, 33]. The hydrothermal pretreatment of these pretreatment methods has been found to be useful technique which makes biomass weaken structurally, enhancing solubilization and subsequent degradation during AD.

Currently, some studies have been focused on the HTP with AD technique for increasing biogas production. (Azizi A., *et al.* 2019) [4]. They have studied the effect of hydrothermal pretreatment on two-stage anaerobic digestion of source separated organics (SSO) in comparison to single stage AD. It mentioned that HTP was the efficient treatment method on two stage of AD process after the acidogenic fermentation stage at a hydrothermal

pretreatment temperature 170 °C, pressure 115 psi and retention time of 30 min. The hydrothermal pretreated wastes was produced in a continuous reactor using an autohydrolysis (steam explosion) at 150 °C, 45 psi and retention time of 40 min.

Then other compounds such as humic substances (humic acids and fulvic acids) were produced during the hydrothermal pretreatment of lignocellulosic biomass and humic acids was formed by phenolic compounds released from lignin decomposition. It was recently found that humic acids could be stimulated during the acidogenic stage, which it is a good electron transferring chains for catalyzing the reaction rate, but it has still the inhibiting effect on the methanogenic stage (Li *et al.*, 2019) [21]. Then, it was also found that humic substances can be used as the terminal electron acceptor during the anaerobic oxidation of methane (Valenzuela *et al.*, 2019) [29]. It could be explained the low biochemical methane potential (BMP) produced by hydrothermal pretreated lignocellulosic wastes could be acted the inhibition of AD process. Therefore, it needs to study how the low methane production yields was produced by hydrothermal pretreated lignocellulosic wastes.

This study focuses on two ways of waste management method by using hydrothermal pretreatment at different temperature (100-180 °C) on anaerobic co-digestion.

The aim of this study was to: (1) investigate the effect of hydrothermal treatment temperature on the variation of lignocellulosic biomass components; (2) assess the HTP temperature for rice straw and tofu residue on biogas and methane yield during anaerobic co-digestion; (3) explain the variation of the fermentation liquid after anaerobic co-digestion.

2. Materials and methods

2.1. Substrate and inoculum for anaerobic digestion and HTC substrate

Substrate 1. Tofu residue as raw substrate employed in this study was supplied from Hangzhou Soy Foods Co., Ltd, Hangzhou in China (Hongguanglanghua Co., Ltd). After being brought to laboratory, it was immediately separated into every sealing bag about 1kg and then stored in a refrigerator (-20 °C) for further use. Before the experiment, the frozen tofu residue was melted and whisked by a blender (CPEL-23, Shanghai Guosheng, China), to use as substrate 1. The mainly characteristics of tofu residue are as follows: moisture content 86.9±0.4%, pH 6.8±0.2, total solid (TS) 12.7±0.5%, volatile solid (VS) 12.2±0.3%, VS/TS 97.4±3.8%, soluble COD 3340±30 mg/L, NH₄-N content 367.8±15.7 mg/L. The characterization of the proximate and ultimate analysis were showed more detail in Table 1.

Table 1: Characteristics of hydrothermal pretreated rice straw, raw rice straw and inoculum.

Parameters	RS-100	RS-120	RS-140	RS-160	RS-180	Rice straw	Inoculum
TS (% w.b.)	11.6±0.4	12.3±0.5	11.8±0.3	10.9±0.2	10.5±0.3	90.2±0.4	3.45±0.18
VS (% w.b.)	10.7±0.3	11.5±0.3	11.0±0.4	10.1±0.3	9.4±0.3	9.3±0.3	1.73±0.12
VS/TS (%)	92.5±0.4	92.9±0.4	92.8±0.3	92.9±0.3	89.7±0.3	85.6±0.2	50.40±0.22
Moisture content (%)	88.4±0.4	87.7±0.3	88.2±0.4	89.1±0.3	89.5±0.4	9.8±0.3	96.55±3.27
pH	6.8±0.2	6.7±0.3	6.4±0.3	6.1±0.2	5.9±0.2	-	8.08±0.18
Ash content (% d.b.)	0.9±0.1	0.7±0.1	0.9±0.1	0.7±0.1	1.1±0.2	-	49.60±0.26

Note: All data were tested or measured on dry basis. Data are mean values ± standard error of three replicates. -: not measured.

RS: rice straw. d.b.: dry basis.

Substrate 2. Rice straw: The rice straw was collected from a farm in Haiyan, Zhejiang Province, China. One week before the experiment, rice straw was initially dried in the air to decrease the moisture content less than 10%, then subsequently cut into pieces of 425µm in length using a blender (CPEL-23, Shanghai Guosheng, China) and stored in plastic bags at room temperature for further use.

Hydrothermal pretreatment

Hydrothermal pretreated tofu residues and rice straw produced from different temperatures between 130 °C and 180 °C were used as substrates for anaerobic co-digestion. The method for obtaining hydrothermal pretreated tofu residues and tofu residue are as follows;

Before the experiment, substrate and deionized water was directly mixed and evenly stirred, which the ratio of substrate to deionized water is 1:1. Mixed material was transferred into a cylindrical stainless-steel reactor (4848,

Parr Instrument Company, USA) and poured into the Parr stirred reactor. The HTC process conditions were set between 130 and 180 °C, 3 °C/min heating rate and retention time was 1 h at the desired temperature. The reaction temperature was controlled by a Parr PID (proportional-integral-differentiate) temperature controller (4848 Parr Instrument Company, USA). The accuracy of the controller was ±2 °C. The pressure was not controlled, but monitored during hydrothermal carbonization. In this case it was around 8-12 bar. The content was stirred at 90 rpm continuously, stir rate was controlled by Parr stir speed controller. At the end of hydrothermal carbonization reaction, the heater was turned off and the Parr reactor was cooled down to a room temperature naturally. It usually takes 4-5 hours to cool down from experimental temperature to 25 °C, while pressure drops from 8-12 to 3-4 bar. After the temperature was cooled down to 25 °C, gas valve was turned on. Gaseous product was removed and liquid product was separated by vacuum filtration with 0.45 µm cellulose filter paper. The filtered LFHTC (liquid fraction of hydrothermal carbonization) of tofu residues and rice straw was stored in a -20 °C refrigerator for analysis and further use. The characteristics of hydrothermal pretreated tofu residues and rice straw are listed in Table 2.

Table 2: Characteristics of hydrothermal pretreated tofu residue, raw tofu residue and inoculum.

Parameters	HPTR-100	HPTR-120	HPTR-140	HPTR-160	HPTR-180	Raw tofu residue	Inoculum
TS (% w.b.)	8.25±0.27	7.24±0.27	7.14±0.25	6.63±0.24	6.12±0.26	13.28±0.46	3.45±0.18
VS (% w.b.)	6.8±0.23	6.93±0.23	7.02±0.27	6.33±0.25	5.71±0.23	12.94±0.45	1.73±0.12
VS/TS (%)	83.4±0.27	95.8±0.27	98.25±0.25	95.54±0.28	91.71±0.29	97.43±0.35	50.40±0.22
Moisture content (%)	91.8±3.52	92.8±3.52	92.86±2.94	93.37±3.52	93.88±2.96	86.72±3.43	96.55±3.27
pH	5.36±0.19	4.95±0.19	4.57±0.17	4.39±0.17	4.0±0.16	5.76±0.14	8.08±0.18
Ash content (% d.b.)	0.10±0.04	0.11±0.04	0.17±0.04	0.29±0.06	0.29±0.07	2.57±0.15	49.60±0.26

Note: All data were tested or measured on dry basis. Data are mean values ± standard error of three replicates. -: not measured.

RS: rice straw. d.b.: dry basis.

Inoculum

The digested sewage sludge was used as inoculum and supplied by an anaerobic fermentation tank that was normally running in Hangzhou Xiaoshan environment, Co. LTD (Hangzhou Environment Group Co., Ltd, China). The digested sewage sludge was characterized as follows: it contained 3.5±0.4% total solids (TS), 1.7±0.3% volatile solids (VS) and VS to TS was 50.4±0.8%, pH 8.1±0.1, the C/N ratio of inoculum was 7.44±0.14. The characteristics of inoculum are listed in Table 1.

2. 2 Anaerobic digestion and design of experiment

The experiment was carried out in batch mode, 500-mL glass bottles were used as vessels of the bioreactor. The TS of all AD bioreactors was set to 7% by adding the designated amount of substrate, inoculum and tap water. All bioreactors were sealed using rubber plugs and screw caps that was fitted with connection tubes, then there were flushed with nitrogen gas for 5 min to remove oxygen. It was carried out the incubation condition at 37 ± 1 °C for 25 days in a water bath (Shanghai Jinghong Company, China), shaking manually for 1 min twice a day before taking biogas for production yield and methane content. The produced biogas was trapped in a 1-L glass bottle, loading with a diluted hydrochloric acid solution (pH < 3 to prevent CO₂ dissolution). Daily biogas production yield was measured by using the water displacement method and it was produced in the bioreactor displaced acid liquid which in turn was gathered in a 1-L glass bottle. (Mustafa *et al.*, 2016) [24].

The batch anaerobic co-digestion of rice straw with hydrothermal pretreated tofu residue at different temperature of 100, 120, 140, 160 and 180 °C was carried out with a mixture ratio in the condition of volatile solid (3gVS). (Choe *et al.*, 2020 [11]). One group as the control group was fermented with tofu residue and rice straw with inoculum. 11 experiment groups for anaerobic co-digestion treatments was carried out twice.

Substrate/inoculum VS ratio was 0.5. In this study it was decided as 1.5 gVS of substrate and 3.0 gVS of inoculum (Choe *et al.*, 2019, Xu *et al.*, 2016) [10, 30]. The pH of all bioreactors was adjusted to around 7 by using 1 M NaOH and 1 M HCl solutions. Cumulative biogas production yield was observed by daily measurement for the first eight days, and every 2 days thereafter until there was no biogas yield for 3 days. Special biogas production was adjusted to standard temperature (0 °C) and pressure (1 atm) using equation (1) (Kafle & Kim, 2013) [18].

$$V_{STP} = \frac{V_T \times 273 \times (760 - p_w)}{(273 + T) \times 760} \quad (1)$$

Where V_{STP} is the volume of biogas (L) measured at standard temperature (0 °C) and pressure (1 atm), V_T is the volume of biogas measured at temperature T (L), p_w is the water vapor pressure as a function of temperature (mm Hg). T is the temperature of the gas (°C).

2.3 Chemical analysis

The total solid (TS), volatile solid (VS) and ash content were analyzed by heating the samples in an oven (SHANGHAI BOXUN INDUSTRY & COMMERCE Co., Ltd, China) at 105 °C, for 24 h, a furnace (Tianjin Zhonghuan Experiment Electric Furnaco Co., Ltd, China) at 900 °C for 7 min, and a furnace (Tianjin Zhonghuan Experiment Electric Furnaco Co., Ltd, China) at 575 °C for 2 h, respectively (APHA, 2005) [3]. The pH was measured by using a pH meter (Testo 205 AG Germany) and the samples were stirred manually before the measuring. LFHTC of tofu residues was used to determine the chemical oxygen demand (COD) by COD measurement device (Hach, LCK 514) and determination of COD (Hach, LCK 514) was performed by using the test kits containing dichromate. The method involved a simple digestion and the changes in color were measured by using quantitative Hach spectrophotometer (DR/2800, APHA, 2005) [3]. Then, proximate and ultimate analyses were carried out using the method that the carbon, hydrogen, nitrogen and sulphur content of LFHTC of tofu residue was calculated by drying a known volume of process water at 105 °C over a period of 48 h to reduce loss of volatile organic compounds. The elemental determination of carbon, nitrogen, hydrogen and sulfur content of the sample was measured with a Vario EL elementary analyzer (Elementar Trading Shanghai Co., Ltd. China) (Carter and Barwick, 2011) [9]. The pH of raw tofu residue in the water-extractable fraction was determined around 5.76-5.90 (dry tofu residue mixed with deionized water at a ratio of 1:10, w/v). (Zhang *et al.*, 2014) [31].

Biogas components were determined by using a gas chromatograph (GC-2014, Shimadzu, Japan) equipping with a thermal conductivity detector. The temperatures of the column oven, injector port, and detector were 100, 120, and 120 °C respectively. Biogas samples were taken from the bioreactors by using a special syringe (injection volume was 200 µL) and injected to the thermal conductivity detector of the gas chromatograph. Biogas composition was measured using standard biogas (CH₄: 50.17% v/v, CO₂: 9.96% v/v, H₂: 30.04% v/v, and N₂: 9.83% v/v) after 5 days of digestion, and then every 3 days thereafter and carried out in three replicates.

The biomass components of raw rice straw, tofu residue, HTP tofu residue and HTP rice straw were measured by the

National Renewable Energy Laboratory (NREL) standard analytical procedure (Sluiter *et al.*, 2008) [27]. Soluble sugars in supernatant, acid hydrolysate and enzymatic hydrolysate, and 5-HMF in filtered liquid after catalytic reaction were determined by HPLC (Waters e2695, USA) equipped with a Bio-Rad Aminex HPX-87H column (300 mm × 7.8 mm, USA) and 2414 refractive index detector (USA). The column was kept at 50 °C with 5 mm H₂SO₄ solution as mobile phase at a flow rate of 0.6 mL/min, and the detection time was 30 or 60 min. The quantitative analysis was conducted by calibration with standard sugar concentration. Soluble lignin in acid hydrolysate and supernatant was determined by UV-Vis Spectrophotometer (Cary 60, Agilent Technologies, USA) (Jin *et al.*, 2020) [17]. The conversions of cellulose, hemicellulose, acid-soluble lignin and acid-insoluble lignin content after catalytic reaction were calculated as follows:

$$\text{Water content } A = \frac{m' - m_0}{m_0} \times 100\% \quad (A < 10\%) \quad (2)$$

Where

m represents the mass of corresponding components (g).

m₀ means the mass of substrate before drying.

m' means the mass of substrate after drying.

Standard curve

- Standard curve of glucose : Y=101937X-5015.6 R²=0.9999 (3)
- Standard curve of xylose : Y=115172X-8942.6 R²=0.9982 (4)
- Standard curve of arabinose : Y=102193X-8802.1 R²=0.9845 (5)

**Linear relationship between peak area and concentration

C glucose=X₁, X₂, X₃

Cellulose content (%) =

$$\frac{C_{glucose} \times 87 \times 10^{-3} \times 0.9}{0.300 \times (100\% - A)} \times 100\% \quad (6)$$

Hemicellulose content (%) =

$$\frac{(C_{xylose} + C_{arabinose}) \times 87 \times 10^{-3} \times 0.88}{0.300 \times (100\% - A)} \times 100\% \quad (7)$$

Lignin content

Acid-soluble lignin content (%) =

$$\frac{U_{vabs} \times 87 \times 10^{-3} \times \text{Dilution factor}}{110 \times 0.3 \times (100\% - A)} \times 100\% \quad (8)$$

Acid insoluble lignin content (%) =

$$\frac{m_1 - m_2}{0.300 \times (100\% - A)} \times 100\% \quad (9)$$

Calculation of total C/N of the mixed materials

The total C/N ratio of the mixed material was calculated using Eq. (10).

$$\frac{C}{N_{Total}} = \frac{C_1 X_1 + C_2 X_2 + C_3 X_3 + \dots}{N_1 X_1 + N_2 X_2 + N_3 X_3 + \dots} = \frac{\sum C_i X_i}{\sum N_i X_i} \quad (10)$$

Where C (% TS) is the mass fraction of carbon in an individual material, N (% TS) is the mass fraction of nitrogen in an individual material, and X_n (g TS) is the amount of the individual material in the mixture (n = 1, 2, 3.i).

After anaerobic digestion, the ammonium nitrogen content in the digested liquid was analyzed using ammonium nitrogen (NH₄-N) measuring equipment (Cary 60 UV-Vis, Agilent Technologies CO., LTD, China). The LFHTC was used to determine the chemical oxygen demand (COD) by dichromate oxidation method (Hach, LCK 514 and Hach spectrophotometer DR/2800, China) (APHA, 2005) [3].

The surface structures of untreated and hydrothermal pretreated rice straw were further examined employing field launch scanning electron microscope (SEM) using an SU8010 microscope (Hitachi, Japan).

3. Results and discussion

3.1 Variation of cellulose, hemicellulose and lignin after hydrothermal pretreatment of rice straw

The Variation of cellulose, hemicellulose and lignin after hydrothermal pretreatment was shown in Table. 3. As shown in Table 3, the cellulose contents of rice straw were gradually decreased from 22.04 to 18.79%, which was treated at hydrothermal pretreatment temperature between 100 to 180 °C. And the hemicellulose contents of hydrothermal treated rice straw were also reduced when the HTP temperature was increased from 100 to 180 °C. However, the acid soluble-lignin contents were increased from 4.30 to 6.21%, while the acid insoluble-lignin contents were gradually decreased, when the hydrothermal pretreatment temperature was increased from 100 to 180 °C. Antwi E. *et al.* (2019) [2] recently carried out the anaerobic digestion of cocoa pods residues by using hydrothermal pretreatment at different temperature and retention time (150-220 °C, 5-15min), which was found that the effect of hydrothermal pretreatment on the biogas yield. They have reported that the optimal condition for biogas yield was 150 °C, 15 min, which was due to the hydrolysis and liquefaction of lignocellulosic biomass.

Table 3: Variation of biomass components of rice straw and tofu residue after hydrothermal pretreatment

Number	Symbol	Cellulose content (%)	Hemicellulose content (%)	Acid-soluble lignin content (%)	Acid-insoluble lignin content (%)
1	RS(rice straw)	22.04	14.02	4.30	20.94
2	TR(tofu residue)	11.64	14.88	4.59	2.39
3	RS-100	21.47	13.94	4.74	20.63
4	RS-120	20.83	13.17	5.31	20.28
5	RS-140	19.27	12.62	5.62	19.71
6	RS-160	18.92	12.43	5.93	19.04
7	RS-180	18.79	12.18	6.21	18.79

8	TR-100	11.64	14.72	4.68	2.27
9	TR-120	11.42	14.48	4.92	2.05
10	TR-140	11.34	14.23	5.16	1.87
11	TR-160	11.17	14.09	5.24	1.81
12	TR-180	11.09	13.98	5.31	1.79

Table. 3 also showed the variation of biomass components after hydrothermal pretreatment of tofu residue. As shown in Table. 3, the cellulose contents of tofu residue were gradually decreased from 11.64 to 11.09% after hydrothermal pretreatment between 100 to 180 °C. Also the hemicellulose contents were decreased from 14.02 to 13.98% at the same HTP condition. Anyhow, the acid soluble lignin contents were increased from 4.59 to 5.31%, while the acid insoluble lignin contents were decreased from 2.39 to 1.79%, respectively. This result was matched exactly with the research of Ding *et al.* (2019) [13], who have already reported that the hydrogen and methane yields from hydrothermally pretreated food waste under the optimum condition (140 °C, 20 min) through two-stage fermentation. They found that the increase of soluble carbohydrates at the temperatures of 100-140 °C could be explained by the following reasons: when food waste was subjected to hydrothermal pretreatment, the large-molecular-weight carbohydrate polymers (e.g., starch, cellulose, and hemicellulose) were hydrolyzed into small-molecular weight oligosaccharides and monosaccharides (e.g., glucose and xylose) (Li *et al.*, 2016) [20]. However, some soluble

sugars, such as hemicellulose derivatives, were further degraded into short-chain VFAs, such as acetic acid (Monlau *et al.*, 2014) [23], thus decreasing total carbohydrates. So, it was demonstrated that hydrothermal pretreatment was a suitable approach in pretreatment of tofu residue to promote anaerobic fermentation performance. Author's knowledge, subjecting to hydrothermal pretreatment at 120 °C for 30 min facilitates the solubilization of tofu residue (i.e., carbohydrates and proteins), thus benefiting the anaerobic co-digestion with lignocellulosic biomass for methane production.

3.2 SEM observation

Fig. 1 showed SEM images of hydrothermal pretreated rice straw at different temperature of 160 °C for 30 min and untreated rice straw. For untreated rice straw the rigid, arranged fibrils and compact structure were clearly visible. (Fig. 1) Rice straw of hydrothermal pretreated at low temperature 100, 120 and 140 °C induced a slight level of damage to the structure and caused micro cracking of the surface.

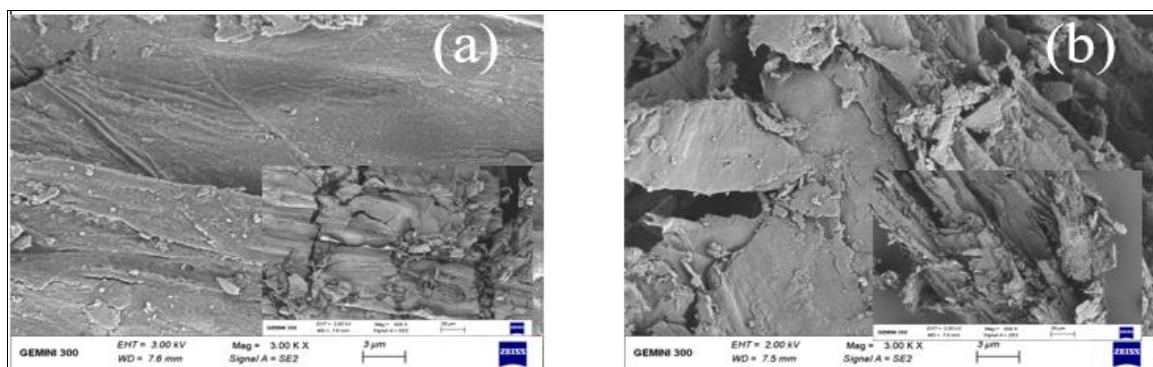


Fig 1: SEM image of untreated rice straw (a) and hydrothermal pretreated rice straw at temperature of 160 °C (b)

However, rice straw of hydrothermal pretreated temperature at 160 °C resulted in increased structural decomposition. As shown Fig. 1, lignin fibers were damaged, while the secondary cell walls were exposed. It was caused due to breaking down the surface structure of rice straw by high hydrothermal temperature and resulted into the increasing surface area of biomass. It was suggested that the surface area extension of biomass might be more useful for microorganisms on digestion of biomass during anaerobic fermentation. Comparing images of rice straw at different hydrothermal pretreatment temperatures showed increasing structural variation like pores size and gaps in the rice straw surface structure. The surface area obviously increased while hydrothermal pretreatment temperature increased from 100 to 160 °C. In the case of rice straw of HTP temperature at 180 °C, the variation of surface structure was not obviously compared with the rice straw at HTP temperature of 160 °C. Anyhow, slurry of HTP rice straw could contain more inhibition materials such as phenol (Ding L. *et al.*, 2017) [13], so it could cause inhibition of AD, decreasing biogas production. These results were agreed

with the cumulative biogas production yield on anaerobic co-digestion of tofu with HTP at different temperature between 100 and 180 °C. Fig. 1 showed the increasing surface area of rice straw, large pore size and lignin removal, while hydrothermal pretreat temperature was increased gradually.

Antwi E *et al.* (2019) [2] recently reported the effect of hydrothermal pretreatment of cocoa pods residues on anaerobic digestion for possibility of increasing biogas production yield. It was found that the optimal HTP temperature and retention time was 150 °C and 15 min, respectively. Therefore, this result was matched exactly with the recent research (Antwi E *et al.* 2019) [2], which was the optimal pretreatment condition was HTP temperature of 160 °C and retention time of 30 min for rice straw hydrothermal pretreatment for anaerobic co-digestion with tofu residues.

3.3 Anaerobic co-digestion of HTC pretreated rice straw and raw tofu residue

3.3.1 Effect the HTC temperature of rice straw on biogas yield

The anaerobic co-digestion of hydrothermal pretreated rice straw (produced at various HTC temperature of 100, 120, 140, 160 and 180 °C) and raw tofu residue was conducted with entitled RS-100+TR, RS-120+TR, RS-140+TR, RS-160+TR and RS-180+TR. Anaerobic co-digestion of raw rice straw and raw tofu residue as the control group was used with inoculum. Fig. 2 shows the cumulative biogas yield (expressed as mL/g VS) during 25 days of anaerobic co-digestion of mixed HTP (hydrothermal pretreated) rice straw and raw tofu residue.

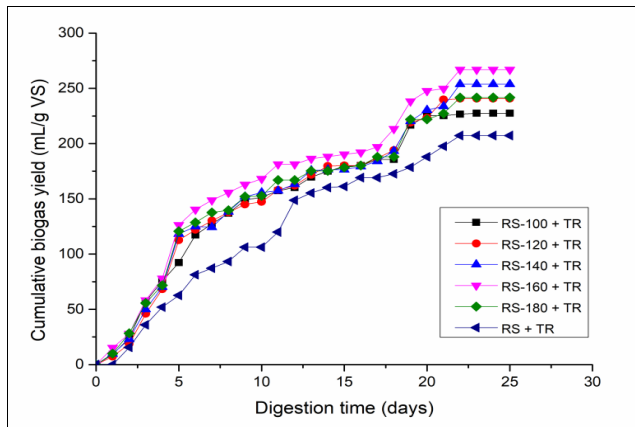


Fig 2: Cumulative biogas yield during 25days anaerobic co-digestion of mixed HTP rice straw and raw tofu residue at HTC temperature of 100, 120, 140, 160 and 180 °C. Data are means of two replicates.

As shown Fig. 2, cumulative biogas yield for anaerobic co-digestion of RS-100+TR, RS-120+TR, RS-140+TR, RS-160+TR and RS-180+TR was 236, 240.7, 253.7, 267.0 and 241.7 mL/g VS, respectively. All anaerobic co-digestion of HTP rice straw with tofu residue showed increasing tendency in cumulative biogas yield more than control. Then cumulative biogas yield of the control group for anaerobic co-digestion of rice straw and tofu residue was 207.3 mL/g VS and it was detected as the lowest biogas production yield. The highest cumulative biogas yield was detected with RS-160+TR and it was increased 28.8% more than control. Then, the increased biogas yield was ranged RS-140+TR, RS-180+TR, RS-120+TR and RS-100+TR. It was found that biogas production potential could be increased after hydrothermal pretreatment, and hydrothermally pretreated rice straw was useful for anaerobic co-digestion with tofu residue. The increasing cumulative biogas yield may be due to enhanced hydrolysis of rice straw through hydrothermal treatment process and it could improve the biodegradability during anaerobic co-digestion of HTP rice straw with tofu residue.

Jin et al (2014) [17] found that under subcritical conditions the relatively strength of hydrogen bonds of the water molecules of evaporated water have been reduced, meanwhile it was decreased the dielectric constant. Then the ionic products $[H^+]$ and $[OH^-]$ were also increased to a maximum to 10^{-12} under the subcritical condition considerably (Pedersen *et al.*, 2016) [26] and two factors have been made the efficient solvent for hydrolysis and liquidation of lignocellulosic biomass such as rice straw. This result was agreed to the opinions of Antwi E. *et al.* (2019) [2] who recently have carried out the hydrothermal pretreatment of cocoa pods for increasing biogas yield during anaerobic digestion, which was confirmed by using a

central composite surface response methodology method and effect of hydrothermal treatment temperature and retention time for hydrolysis of lignocellulosic biomass. And it was reported that the optimum biogas yield was obtained at hydrothermal pretreatment condition of 150 °C and 15 min, measuring the highest biogas yield of 526.38 L (N)/kgVS with a severity of 2.65.

Fig. 3 showed the cumulative biogas yield for anaerobic co-digestion of rice straw with hydrothermal pretreated tofu residue.

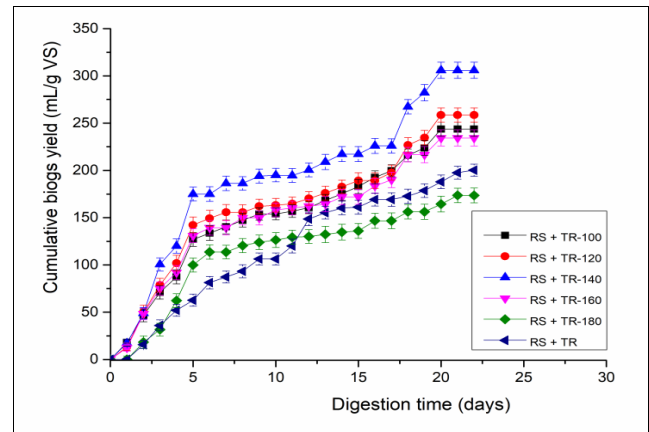


Fig 3: Cumulative biogas yield during 25days anaerobic co-digestion of mixed rice straw and HTP tofu residue at HTC temperature of 100, 120, 140, 160 and 180 °C. Data are means of two replicates.

As shown Fig. 3, it was obtained the cumulative biogas yields for RS+TR-100, RS+TR-120, RS+TR-140, RS+TR-160 and RS+TR-180 were 243.7, 258.7, 306, 234.3 and 173.6 mL/g VS, respectively. Ultimate biogas production for anaerobic co-digestion of rice straw with HTP tofu residue ranged from 173.6 mL/g VS (RS+TR-180) to 306 mL/g VS (RS+TR-140). Fig. 3 showed that the highest cumulative biogas yield was obtained during anaerobic co-digestion of RS+TR-140 experimental group, which was treated at hydrothermal pretreatment temperature of 140 °C, while the lowest cumulative biogas yield was RS+TR-180. It was found that the cumulative biogas yield was rose initially from 243.7 to 306 mL/g VS, then it was decreased gradually from 306 to 173.6 mL/g VS. It indicated that HTP was used to enhance tofu residue solubilization and the solubiliziation of tofu residue was increased at HTP temperature of 100 and 140 °C while it was decreased when HTP temperature increased from 140 to 180 °C.

Ding L. *et al.* (2017) [13] already have investigated the hydrothermal pretreatment of food waste to enhance hydrogen and methane co-production during anaerobic digestion and it was found that hydrothermal pretreatment temperature from 100 to 140 °C could be increased the soluble carbohydrate content of food waste and then decreased it from 140 to 200 °C. So it was demonstrated that soluble carbohydrate content of food waste was negatively co-related with increasing HTP temperature due to enhancing degradation and Maillard reaction through hydrothermal pretreatment at temperature of 100 to 200 °C. These results were agreed to Ding L. *et al.* (2017) [13], who have already conducted anaerobic digestion of food waste through hydrothermal pretreatment process at 140 °C, 20 min for two-stage H_2 and CH_4 co-production while increasing soluble carbohydrate content. According to the

author's knowledge, the co-digestion of rice straw with tofu residue which was treated at 140 °C, 30 min provides a practical residues management method for bioenergy production.

3.3.2 Effect the HTC temperature of rice straw on methane yield

Methane content and final methane yield for the anaerobic co-digestion of hydrothermal pretreated rice straw with tofu residue during 25 days was shown Fig. 4.

For all experimental groups in Fig.4, the average methane content was ranged from 58.6 to 65.4%. The highest methane content was 65.4% obtained from RS-160+TR, while the lowest methane content was 58.6% from RS-100+TR, which was in the normal range of methane content produced during anaerobic digestion (Campuzano, R., 2016) [7]. It was found that methane content was included the range

of normal methane content in general, so it was supposed that anaerobic co-digestion of HTP rice straw with tofu residue was carried out normally. Then, the range of methane content was similar to a recent study, which has studied hydrothermal pretreatment on the biogas yield of cocoa pods residues on anaerobic digestion (Antwi E. *et al.*, 2019). And Nyktari *et al.* (2017) [2, 25] have reported that methane content in anaerobic fermentation using hydrothermal carbonization of sewage sludge with different reaction conditions (140°C-200 °C for 30-240 min) was 63.5-77.7%.

The final methane yield for anaerobic co-digestion of tofu residue with hydrothermal pretreated rice straw (produced at various HTP temperature of 100, 120, 140, 160 and 180 °C) was ranged 138.3, 145.1, 159, 174.6 and 148.6 mL CH₄/g VS.

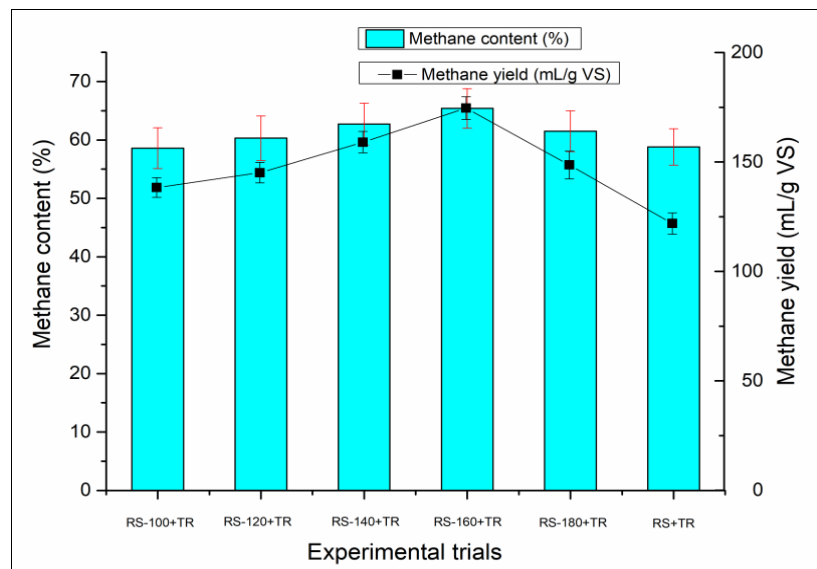


Fig 4: Average methane content and total methane yield produced from anaerobic co-digestion of hydrothermal pretreated rice straw and tofu residue. Data are means of two replicates.

As shown Fig. 4, the highest final methane yield was observed with RS-160+TR (174.6 mL CH₄/g VS), while the lowest methane yield 121.9 mL CH₄/g VS was noticed with RS+TR (control). It was found that the methane yield was gradually increased from 138.3 (RS-100+TR) to 174.6 mL CH₄/g VS (RS-160+TR) and it was supposed that increasing HTP temperature between 100 and 160 °C, enhancing the final methane yield gradually. However, the final methane yield was decreased at the HTP temperature of 160 °C, in the case of RS-180+TR experimental groups. It was observed with 148.6 mL CH₄/g VS, which was decreased 14.9% more than optimal condition of RS-180+TR. In summary, it was found that the final methane yield was increased after hydrothermal pretreatment compared with control group (anaerobic co-digestion of raw rice straw with raw tofu residue). So according to the author's knowledge, the hydrothermal pretreatment of rice straw at 100 to 180 °C for anaerobic co-digestion with tofu residue could increase final methane yield, then it will provide a practical lignocellulosic biomass and food waste management method for renewable bioenergy production.

Fig. 5 showed the methane content and final methane yield for anaerobic co-digestion of rice straw with HTP tofu residue during 25 days fermentation.

As shown in Fig. 5, the methane contents in all experimental groups were ranged 64.9, 65.4, 68.3, 63.2, 59.7 and 58.8%, which were obtained with RS+TR-100, RS+TR-120, RS+TR-140, RS+TR-160, RS+TR-180 and RS+TR, respectively. The highest methane content was noticed with 68.3% (RS+TR-140), while the lowest methane content was 58.8% (RS+TR), which were within the normal range of methane content during anaerobic digestion (Campuzano, R., 2016) [7]. It was also demonstrated that methane content was included the range of normal methane content in all experimental groups, so it was suggested that anaerobic co-digestion of rice straw with hydrothermal pretreated tofu residue was carried out regularly. Also, the scope of methane content was similar to a recent study, which has studied hydrothermal pretreatment on the biogas yield of cocoa pods residues on anaerobic digestion. (Antwi E. *et al.*, 2019) [2].

The final methane yield for anaerobic co-digestion of rice straw with HTP tofu residue were ranged 158.1, 176.7, 200.1, 148.1 and 119.2 mL CH₄/g VS, which was observed with RS+TR-100, RS+TR-120, RS+TR-140, RS+TR-160 and RS+TR-180, respectively. (Fig. 5)

As shown Fig.5, the highest methane yield was noticed with 200.1 mL CH₄/g VS (RS+TR-140), while the lowest

methane yield was 119.2 mL CH₄/g VS (RS+TR-140). It was found that the final methane yield was increased from 158.1 mL CH₄/g VS to 200.1 mL CH₄/g VS after hydrothermal pretreatment at temperature of 100 and 140 °C and then decreased gradually 200.1 mL CH₄/g VS to 119.2

mL CH₄/g VS, which was pretreated at temperature of 140 to 180 °C. However, the methane yield of RS+TR-180 experimental groups was decreased 2.3% compared with control (RS+TR) 121.9 mL CH₄/g VS.

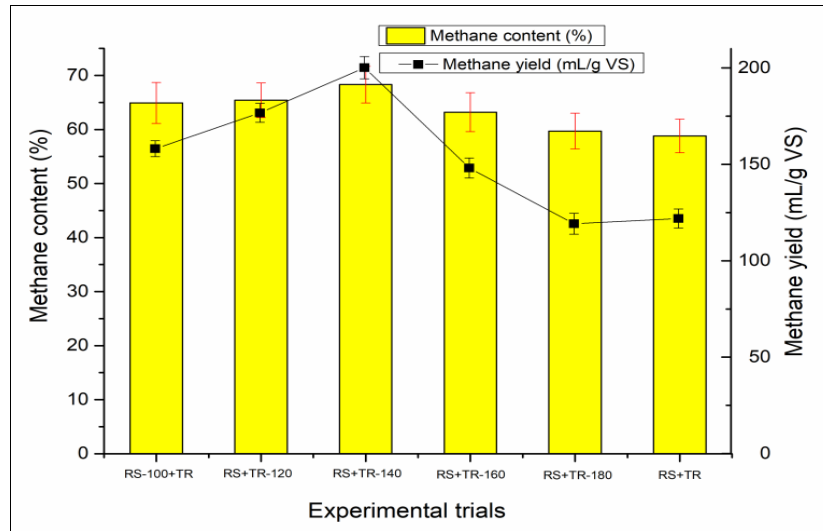


Fig. 5. Average methane content and total methane yield produced from anaerobic co-digestion of rice straw and hydrothermal pretreated tofu residue. Data are means of two replicates.

In the case of RS+TR-180, the final methane yield was lower than the control, and it was due to the components produced during Maillard reaction at hydrothermal pretreatment temperature 180 °C. (Danso-Boateng *et al.* 2015) [12]. They have reported that Maillard reaction products were identified in the liquid fractions following hydrothermal carbonization at 180 and 200 °C, so it was suggested that methane production process could be inhibited by Maillard reaction products produced at HTC pretreatment temperature 180 °C, and the reaction products could inhibit the anaerobic fermentation for methane production by methanogenic microorganisms, so the methane yield could be decreased lower than control. Finally, it was suggested that hydrothermal pretreatment between 100 and 160 °C could increase the tofu residue anaerobic fermentation for methane yield, but methane yield

initially increased from 100 to 140 °C and then decreased while HTP temperature increased. So, these results indicate that hydrothermal pretreatment could be used to facilitate tofu residue solubilization for increasing biogas production, then hydrothermal pretreatment temperature of 140 °C would be provided a practical anaerobic co-digestion method of tofu residue with rice straw to treat waste for renewable biogas production.

3.3. The characteristics of co-digested liquid of rice straw and tofu residue with hydrothermal pretreatment

3.3.1. Variation of sCOD after anaerobic co-digestion of HTP rice straw with tofu residue

The soluble COD of co-digested liquid of hydrothermal pretreated (HTP temperature from 100 to 180 °C) rice straw and tofu residue was shown in Fig.6

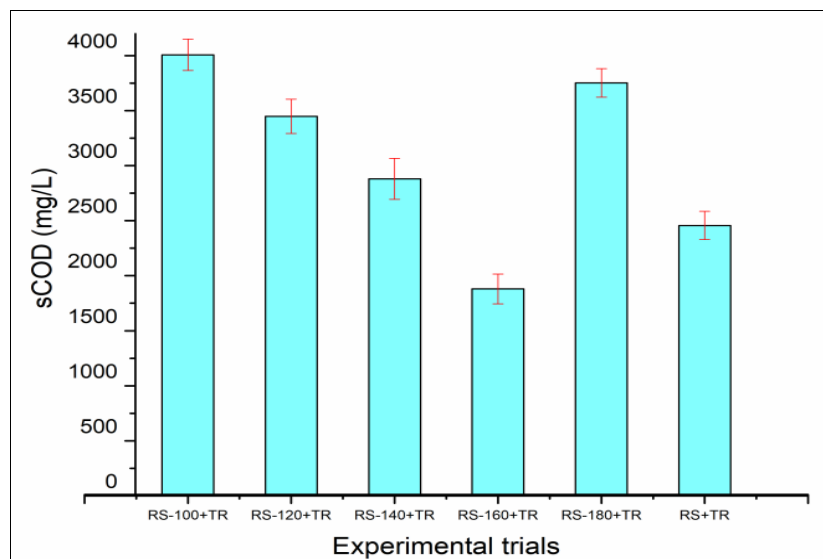


Fig 6: The soluble COD of fermentation liquid after anaerobic co-digestion of HTP rice straw with tofu residue

The soluble COD of co-digested liquid of HTP rice straw and tofu residue at different HTP temperature were ranged to 4008, 3449, 2880, 1880 and 3753 mg/L, which were obtained with RS-100+TR, RS-120+TR, RS-140+TR, RS-160+TR and RS-180+TR, respectively (Fig. 6). The highest sCOD value was noticed with RS-100+TR experimental group, while the lowest sCOD was obtained with RS-160+TR 1880 mg/L. During the period of anaerobic co-digestion of HTP rice straw with tofu residue, HTP RS (hydrothermal pretreated rice straw) and tofu residue are hydrolyzed by hydrolysis bacteria into sCOD. Through the sCOD fermentation, sCOD was changed into VFAs (volatile fatty acids) and then finally produced biogas by methanogenic microorganism on anaerobic fermentation.

As shown in Fig.6, the sCOD values were gradually decreased when hydrothermal pretreatment temperature of rice straw were increased from 100 to 160 °C, then it was suddenly increased from 1880 to 3753 mg/L at HTP temperature of 180 °C. In the case of RS+TR-100 and RS+TR-180, sCOD values were higher than the control (RS+TR), it was indicated that anaerobic co-digestion of rice straw with combination of hydrothermal pretreatment at 100 and 180 °C could not be carried out normally. So, it was supposed that hydrothermal pretreated rice straw could contain some indigestible components produced during the hydrothermal carbonization, which inhibited the anaerobic

co-digestion with tofu residue. Therefore, soluble COD of fermented liquid during anaerobic co-digestion of HTP rice straw with tofu residue was found to be higher than control experimental groups. This result was agreed to Antwi E. *et al.* (2019) [2] who reported lignocellulosic biomass had the possibility of increasing the biogas yield by hydrothermal pretreatment through anaerobic fermentation.

3.3.2. Variation of sCOD after anaerobic co-digestion of rice straw with HTP tofu residue

Fig. 7 shows the variation of soluble COD (sCOD) after anaerobic co-digestion of rice straw with HTP tofu residue. As shown in Fig. 7, sCOD values of rice straw and HTP tofu residue were ranged to 2153, 1950, 1823, 2110 and 3056 mg/L, which were observed with RS+TR-100, RS+TR-120, RS+TR-140, RS+TR-160 and RS+TR-180. The lowest sCOD was observed with RS+TR-140 as 1823 mg/L, while the highest sCOD was observed with RS+TR-180 as 3056 mg/L, respectively. It was found that sCOD value was gradually decreased when hydrothermal treatment temperature was increased from 100 to 140 °C, so it could be useful for hydrolysis on anaerobic bacteria into soluble COD. However, increasing the HTP temperature over 160 and 180 °C, sCOD value was 2110 and 3056 mg/L and then sCOD of RS+TR-180 experimental group was increased more than control (RS+TR 2457 mg/L).

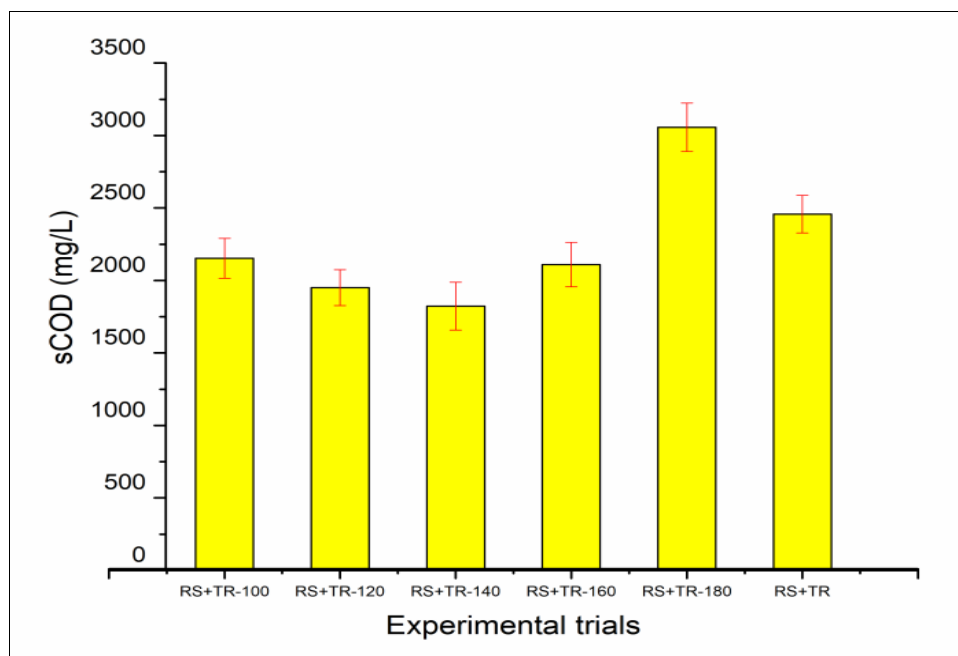


Fig 7: The soluble COD of fermentation liquid after anaerobic co-digestion of rice straw with HTP tofu residue

Therefore, it was supposed that hydrothermal pretreated tofu residue at temperature over 140 °C could contain some indigestible components produced during the hydrothermal carbonization, which inhibited the anaerobic co-digestion with rice straw. So, the sCOD of fermented liquid during anaerobic co-digestion of rice straw with HTP tofu residue at HTP temperature more than 140 °C was found to be higher than control groups. These results were quite similar to the recent report of Huang *et al.* (2019) [16] and showed

that the synergistic effect of microorganisms contained in TR and WAS may be responsible for the enhancement of lignocellulose hydrolysis and VFA generation.

3.3.3 Variation of NH₄-N in fermentation liquid after anaerobic co-digestion

The variation of NH₄-N contents of fermentation liquid after anaerobic co-digestion of HTP rice straw and tofu residue is shown in Fig. 8

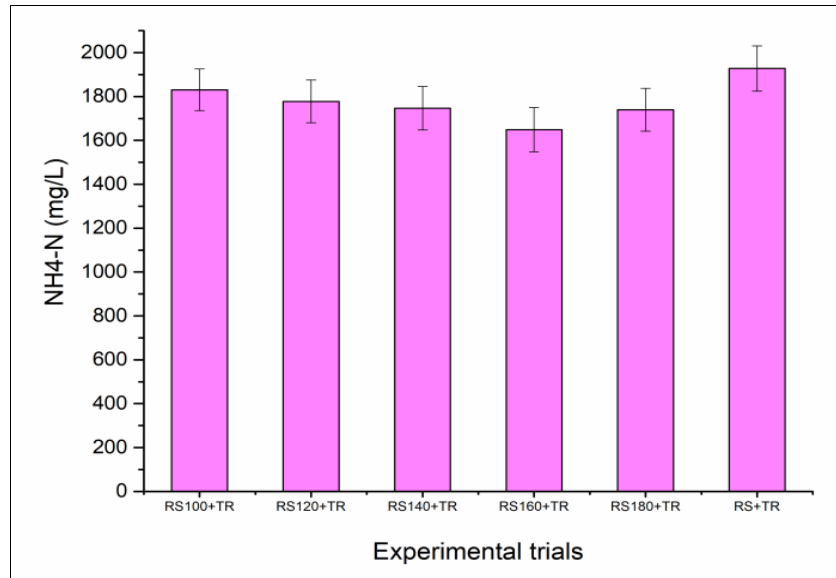


Fig 8: NH₄-N contents of fermentation liquid after anaerobic co-digestion of HTP rice straw and tofu residue. Data are means of two replicates.

As shown in Fig. 8, the NH₄-N contents of fermentation liquid were ranged to 1830.7, 1777.8, 1746.6, 1648.9 and 1739.8 mg/L, which were measured with RS-100+TR, RS-120+TR, RS-140+TR, RS-160+TR and RS-180+TR, respectively. The highest NH₄-N content was observed with RS-100+TR experimental group, while the lowest NH₄-N content was observed with RS-160+TR. It was found that the NH₄-N content was gradually decreased with the increasing hydrothermal pretreatment temperature from 100 to 160 °C, then was increased from 1648.9 mg/L (RS-160+TR) to 1739.8 mg/L (RS-180+TR).

Then it was found that except the RS-160+TR experimental group, all experimental groups for anaerobic co-digestion of hydrothermal pretreated rice straw with tofu residue were higher than the inhibition threshold of 1700 mg/L (Bujoczek G., 2000). Villamil J. A. (2019) ^[6] and the final total NH₄-N content was within the range of 600-800 and 460-650 mg N/L for flocculent sludge (FS) and granular sludge (GS) in the anaerobic co-digestion of LF of dewatered waste activated sludge with PSS, respectively. It was found out that NH₄-N content was much higher than the critical inhibitory concentration (1700 mg/L) for

methanogenic microorganism. It showed that the anaerobic co-digestion of LF of dewatered waste activated sludge with PSS were carried out to estimate total ammonium nitrogen after anaerobic co-digestion, which was found that AD process was not inhibited by NH₄-N during the anaerobic co-digestion of LF with PSS. However, in this study the NH₄-N contents of RS-100+TR, RS-120+TR, RS-140+TR and RS-180+TR experimental groups were higher than 1700 mg/L, demonstrating that the anaerobic co-digestion of HTP rice straw (at temperature of 100, 120, 140 and 180 °C) with tofu residue may have some inhibition for biogas production. But, NH₄-N content of RS-160+TR experimental group was lower than inhibition content of 1700 mg/L. It was suggested that the anaerobic co-digestion of RS-160+TR was not inhibited for biogas production during anaerobic fermentation. This result agreed with Antwi E. (2019) ^[2], who has carried out anaerobic digestion and found the possibility of increasing the biogas yield by hydrothermal pretreatment at 150 °C, 30 min.

Fig. 9 showed the NH₄-N contents of fermentation liquid after anaerobic co-digestion of rice straw and HTP tofu residue.

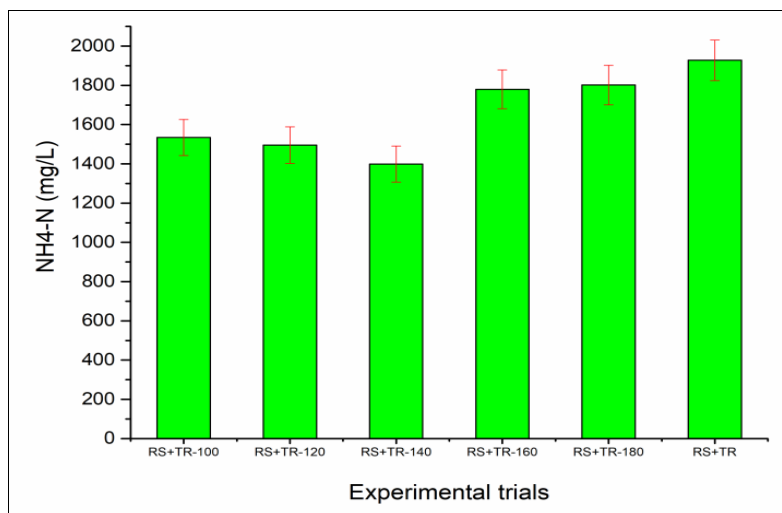


Fig 9: NH₄-N contents of fermentation liquid after anaerobic co-digestion of rice straw and HTP tofu residue. Data are means of two replicates.

As shown Fig. 9, the $\text{NH}_4\text{-N}$ contents of fermentation liquid after anaerobic co-digestion of rice straw and HTP tofu residue were ranged to 1534.7, 1495.4, 1399.4, 1779.5 and 1801.7 mg/L which was measured with RS+TR-100, RS+TR-120, RS+TR-140, RS+TR-160 and RS+TR-180, respectively. It was found that the highest $\text{NH}_4\text{-N}$ content was observed with RS+TR-180, while the lowest $\text{NH}_4\text{-N}$ content was RS+TR-140, 1399.4 mg/L. In the case of RS+TR-100, RS+TR-120, RS+TR-140 experimental groups, the $\text{NH}_4\text{-N}$ content was lower than the inhibition threshold of 1700 mg/L (Bujoczek G., 2000) [6]. When increasing hydrothermal pretreatment temperature of tofu residue, the $\text{NH}_4\text{-N}$ content was increased more than 1700 mg/L. It was suggested that the anaerobic co-digestion of rice straw with HTP tofu residue at temperature between 100 and 140 °C could not inhibit during biogas production, while RS+TR-160 and RS+TR-180 experimental groups had inhibited by $\text{NH}_4\text{-N}$ content higher than inhibition threshold (1700 mg/L). This result is also similar to the one of Antwi E. (2019) [12], which has carried out anaerobic digestion and found the possibility of increasing the biogas yield by hydrothermal pretreatment at 150 °C, 30 min. Hashemi *et al.* (2019) [15] recently have reported that hydrothermal pretreatment of safflower straw at 120 °C for

1h of treatment condition promoted biogas production yield during anaerobic digestion. They found that hydrothermal pretreatment resulted in a solid (mainly cellulose) and a liquid (mainly hemicellulosic monomers) and the highest methane yield from solid fraction (191.4 NmL/g VS) was obtained at the least severe pretreatment conditions (120 °C for 1 h), which resulted in 8.3% improvement in comparison to the control.

3.3.4 Variation of VFAs (Volatile fatty acids) in fermentation liquid after anaerobic co-digestion

The different organic acids content in the digested liquid after anaerobic co-digestion of HTP rice straw with tofu residue and rice straw with HTP tofu residue are shown in Table 4. As shown in Table. 4, all digested liquid contained acetic acid and valeric acid, which was ranged from 3.42 to 23.32%, from 32.01 to 48.03%, respectively. Formic acid was noticed with only RS-180+TR and RS+TR-100 experimental groups, while their values were 0.21 and 0.95%, respectively. Lactic acid contents were measured with RS-100+TR, RS-140+TR, RS+TR-180 and RS+TR (control), while their values were 1.48, 4.11, 1.06 and 8.31%, respectively. Then, butyric acid content was different from 0.42 to 0.78%, respectively.

Table 4: VFAs content in fermentation liquid after anaerobic co-digestion of HTP rice straw with tofu residue and rice straw with HTP tofu residue

Digested material	Formic acid (%)	Acetic acid (%)	Valeric acid (%)	Lactic acid (%)	Propionic acid (%)	Butyric acid (%)
RS-100+TR	-	16.07±0.42	34.43±1.58	1.48±0.17	6.28±1.32	0.61±0.12
RS-120+TR	-	11.8±0.24	32.01±1.63	-	4.73±1.18	-
RS-140+TR	-	9.68±0.71	39.6±2.12	4.11±0.69	2.15±0.31	-
RS-160+TR	-	3.42±0.84	33.07±3.31	-	1.73±0.47	-
RS-180+TR	0.21±0.07	21±1.67	38.51±1.95	-	3.45±1.26	0.42±0.17
RS+TR-100	0.95±0.18	11.02±0.84	39.39±1.82	-	3.14±0.84	0.62±0.18
RS+TR-120	-	17±0.97	32.01±3.26	-	2.95±0.78	-
RS+TR-140	-	17.65±0.95	46.46±2.37	-	1.39±0.33	-
RS+TR-160	-	23.32±1.37	48.03±2.48	-	4.48±1.41	0.53±0.12
RS+TR-180	-	20.63±1.28	43.94±2.31	1.06±0.26	5.39±1.52	0.78±0.15
RS+TR	-	13.81±0.98	34.99±2.19	8.31±0.86	6.08±1.67	0.64±0.14

Not detected. Data are mean values ± standard error of three replicates.

It was found that acetic acid content were very low in digested liquid of anaerobic co-digestion of RS-160+TR and RS+TR-120 experimental groups. It was quite agreed with the recent research of Capson-Tojo *et al.* (2018) [8] who reported that formic acid, lactic acid, and butyric acid were firstly hydrolyzed by hydrolysis and methanogenesis microorganisms for biogas production, therefore methanogenic microorganism preferentially could digest the formic acid and acetic acid to produce methane, and consume less lactic acid.

As shown in Table 4, all experimental groups were differently contained propionic acid, which was ranged from 1.39 to 6.28%. Zhang *et al.* (2001) [32] already suggested that acetic acid could be converted and utilized by methanogenic microorganisms, while propionic acid could not be hydrolyzed by the methanogenic bacteria directly, resulting in the accumulation of propionic acid in the digested liquid. Recently, another research (Capson-Tojo *et al.* 2018) [8] have reported that the high concentration of propionic acid in the digested liquid of food waste led to the inhibition of anaerobic fermentation so it caused decreasing biogas production. In the case of RS-160+TR and RS+TR-140

experimental groups, the propionic acid content was low around 1.73 and 1.39% in the fermentation liquid was the lowest two experimental groups. Therefore, it was suggested that the two RS-160+TR and RS+TR-140 experimental groups could not be inhibited by propionic acid, and the cumulative biogas yield was higher than the other experimental groups.

4. Conclusions

This study found that rice straw of the hydrothermal pretreatment temperature of 160°C in the range of tested HTP temperature has promoted the highest amount of biogas yield (267 mL biogas/g VS) and methane (174.6 mL CH_4 /g VS) production yield during anaerobic co-digestion with tofu residue.

The highest biogas yield (306 mL biogas/g VS), methane production yield (200.1 mL CH_4 /g VS) were obtained at HTP temperature of 140 °C for tofu residue with rice straw for anaerobic co-fermentation. Therefore, HTP of 160 °C for rice straw and 140 °C for tofu residue can be used by anaerobic co-digestion for renewable bio-energy production effectively.

Acknowledgments

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