

# High-Solids Anaerobic Digestion Followed by Ultrasonication of Digestate and Wet-Type Anaerobic Digestion for Enhancing Methane Yield from OFMSW

## Authors:

Bappi Chowdhury, Sarmad Bilal Magsi, Hok Nam Joey Ting, Bipro Ranjan Dhar

Date Submitted: 2020-07-17

Keywords: digestate, ultrasonication, wet-type anaerobic digestion, organic fraction of municipal solid waste (OFMSW), high-solids anaerobic digestion (HSAD)

## Abstract:

High-solids anaerobic digestion of organic fraction of municipal solid waste often shows inefficient biomethane recovery due to mass transfer limitations. Consequently, this study presents a two-stage anaerobic digestion process combining high-solids anaerobic digestion followed by ultrasonication of digestate and wet-type anaerobic digestion for effective biomethane recovery from the organic fraction of municipal solid waste. The high-solids anaerobic digestion yielded methane production of 210 L CH<sub>4</sub>/kg volatile solids (VS). The digestate from the high-solids anaerobic digestion process was ultrasonicated at three different specific energy inputs (1000, 2500, and 5000 kJ/kg total solids (TS)). The increases in the soluble chemical oxygen demand (SCOD) concentrations (8%–32%) and volatile solids (VS) removal efficiencies (3.5%–10%) at different specific energy inputs were linearly correlated ( $R^2 = 0.9356$ ). Thus, ultrasonication led to the solubilization of particulate organics and released soluble organic matters. All ultrasonicated digestate samples showed significantly higher biomethane yields than that observed for the untreated digestate samples. The highest methane yield of 132 L CH<sub>4</sub>/kg VS was observed for a specific energy input of 5000 kJ/kg TS, which was 1.94 times higher than the control (68 L CH<sub>4</sub>/kg VS). Although specific energy inputs of 1000 kJ/kg TS and 2500 kJ/kg TS showed comparable methane yields (113–114 L CH<sub>4</sub>/kg VS), they were ~1.67 times higher than the control. Overall, our results suggest that an integrated system of high-solids and wet-type anaerobic digestion with pre-ultrasonication of digestate has the potential to provide a technically viable solution to enhance biomethane recovery from the organic fraction of municipal solid waste.

Record Type: Published Article

Submitted To: LAPSE (Living Archive for Process Systems Engineering)

Citation (overall record, always the latest version):

LAPSE:2020.0824

Citation (this specific file, latest version):

LAPSE:2020.0824-1

Citation (this specific file, this version):

LAPSE:2020.0824-1v1

DOI of Published Version: <https://doi.org/10.3390/pr8050555>

License: Creative Commons Attribution 4.0 International (CC BY 4.0)

Article

# High-Solids Anaerobic Digestion Followed by Ultrasonication of Digestate and Wet-Type Anaerobic Digestion for Enhancing Methane Yield from OFMSW

Bappi Chowdhury, Sarmad Bilal Magsi, Hok Nam Joey Ting and Bipro Ranjan Dhar \*

Civil and Environmental Engineering, University of Alberta, Edmonton, AB T6G 1H9, Canada; bappi@ualberta.ca (B.C.); smagsi@ualberta.ca (S.B.M.); hoknamjo@ualberta.ca (H.N.J.T.)

\* Correspondence: bipro@ualberta.ca

Received: 10 April 2020; Accepted: 6 May 2020; Published: 9 May 2020



**Abstract:** High-solids anaerobic digestion of organic fraction of municipal solid waste often shows inefficient biomethane recovery due to mass transfer limitations. Consequently, this study presents a two-stage anaerobic digestion process combining high-solids anaerobic digestion followed by ultrasonication of digestate and wet-type anaerobic digestion for effective biomethane recovery from the organic fraction of municipal solid waste. The high-solids anaerobic digestion yielded methane production of 210 L CH<sub>4</sub>/kg volatile solids (VS). The digestate from the high-solids anaerobic digestion process was ultrasonicated at three different specific energy inputs (1000, 2500, and 5000 kJ/kg total solids (TS)). The increases in the soluble chemical oxygen demand (SCOD) concentrations (8%–32%) and volatile solids (VS) removal efficiencies (3.5%–10%) at different specific energy inputs were linearly correlated ( $R^2 = 0.9356$ ). Thus, ultrasonication led to the solubilization of particulate organics and released soluble organic matters. All ultrasonicated digestate samples showed significantly higher biomethane yields than that observed for the untreated digestate samples. The highest methane yield of 132 L CH<sub>4</sub>/kg VS was observed for a specific energy input of 5000 kJ/kg TS, which was 1.94 times higher than the control (68 L CH<sub>4</sub>/kg VS). Although specific energy inputs of 1000 kJ/kg TS and 2500 kJ/kg TS showed comparable methane yields (113–114 L CH<sub>4</sub>/kg VS), they were ~1.67 times higher than the control. Overall, our results suggest that an integrated system of high-solids and wet-type anaerobic digestion with pre-ultrasonication of digestate has the potential to provide a technically viable solution to enhance biomethane recovery from the organic fraction of municipal solid waste.

**Keywords:** high-solids anaerobic digestion (HSAD); organic fraction of municipal solid waste (OFMSW); wet-type anaerobic digestion; ultrasonication; digestate

## 1. Introduction

The rapid urbanization and increased population levels have considerably accelerated the global municipal solid waste (MSW) generation. According to a report published by the Organisation for Economic Co-operation and Development (OECD), the global MSW generation is expected to be increased from 1.3 million tons to 2.2 billion tons over the period 2015–2025 [1]. Currently, only a small fraction of MSW is recycled or converted to value-added resources, with the rest being burned or landfilled due to a lack of suitable technologies and economic constraints [2]. The landfilling of MSW nowadays is under a challenge of requisite space due to the decreasing land availability [3]. Moreover, the landfilling of organic fraction of municipal solid waste (OFMSW), which was estimated to be ~46% of the global MSW [4], is criticized because of its high environmental impacts.

The OFMSW is a heterogeneous mixture of food scraps, paper, yard waste, wood, and other organic wastes. Due to a high proportion of biodegradable materials, many cities in developed and developing countries are now in the process of implementing biological treatment processes, such as anaerobic digestion and composting for the management of OFMSW [5,6]. Particularly, anaerobic digestion (AD) of OFMSW for renewable biomethane recovery is being increasingly adopted by waste management facilities throughout the world [7,8]. AD represents a biological decomposition of organic compounds to biomethane by the action of groups of hydrolytic/fermentative bacteria and methanogenic archaea. However, based on the solids content, the process can be classified into two major categories, wet-type or low-solids anaerobic digestion (LSAD) and dry-type or high-solids anaerobic digestion (HSAD) [9]. However, LSAD (<15% total solids, TS) is not suitable for many waste streams with a high solids content, such as lignocellulosic and yard wastes [7,10]. In contrast, HSAD (>15% TS) systems are more flexible in handling organic wastes with high solids content [8,10]. Therefore, OFMSW, which is composed of diverse wastes with high solids content, is primarily treated with the HSAD process [9,11,12].

Compared to the wet-type digesters, high-solids digesters can provide several benefits, including smaller digester footprint, lower heating cost, and higher volumetric methane productivities [9]. Despite these benefits, various operational challenges still limit HSAD systems from being widely applied. The performance of HSAD systems depends on the operating TS content and biodegradability of feedstocks [9]. Notably, the presence of lignocellulosic materials (e.g., yard waste and paper) in OFMSW may lead to slow process kinetics due to the low biodegradability of these substrates [9]. Additionally, mass transfer limitations associated with the operation without any mixing poses challenges in achieving effective solids removal and biomethane recovery from the HSAD process [13].

Additionally, HSAD systems often show process instability because of localized inhibition of microbial communities by high ammonia levels and the accumulation of various volatile fatty acids (VFAs) [13,14]. For instance, Brown et al. [15] reported that the LSAD could provide higher methane yield (312.4 L CH<sub>4</sub>/kg volatile solids (VS)) from waste paper than that could be achieved with the HSAD process (15 L CH<sub>4</sub>/kg VS). Thus, a significant portion of biodegradable organics would be remaining in the digestate from HSAD systems. Currently, digestate from HSAD systems are most often composted or directly used for land application [6,16], while efforts have been limited in developing downstream processes to promote further biotransformation of residual organics to bioenergy.

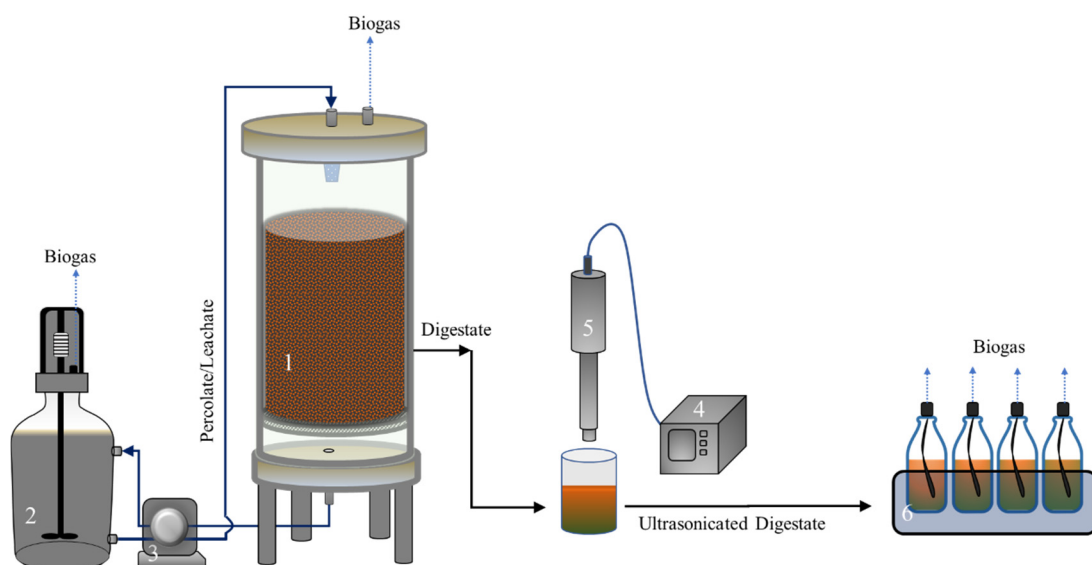
In general, the biodegradability of the digestate from anaerobic digesters is very low [17]. However, a few studies suggested that digestate from one digester can be hydrolyzed and subsequently used as a feedstock for a second digester or can be recycled as an additional feedstock to an existing digester [17–20]. For instance, Cesaro et al. [20] investigated the ultrasonication of lignocellulosic and protein-rich digestate. Their results demonstrated that ultrasonication of digestate with lignocellulosic-based materials could enhance biogas production by up to 71%. In contrast, ultrasonication of protein-rich digestate showed a relatively lower increase (3%–23%) in methane production. Somers et al. [19] studied three different hydrolysis methods (ultrasonication, hydrogen peroxidation, and ozone treatment) on manure, potato waste, and mixed organic waste digestate. Their results suggested that ultrasonication (15,000 kJ/kg TS) of potato waste digestate could increase biogas yield up to 22.5% in a second-stage digester. Boni et al. [18] investigated that ultrasonication (6000 kJ/kg TS) of food waste digestate could increase biomethane production by up to 30%. Previous studies [7–21] suggested that ultrasonication could enhance anaerobic biodegradability due to the physical disintegration of particulate organics via acoustic cavitation phenomena along with the enhancement of enzymatic activity. Although ultrasonication seems to be a highly promising approach for hydrolysis of digestate to enhance additional biomethane recovery, it is expected that the characteristics of feedstocks would significantly influence the optimum operating conditions for ultrasound applications [21]. To date, very limited information is available on the ability of ultrasonication to promote further methane recovery from the digestate from HSAD systems operated with OFMSW.

The primary goal of this study was to investigate an integrated process of HSAD followed by ultrasonication of digestate and wet-type anaerobic digestion for enhanced biomethane recovery from OFMSW. First, we operated a lab-scale HSAD system with OFMSW collected from a full-scale waste management facility. Second, ultrasonication was applied at different specific energy inputs for the hydrolysis of the digestate from the HSAD system. Third, methane potentials of ultrasonicated digestate samples were evaluated with biochemical methane potential (BMP) tests. The results of this study would be useful in engineering strategies to enhance methane yield from OFMSW.

## 2. Materials and Method

### 2.1. High-Solids Anaerobic Digestion of OFMSW

We used a lab-scale HSAD system that consisted of a digester tank (working volume of 15 L) connected to a leachate/percolate storage tank (working volume of 2 L) (Figure 1). A stainless-steel mesh was placed at the bottom of the digester tank to allow the leachate to pass through. The leachate storage tank was equipped with an agitator for mixing of leachate. The gas outlets from both tanks were connected to an individual absorption trap for capturing acidic gases (e.g., CO<sub>2</sub>, H<sub>2</sub>S, etc.) from biogas with 3 M NaOH solution having thymolphthalein pH-indicator, which allowed direct measurement of methane volume with wet-tip gas meters [22].



**Figure 1.** Schematic diagram of experimental setup. Notes: (1) digester tank; (2) percolate/leachate tank; (3) pump; (4) ultrasound generator; (5) ultrasonic probe; (6) batch anaerobic bioreactor system.

The OFMSW, solid, and liquid inoculums were obtained from the Edmonton Waste Management Centre (EWMC) located in Edmonton, AB, Canada. The OFMSW sample contained mainly grass clippings, yard waste, paper, food waste, and some plastics. The total solids (TS) and volatile solids (VS) content in the sample were  $54 \pm 5\%$  and  $26 \pm 3.4\%$ , respectively. The dewatered biosolids (TS:  $24.63 \pm 0.10\%$ , VS:  $14.45 \pm 0.002\%$ ) were used as the solid inoculum. The characteristics of liquid inoculum are as follows: chemical oxygen demand (COD):  $16.04 \pm 0.05$  g/L, total ammonia nitrogen (TAN):  $3.60 \pm 0.15$  g/L, alkalinity:  $18.43 \pm 0.36$  g/L, pH:  $8.40 \pm 0.00$ . During start-up, the digester tank was loaded with 1.9 kg of OFMSW and 2.1 kg of solid inoculum. They were premixed before loading in the digester tank. The leachate tank was loaded with 2 L of liquid inoculum. The leachate was circulated between the digester tank and leachate storage tank using a peristaltic pump (Longer Pump BT100-2J, Langer Instruments Corp, Tucson, AZ, USA). For the first four days, the flow rate was set at 2 mL/s for 10 min per day and then increased to 4 h per day until the end of the experiment. However, a considerable decrease in the leachate volume was observed over time due to the evaporation and

accumulation of liquid in the digester tank. The temperature of both tanks was maintained at  $37 \pm 2$  °C with electrical heating tapes. The liquid in the leachate storage tank was continuously stirred with a mechanical agitator connected with electric motor (ISES-Canada, Vaughan, ON, Canada) for continuous mixing at 300 rpm during the experiment. The volumes of methane gas produced from both tanks were monitored on a regular basis.

## 2.2. Ultrasound Treatment and BMP Test of HSAD Digestate

The ultrasound treatment of HSAD digestate was performed with a bench-scale ultrasound generator (Model Q500, QSonica LLC, Newtown, CT, USA) equipped with an ultrasonic probe (see Figure 1). The amplitude of the device was set at 120  $\mu\text{m}$ , and sonication pulses were set to 5 s on and 5 s off to avoid excessive heat generation. The digestate sample was pretreated at three different specific energy inputs (1000, 2500, and 5000 kJ/kg TS), which corresponded to sonication times of 12, 32, and 65 min, respectively. The digestate sample was diluted by ~40% with deionized water for the proper dispersion of sonication waves. Then, 250 mL of sample was taken in a beaker, and the probe of the device was immersed into the sample; immersion depth was fixed at 2.5 cm. It should be noted that we applied ultrasonication at the specific energy input of up to 5000 kJ/kg TS, because a previous study suggested that specific energy input  $\geq 5000$  kJ/kg TS may not be economically feasible [23].

The sonicated samples were then used for the BMP test with a batch anaerobic bioreactor system (ISES-Canada, Vaughan, ON, Canada) (Figure 1). The system consisted of glass anaerobic bioreactors with a working volume of 600 mL. These reactors were equipped with agitators for mixing liquids. Each reactor was connected with an individual absorption trap for capturing acidic gases from biogas, as described above. The tests were conducted for three different conditions: control (untreated digestate + inoculum), ultrasonication (sonicated digestate + inoculum), and blank (deionized water + inoculum). We operated triplicate bioreactors for each test condition. The inoculum (anaerobic digester sludge) was collected from the Gold Bar wastewater treatment plant located in Edmonton, Alberta, Canada. The average characteristics of inoculum are as follows: TCOD:  $28,080 \pm 47$  mg/L; TS:  $25,085 \pm 232$  mg/L; VS:  $15,179 \pm 58$  mg/L; pH: 6.8. Prior to the BMP test, the inoculum was acclimatized at 37 °C for 5 days. The volumes of substrate and inoculum were estimated based on food to microorganism ratio (F/M) of 2 (g of VS of sample/g of VS of inoculum). The initial pH of the mixture (sample + inoculum) was measured in the range of 7.5–8.0. However, sodium bicarbonate (6 g/L) was added as an additional buffer to avoid any pH drop during batch operation. All bioreactors were purged with ultrapure nitrogen gas for 2 min to create an anaerobic environment. Then, the bioreactors were placed in a water bath, and the temperature was maintained at  $37 \pm 2$  °C. Additionally, the mixing was set at 300 rpm. Methane produced from each reactor was collected with a gas bag connected to the absorption trap for acidic gases, and volume was measured on a regular basis with a frictionless glass syringe.

## 2.3. Analytical Method

The COD and TAN concentrations were measured with HACH reagent kits (HACH, Loveland, CO, USA). For the determination of soluble chemical oxygen demand (SCOD), the samples were filtered through a 0.45  $\mu\text{m}$  membrane. The pH values were measured using a bench-top pH meter (AR15 pH meter, Fisher Scientific, Pittsburgh, PA, USA). The concentrations of various volatile fatty acids (acetate, propionate, and butyrate) were measured using ion chromatography (Dionex ICS-2100, Dionex, Sunnyvale, CA, USA), which was equipped with an electrochemical detector (ECD) and a microbore AS19 column. The TS and VS concentrations were measured according to the standard method (APHA, 1999).

## 2.4. Kinetic and Statistical Analyses

The modified Gompertz model was used to predict kinetic parameters as described in Equation (1) [24]:

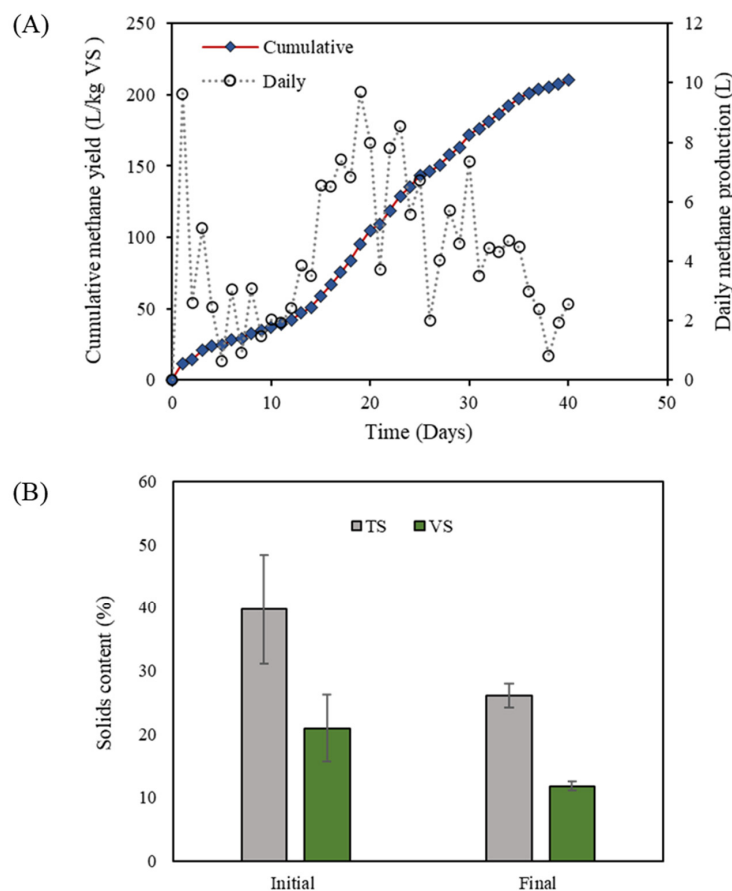
$$H(t) = H_{\max} \times \exp\left\{-\exp\left[\frac{R_{\max} \times e}{H_{\max}} (\lambda - t) + 1\right]\right\} \quad (1)$$

where  $H(t)$  is the cumulative methane production (L CH<sub>4</sub>/kg VS) at time  $t$ ;  $H_{\max}$  is the methane potential (L CH<sub>4</sub>/kg VS);  $R_{\max}$  is the maximum methane production rate (L CH<sub>4</sub>/kg VS-d);  $\lambda$  is the time of lag phase (d). The cumulative methane from data from BMP tests were fitted with Equation (1) for estimation of kinetic parameters, including methane production rate (L CH<sub>4</sub>/kg VS-d) and lag phase (d). Statistical significance was tested using analysis of variance (ANOVA) and student's  $t$ -test in R project (v.3.5.1) with a threshold  $p$ -value of 0.05.

### 3. Results and Discussion

#### 3.1. Performance of HSAD

Figure 2A shows the cumulative methane yield and daily methane production from the HSAD operation. The system was operated for 40 days until the methane production was negligible. Although no lag phase was observed, daily methane production sharply decreased after day 1 and remained low until day 14. The maximum daily methane production during this period was 5.1 L on day 3. The exponential phase of methane production commenced on day 15 and showed a maximum daily methane production of 9.7 L on day 19. Such methane generation pattern is quite typical for batch high-solids anaerobic digestion due to an early hydrolysis/fermentation phase followed by the methanogenic phase [8,10]. From day 20, methane gas production started to decline with some fluctuations. A minimum methane production of 0.83 L was observed on day 38. After 40 days of operation, the total methane production was 176.8 L, resulting in a specific methane yield of 210 L CH<sub>4</sub>/kg VS. This methane yield was within the range of values (109–211 L CH<sub>4</sub>/kg VS) previously reported for high-solids anaerobic digestion of OFMSW [8,10,25].



**Figure 2.** (A) Cumulative methane and daily methane production from the high-solids anaerobic digestion (HSAD) reactor; (B) initial and final solids contents.

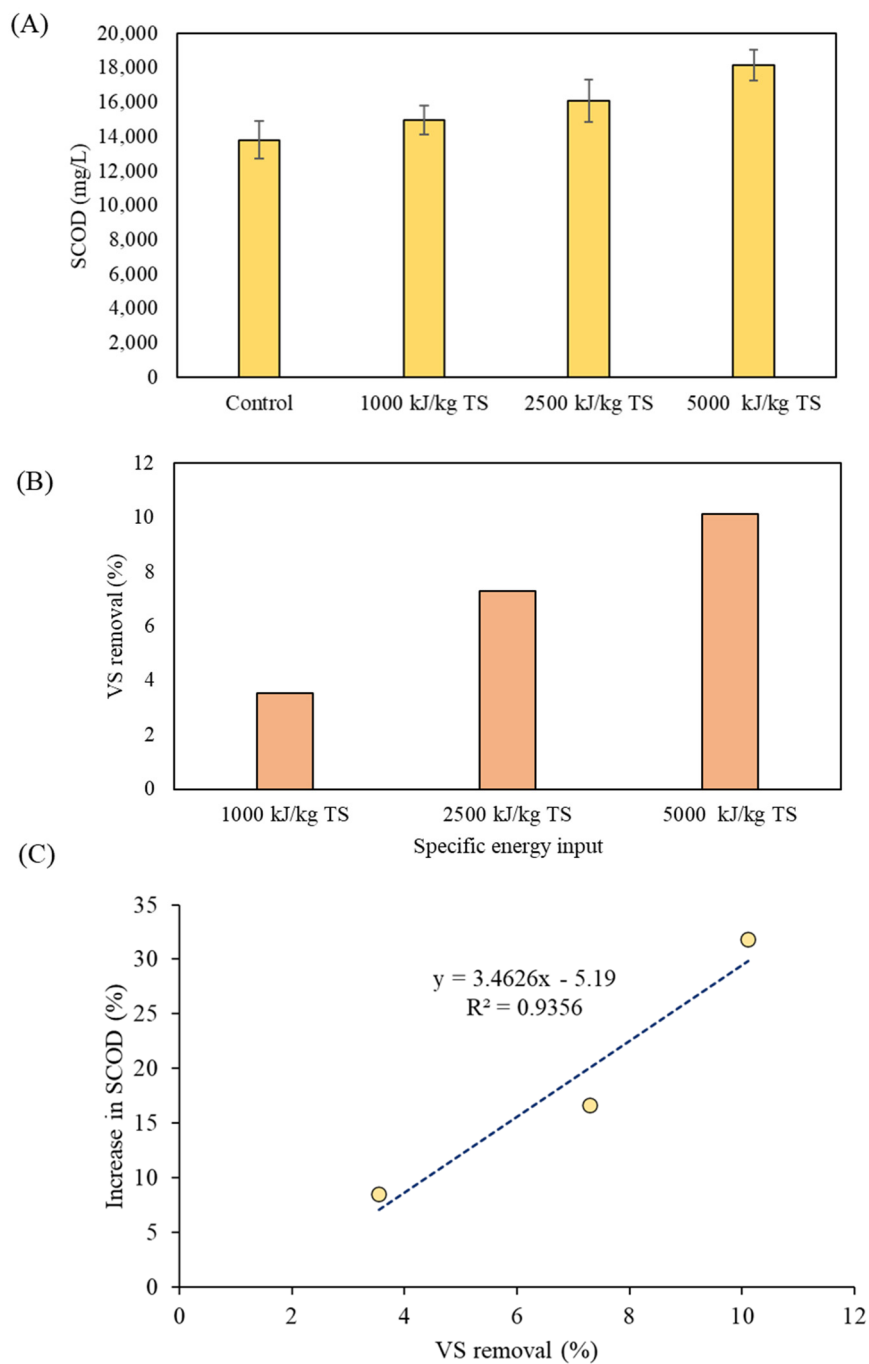
Figure 2B shows TS and VS contents for an initial mixture of OFMSW plus biosolids and final digestate. Initial TS and VS contents were  $39.70 \pm 8.60\%$  and  $20.91 \pm 5.31\%$ , respectively. In the final digestate, solids content decreased to  $26.04 \pm 1.90\%$  (TS) and  $11.77 \pm 0.74\%$  (VS), which led to the removal efficiency of 34% (TS) and 44% (VS), respectively. The VS removal efficiency was slightly higher than 38% VS removal efficiency recently reported for an OFMSW fed HSAD system with percolate recirculation [10].

### 3.2. Effects of Ultrasonication of Digestate

The changes in SCOD, VS, and VFA concentrations were considered as an indicator of the effectiveness of ultrasonication in terms of particulate organics solubilization [26], which can potentially benefit biomethane recovery. These parameters were measured before and after ultrasound treatment. Figure 3A shows the changes in SCOD concentrations in digestate samples due to ultrasonication. Ultrasound treatment at different specific energy inputs significantly increased SCOD concentrations, which could be attributed to the solubilization of particulate organics in the digestate [21]. Notably, ultrasound treatment at 5000 kJ/kg TS had a significant impact on organics solubilization as reflected by the maximum increase (32%) in SCOD concentration from  $13,789 \pm 1094$  mg/L (untreated control sample) to  $18,171 \pm 1100$  mg/L (ultrasonicated sample). In contrast, SCOD increased only by 8% for a specific energy input of 1000 kJ/kg TS. Nonetheless, under the investigated conditions, SCOD concentrations increased linearly with an increase in ultrasound specific energy inputs. Previous studies also reported that ultrasound treatment <10,000 kJ/kg TS could linearly increase COD solubilization for different feedstocks, including sewage sludge, lignocellulosic waste, manure, etc. [20,27–29]. Additionally, SCOD solubilization efficiencies achieved in this study (0.9–1.2 mg SCOD/(kJ/kg TS)) were within the range of values (0.12–1.35 mg SCOD/(kJ/kg TS)) previously reported in the literature [21,30]. Interestingly, the specific energy input of 1000 kJ/kg TS provided highest SCOD solubilization efficiency of 1.2 mg SCOD/(kJ/kg TS) in this study, which may have been caused by the fact that higher ultrasound specific energy input can lead to substantial energy loss in the form of heat [31].

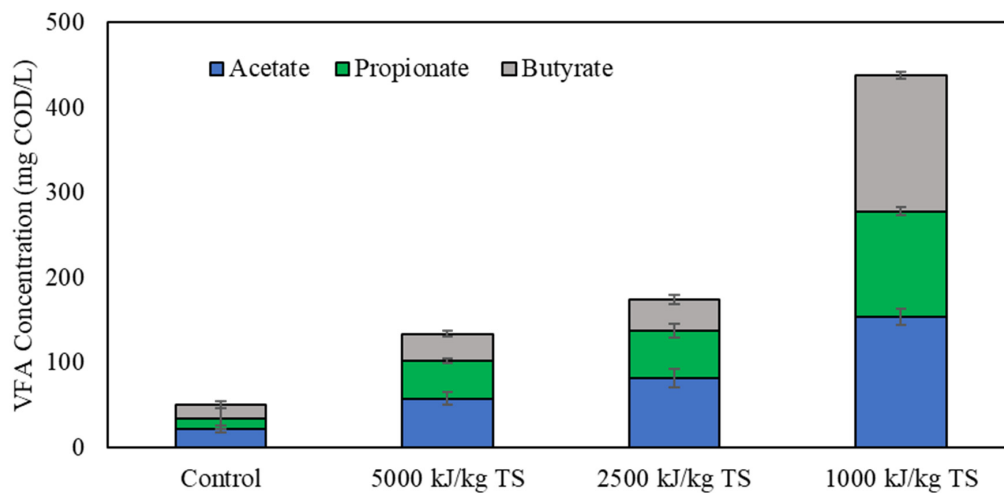
A similar trend was observed in terms of VS removal efficiencies (see Figure 3B); VS removal efficiency increased with the increase in specific energy inputs. VS removal efficiencies were ranged from 3.5% (1000 kJ/kg TS) to 10% (5000 kJ/kg TS). The VS removal efficiencies for treated samples were linearly correlated with an increase in SCOD concentrations ( $R^2 = 0.9356$ ) (see Figure 3C). Thus, these results suggested that an increase in SCOD might have resulted from the solubilization of particulate organic matters during ultrasound treatment. Several studies previously reported a similar trend of linearity between SCOD and VS concentrations for ultrasound treatment at different specific energy inputs [7,32,33]. Conversely, some studies reported a non-linear relationship between the degree of COD solubilization and VS removal efficiencies [20,34].

As shown in Figure 4, concentrations of various VFAs increased considerably after ultrasonication. TVFA concentration increased by 8.75 times after ultrasonication at 5000 kJ/kg TS, as compared to the control. At specific energy inputs of 1000 kJ/kg TS and 2500 kJ/kg TS, TVFA concentrations increased by 2.68 and 3.48 times, respectively. Although SCOD and VFAs concentrations increased in all ultrasonicated samples, TVFA/SCOD ratios remained very low (0.48–0.15) in those samples. Thus, ultrasonication was effective in the hydrolysis of other particulate organic matters, while it needs a further metabolic step to convert soluble organics into VFAs. Previous studies also suggested that various pre-treatments methods used for organic feedstocks could increase VFAs concentration slightly due to the disintegration of particulate organics [18,35,36].



**Figure 3.** (A) Changes in soluble chemical oxygen demand (SCOD) concentrations; (B) volatile solids (VS) removal efficiencies; and (C) relationship between increase in SCOD concentrations and VS removal efficiencies for different ultrasonication conditions.



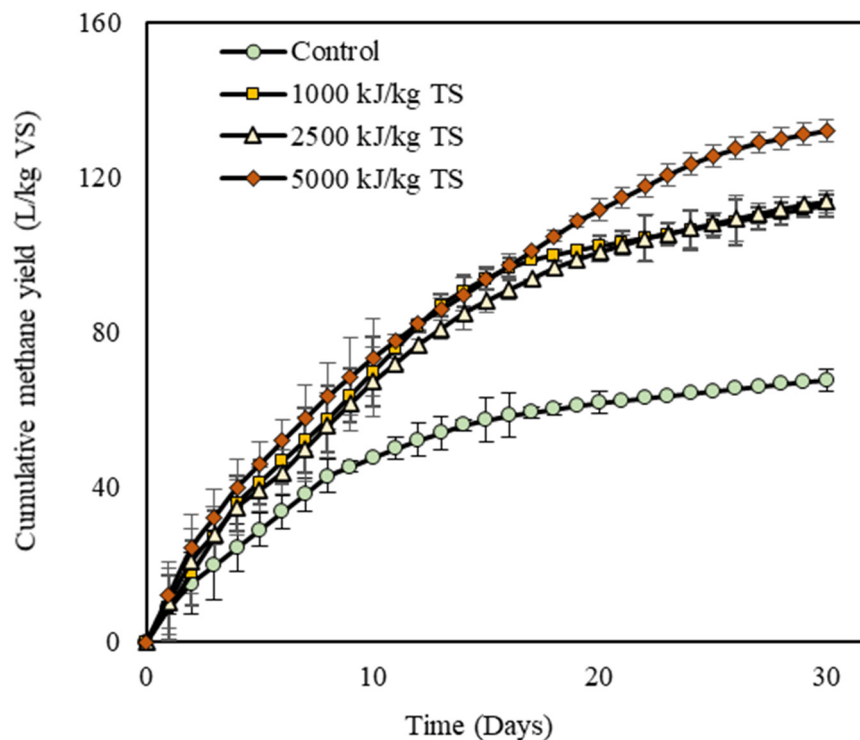


**Figure 4.** Changes in volatile fatty acids (VFAs) concentrations for different ultrasonication conditions.

### 3.3. Biomethane Recovery from Digestate

Figure 5 shows the cumulative methane yields from three ultrasonicated and one untreated HSAD digestate samples. None of the samples showed any lag phases. For the untreated control sample, the ultimate methane yield reached a plateau after 15 days. In contrast, for samples sonicated at 1000 and 5000 kJ/kg TS, ultimate methane yields reached a plateau after 18 days, whereas it took nearly 28 days for the sample sonicated at 5000 kJ/kg TS. As apparent in Figure 5, the ultimate methane yields of the ultrasonicated samples were much higher than the untreated control sample (113–132 vs. 68 L CH<sub>4</sub>/kg VS). Thus, the solubilization of particulate organics and the release of soluble organics (i.e., increase in SCOD) positively affected methane productivity. Of the three ultrasonicated samples, the sample pretreated at 5000 kJ/kg TS showed the highest methane yield of 132 L CH<sub>4</sub>/kg VS, which was 90% higher than the untreated control. However, ultimate methane yields from samples sonicated at 1000 and 2500 kJ/kg TS were comparable ( $p > 0.05$ ). Previous studies also reported that the increase in methane production did not correlate linearly with various specific energy inputs in ultrasound treatment of different feedstock [21,23]. For instance, Elbeshbishy et al. [21] observed an increase in methane production for a specific energy input of 500 kJ/kg TS, while methane production decreased for specific energy inputs of 500–10,000 kJ/kg TS before increasing again. Nonetheless, our results suggest that the operation of HSAD for 40 days could partially convert organics to biomethane. Thus, a considerable portion of particulate biodegradable organics would remain in the HSAD digestate. The application of ultrasound followed by wet-type anaerobic digestion as a post-treatment could provide an excellent opportunity to further effective methane recovery from residual organics in HSAD digestate. Additionally, 5000 kJ/kg TS was the most effective in enhancing methane yield.

Table 1 summarizes the kinetic parameters estimated by fitting cumulative methane yield data. The values of methane potentials, maximum methane production rates, and lag phases for samples treated at 1000 and 2500 kJ/kg TS were comparable ( $p > 0.05$ ). The sample treated at 5000 kJ/kg TS showed a maximum methane production rate of  $6 \pm 1$  L CH<sub>4</sub>/kg VS/d, which was slightly higher than the other ultrasonicated samples. The lowest methane production rate of  $1.5 \pm 0.4$  L CH<sub>4</sub>/kg VS/d was estimated for the untreated control sample. Thus, ultrasonication could increase the methane production rate during the second-stage digestion of the HSAD digestate.



**Figure 5.** Cumulative methane yields from digestate ultrasonicated under different specific energy inputs.

**Table 1.** Kinetic parameters estimated with biochemical methane potential (BMP) test data fitting. TS: total solids.

Condition	Modified Gompertz Model			
	CH <sub>4</sub> Potential (L CH <sub>4</sub> /kg VS)	Maximum CH <sub>4</sub> Production Rate (L CH <sub>4</sub> /kg VS/d)	Lag Phase (d)	R <sup>2</sup>
Control	74 ± 7	1.5 ± 0.4	2 ± 1	0.9734
1000 kJ/kg TS	127 ± 11	4.7 ± 1	1 ± 1	0.9822
2500 kJ/kg TS	132 ± 8	4.8 ± 0.8	1 ± 1	0.9945
5000 kJ/kg TS	158 ± 15	6 ± 1	0	0.9832

At the end of the BMP test, TAN and VFAs concentrations were measured in the final digestate, since they may provide valuable information on methanogenesis efficiencies and the possibility of inhibition (Table 2). Before starting the BMP test, TAN concentrations in all bottles were ranged from 1598–1756 mg N/L (data not shown). After the completion of the BMP test, TAN concentrations increased in the final digestate samples (see Table 2), which could be contributed to the hydrolysis of proteins [8]. The final TAN concentrations in samples sonicated at 2500 and 5000 kJ/kg TS were slightly higher than the sample sonicated at 1000 kJ/kg TS. The final TAN concentration in the digestate from the control ( $1787 \pm 33$  mg N/L) was slightly lower than all ultrasonicated samples (1977–2181 mg N/L). The unionized form of FAN has been suggested to be inhibitory to methanogenesis [37–39]. Therefore, FAN concentrations were estimated for all samples. The estimated FAN concentrations varied between 179 to 210 mg N/L (see Table 2). Based on the previous studies, a threshold of FAN concentrations for introducing inhibition may vary widely from 100 to 1100 mg N/L [40]. Thus, we cannot completely rule out the possibility of FAN inhibition during the BMP test. However, it was evident that the hydrolysis of digestate with ultrasound did not impose any additional risk of ammonia inhibition. Some previous studies reported that high FAN levels might lead to VFAs accumulation in the digester [41]. As shown in Table 2, the control showed the least TVFA concentration followed by sample sonicated at 1000 kJ/kg

TS, 2500 kJ/kg TS, and 5000 kJ/kg TS. However, the total VFAs accumulation in all reactors remained below ~300 mg COD/L. Thus, there was no sign of inhibition.

**Table 2.** Nitrogen and volatile fatty acids concentrations in the final digestate from BMP test. COD: chemical oxygen demand.

	Control	1000 kJ/kg TS	2500 kJ/kg TS	5000 kJ/kg TS
Total ammonia nitrogen (mg N/L)	1787 ± 33	1977 ± 32	2056 ± 54	2181 ± 28
Free ammonia nitrogen (mg N/L)	179 ± 3	210 ± 3	200 ± 3	183 ± 2
Total volatile fatty acids (mg COD/L)	73 ± 6	148 ± 23	158 ± 13	239 ± 13
Acetate (mg COD/L)	39 ± 4	79 ± 26	88 ± 2	124 ± 6
Propionate (mg COD/L)	34 ± 2	28 ± 8	34 ± 7	46 ± 4
Butyrate (mg COD/L)	-	41 ± 9	36 ± 4	69 ± 3

#### 4. Conclusions

This study presents high-solids anaerobic digestion followed by wet type anaerobic digestion of ultrasonicated digestate for enhancing the methane yield from OFMSW. Methane yield obtained from HSAD was 210 L CH<sub>4</sub>/kg VS, while methane yield from the digestate in subsequent wet-type anaerobic digestion was only 68 L CH<sub>4</sub>/kg VS. The ultrasonication of digestate linearly increased SCOD solubilization with an increase in specific energy inputs. Additionally, a linear correlation was observed between SCOD solubilization and VS removal efficiencies. The highest methane yield of 132 L CH<sub>4</sub>/kg VS was observed for a specific energy input of 5000 kJ/kg TS, which was 1.94 times higher than that observed for untreated digestate. Thus, these results suggest that ultrasonication of OFMSW digestate from high-solids anaerobic digestion may provide additional energy recovery options with wet-type anaerobic digestion. However, future studies need to involve comprehensive economic feasibility analyses to investigate the potential for practical application. Moreover, the quality of the final digestate should be assessed for potential environmental risks, such as pathogen content, antibiotic resistance genes, etc.

**Author Contributions:** All authors contributed to the design of experiment. B.C., S.B.M., and H.N.J.T. conducted the experiments and collected data. B.R.D. played an advisory role. All authors contributed to the preparation of the manuscript. All authors have read and agreed to the published version of the manuscript.

**Funding:** The authors would like to thank the financial and in-kind contributions from Alberta Innovates (CCITF Clean Technology Development Program), City of Edmonton, Natural Sciences and Engineering Research Council of Canada (Discovery Grant), Faculty of Engineering at the University of Alberta (Start-up Grant), and Future Energy Systems (Early Career Research Grant). The authors also thank Ibrahim Karidio and Hamid Zaman at the Edmonton Waste Management Centre (City of Edmonton) for their assistance.

**Conflicts of Interest:** The authors declare no conflict of interest.

#### References

1. Kawai, K.; Tasaki, T. Revisiting estimates of municipal solid waste generation per capita and their reliability. *J. Mater. Cycles Waste Manag.* **2016**, *18*, 1–13. [[CrossRef](#)]
2. Kaza, S.; Yao, L.; Bhada-Tata, P.; Woerden, F. *Van What a Waste 2.0 Everything You Should Know About Solid Waste Management*; The World Bank: Washington, DC, USA, 2018; ISBN 978-1-4648-1347-4.
3. Aghbashlo, M.; Tabatabaei, M.; Soltanian, S.; Ghanavati, H. Biopower and biofertilizer production from organic municipal solid waste: An exergoenvironmental analysis. *Renew. Energy* **2019**, *143*, 64–76. [[CrossRef](#)]
4. Lim, S.L.; Lee, L.H.; Wu, T.Y. Sustainability of using composting and vermicomposting technologies for organic solid waste biotransformation: Recent overview, greenhouse gases emissions and economic analysis. *J. Clean. Prod.* **2016**, *111*, 262–278. [[CrossRef](#)]
5. Bilgili, M.S.; Demir, A.; Varank, G. Evaluation and modeling of biochemical methane potential (BMP) of landfilled solid waste: A pilot scale study. *Bioresour. Technol.* **2009**, *100*, 4976–4980. [[CrossRef](#)]

6. Tyagi, V.K.; Fdez-Güelfo, L.A.; Zhou, Y.; Álvarez-Gallego, C.J.; Garcia, L.I.R.; Ng, W.J. Anaerobic co-digestion of organic fraction of municipal solid waste (OFMSW): Progress and challenges. *Renew. Sustain. Energy Rev.* **2018**, *93*, 380–399. [CrossRef]
7. Apul, O.G.; Sanin, F.D. Ultrasonic pretreatment and subsequent anaerobic digestion under different operational conditions. *Bioresour. Technol.* **2010**, *101*, 8984–8992. [CrossRef]
8. Michele, P.; Giuliana, D.; Carlo, M.; Sergio, S.; Fabrizio, A. Optimization of solid state anaerobic digestion of the OFMSW by digestate recirculation: A new approach. *Waste Manag.* **2015**, *35*, 111–118. [CrossRef]
9. Pastor-Poquet, V.; Papirio, S.; Trably, E.; Rintala, J.; Escudié, R.; Esposito, G. High-solids anaerobic digestion requires a trade-off between total solids, inoculum-to-substrate ratio and ammonia inhibition. *Int. J. Environ. Sci. Technol.* **2019**, *16*, 7011–7024. [CrossRef]
10. Lee, E.; Bittencourt, P.; Casimir, L.; Jimenez, E.; Wang, M.; Zhang, Q.; Ergas, S.J. Biogas production from high solids anaerobic co-digestion of food waste, yard waste and waste activated sludge. *Waste Manag.* **2019**, *95*, 432–439. [CrossRef]
11. Li, Y.; Park, S.Y.; Zhu, J. Solid-state anaerobic digestion for methane production from organic waste. *Renew. Sustain. Energy Rev.* **2011**, *15*, 821–826. [CrossRef]
12. Anaerobic Digestion of the Organic Fraction of Municipal Solid Waste in Europe—Status, Experience and Prospects. Available online: [https://www.researchgate.net/publication/284229577\\_Anaerobic\\_digestion\\_of\\_the\\_organic\\_fraction\\_of\\_municipal\\_solid\\_waste\\_in\\_Europe\\_-\\_Status\\_experience\\_and\\_prospects](https://www.researchgate.net/publication/284229577_Anaerobic_digestion_of_the_organic_fraction_of_municipal_solid_waste_in_Europe_-_Status_experience_and_prospects) (accessed on 8 April 2020).
13. Rocamora, I.; Wagland, S.T.; Villa, R.; Simpson, E.W.; Fernández, O.; Bajón-Fernández, Y. Dry anaerobic digestion of organic waste: A review of operational parameters and their impact on process performance. *Bioresour. Technol.* **2020**, *299*, 122681. [CrossRef]
14. Mata-Alvarez, J.; Macé, S.; Llabrés, P. Anaerobic digestion of organic solid wastes. An overview of research achievements and perspectives. *Bioresour. Technol.* **2000**, *74*, 3–16. [CrossRef]
15. Brown, D.; Shi, J.; Li, Y. Comparison of solid-state to liquid anaerobic digestion of lignocellulosic feedstocks for biogas production. *Bioresour. Technol.* **2012**, *124*, 379–386. [CrossRef]
16. Cesaro, A.; Belgiorno, V.; Naddeo, V. Comparative technology assessment of anaerobic digestion of organic fraction of MSW. *WIT Trans. Ecol. Environ.* **2010**, *142*, 355–366.
17. Lindner, J.; Zielonka, S.; Oechsner, H.; Lemmer, A. Effects of mechanical treatment of digestate after anaerobic digestion on the degree of degradation. *Bioresour. Technol.* **2015**, *178*, 194–200. [CrossRef] [PubMed]
18. Boni, M.R.; D’Amato, E.; Poletti, A.; Pomi, R.; Rossi, A. Effect of ultrasonication on anaerobic degradability of solid waste digestate. *Waste Manag.* **2016**, *48*, 209–217. [CrossRef] [PubMed]
19. Somers, M.H.; Azman, S.; Sigurnjak, I.; Ghyselbrecht, K.; Meers, E.; Meesschaert, B.; Appels, L. Effect of digestate disintegration on anaerobic digestion of organic waste. *Bioresour. Technol.* **2018**, *268*, 568–576. [CrossRef]
20. Cesaro, A.; Velten, S.; Belgiorno, V.; Kuchta, K. Enhanced anaerobic digestion by ultrasonic pretreatment of organic residues for energy production. *J. Clean. Prod.* **2014**, *74*, 119–124. [CrossRef]
21. Elbeshbishy, E.; Aldin, S.; Hafez, H.; Nakhla, G.; Ray, M. Impact of ultrasonication of hog manure on anaerobic digestability. *Ultrason. Sonochem.* **2011**, *18*, 164–171. [CrossRef]
22. Chowdhury, B.; Lin, L.; Dhar, B.R.; Islam, M.N.; McCartney, D.; Kumar, A. Enhanced biomethane recovery from fat, oil, and grease through co-digestion with food waste and addition of conductive materials. *Chemosphere* **2019**, *236*, 124362. [CrossRef]
23. Dhar, B.R.; Nakhla, G.; Ray, M.B. Techno-economic evaluation of ultrasound and thermal pretreatments for enhanced anaerobic digestion of municipal waste activated sludge. *Waste Manag.* **2012**, *32*, 542–549. [CrossRef] [PubMed]
24. Das, A.; Mondal, C. Comparative Kinetic Study of Anaerobic Treatment of Thermally Pretreated Source-Sorted Organic Market Refuse. *J. Eng.* **2015**, *2015*, 684749. [CrossRef]
25. Qian, M.; Zhang, Y.; Li, R.; Nelles, M.; Stinner, W.; Li, Y. Effects of Percolate Recirculation on Dry Anaerobic Co-digestion of Organic Fraction of Municipal Solid Waste and Corn Straw. *Energy Fuels* **2017**, *31*, 12183–12191. [CrossRef]
26. Wenjing, L.; Chao, P.; Lama, A.; Xindi, F.; Rong, Y.; Dhar, B.R. Effect of pre-treatments on biological methane potential of dewatered sewage sludge under dry anaerobic digestion. *Ultrason. Sonochem.* **2019**, *52*, 224–231. [CrossRef] [PubMed]

27. Bougrier, C.; Carrère, H.; Delgenès, J.P. Solubilisation of waste-activated sludge by ultrasonic treatment. *Chem. Eng. J.* **2005**, *106*, 163–169. [[CrossRef](#)]
28. Lizama, A.C.; Figueiras, C.C.; Herrera, R.R.; Pedreguera, A.Z.; Ruiz Espinoza, J.E. Effects of ultrasonic pretreatment on the solubilization and kinetic study of biogas production from anaerobic digestion of waste activated sludge. *Int. Biodeterior. Biodegrad.* **2017**, *123*, 1–9. [[CrossRef](#)]
29. Zhang, Z.L.; Zhang, L.; Zhou, Y.L.; Chen, J.C.; Liang, Y.M.; Wei, L. Pilot-scale operation of enhanced anaerobic digestion of nutrient-deficient municipal sludge by ultrasonic pretreatment and co-digestion of kitchen garbage. *J. Environ. Chem. Eng.* **2013**, *1*, 73–78. [[CrossRef](#)]
30. Khanal, S.K.; Grewell, D.; Sung, S.; Van Leeuwen, J. Ultrasound applications in wastewater sludge pretreatment: A review. *Crit. Rev. Environ. Sci. Technol.* **2007**, *37*, 277–313. [[CrossRef](#)]
31. Garoma, T.; Pappaterra, D. An investigation of ultrasound effect on digestate solubilization and methane yield. *Waste Manag.* **2017**, *71*, 728–733. [[CrossRef](#)] [[PubMed](#)]
32. Elbeshbishy, E.; Hafez, H.; Ranjan Dhar, B.; Nakhla, G. Single and combined effect of various pretreatment methods for biohydrogen production from food waste. *Int. J. Hydrogen Energy* **2011**, *36*, 11379–11387. [[CrossRef](#)]
33. Xiong, H.; Chen, J.; Wang, H.; Shi, H. Influences of volatile solid concentration, temperature and solid retention time for the hydrolysis of waste activated sludge to recover volatile fatty acids. *Bioresour. Technol.* **2012**, *119*, 285–292. [[CrossRef](#)] [[PubMed](#)]
34. Cesaro, A.; Belgiorno, V. Sonolysis and ozonation as pretreatment for anaerobic digestion of solid organic waste. *Ultrason. Sonochem.* **2013**, *20*, 931–936. [[CrossRef](#)] [[PubMed](#)]
35. Liu, X.; Liu, H.; Chen, J.; Du, G.; Chen, J. Enhancement of solubilization and acidification of waste activated sludge by pretreatment. *Waste Manag.* **2008**, *28*, 2614–2622. [[CrossRef](#)] [[PubMed](#)]
36. Quarmby, J.; Scott, J.R.; Mason, A.K.; Davies, G.; Parsons, S.A. The application of ultrasound as a pre-treatment for anaerobic digestion. *Environ. Technol. (United Kingdom)* **1999**, *20*, 1155–1161. [[CrossRef](#)]
37. Hansen, K.H.; Angelidaki, I.; Ahring, B.K. Anaerobic digestion of swine manure: Inhibition by ammonia. *Water Res.* **1998**, *32*, 5–12. [[CrossRef](#)]
38. Rajagopal, R.; Massé, D.I.; Singh, G. A critical review on inhibition of anaerobic digestion process by excess ammonia. *Bioresour. Technol.* **2013**, *143*, 632–641. [[CrossRef](#)]
39. Arumugam, A.; Ponnusami, V. Biodiesel production from Calophyllum inophyllum oil a potential non-edible feedstock: An overview. *Renew. Energy* **2019**, *131*, 459–471. [[CrossRef](#)]
40. Jiang, Y.; McAdam, E.; Zhang, Y.; Heaven, S.; Banks, C.; Longhurst, P. Ammonia inhibition and toxicity in anaerobic digestion: A critical review. *J. Water Process Eng.* **2019**, *32*, 100899. [[CrossRef](#)]
41. Shi, X.; Lin, J.; Zuo, J.; Li, P.; Li, X.; Guo, X. Effects of free ammonia on volatile fatty acid accumulation and process performance in the anaerobic digestion of two typical bio-wastes. *J. Environ. Sci.* **2017**, *55*, 49–57. [[CrossRef](#)]

