

## Article

# Investigating the Enhancement in Biogas Production by Hydrothermal Carbonization of Organic Solid Waste and Digestate in an Inter-Stage Treatment Configuration

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**Abstract:** In recent years, sewage sludge (SS) and bio-waste management have attracted increasing environmental attention. In this study, hydrothermal carbonization (HTC) technology is investigated in the framework of a co-treatment of sewage sludge digestate (SSD) and an organic fraction of municipal solid waste (OW). The proposed configuration integrates HTC with anaerobic digestion (AD) in an inter-stage configuration (AD1 + HTC + AD2). The effects of different percentages of OW added to SSD in the HTC treatment are evaluated in terms of characteristics and methane yield of the produced HTC liquor (HTCL) and HTC slurry (i.e., the mixture hydrochar-HTCL), as well as dewaterability of the HTC slurry. Results show that, with the increase in the percentage of OW in the OW-SSD mixture fed to the HTC process, production of biogas and biomethane of both HTC slurry and HTCL increases. The highest biogas production is achieved when a mixture consisting of half SSD and half OW is used, reaching  $160 \pm 10$  and  $240 \pm 15$  mL biogas  $g^{-1}$  COD<sub>added</sub>, respectively, for HTCL and HTC slurry. Furthermore, sludge dewaterability is significantly improved by the combined AD1-HTC-AD2 process. Finally, an energy assessment allows estimating that the co-treatment of OW with SSD in HTC can cover up to 100% of the energy consumption of the system.

**Keywords:** anaerobic digestion; hydrothermal carbonization; digestate; organic fraction of municipal solid waste; biogas production; dewaterability; sludge reduction



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## 1. Introduction

The production and handling of municipal sewage sludge (SS) and bio-waste represent a problem worldwide. Although SS, a sludge-like waste resulting from municipal wastewater treatment plants (WWTP), was recently estimated to reach an annual sludge production in the EU-28 of 9.0–9.5 million tons of dried solid (d.s) (2017 data) [1], bio-waste—mainly food and garden wastes—accounts for more than 34% of municipal solid waste (MSW) generated, amounting to 86 million tons in 2017 in the EU-28 [2]. Dealing with these waste feedstocks in an environmentally friendly way can help achieve a few of the Sustainable Development Goals (SDGs) recently adopted by the United Nations (UN). The SDG's aims are to develop renewable energy sources and improve efficiency in the use of resources, to increase the global economy's sustainability, and to implement the circular economy's principles, thus limiting the amount of waste [3]. According to European Parliament Directive 2006/12/EC on waste, "the use of waste as a source of energy" should be encouraged by the Member States as a good practice to avoid landfilling and prevent pollution [4].

In recent years, different technologies or combinations of different processes have been developed for the reduction [5,6], treatment, and reuse [7] of both SS and bio-waste while looking at environmental and energy sustainability options. Among these, the anaerobic digestion (AD) process is, to date, the most widely used technology, producing (i) biogas, a gas rich in methane (typically 55–70% *v/v*) and carbon dioxide (30–45% *v/v*), which can be used to generate renewable energy, and (ii) digestate, a by-product of the process which, depending on its properties, can be employed as it is or after post-treatment as a soil fertilizer due to its NPK (Nitrogen, Phosphorus, and Potassium) content and carbon residue.

A recent analysis [8] has shown that around 46 million tons of digestate are produced from the organic fraction of mixed MSW (mechanical biological treatment—MBT) in the EU-28 per year, of which at least 7 million tons are from source-separated bio-waste, and smaller quantities (ca. 1.7 million tons each) are from SS and agro/food industry by-products. In addition, 120 million tons of digestate are produced in the EU from a mixture of manure and energy crops [8]. The majority of digestate is used directly as fertilizer.

Nevertheless, concerning bio-waste, AD is not always technically feasible, for example, for high shares of garden waste, or for the presence of undesired materials such as glass, metals, or plastic. Furthermore, although AD is a mature technology from a technical and management point of view, it requires long processing times and, consequently, large treatment volumes, especially when bio-wastes are treated. Indeed, the limiting factor of AD is the hydrolysis step, i.e., the conversion of complex organic compounds (i.e., proteins, long-chain fatty acids, and complex carbohydrates) into simpler compounds (such as monosaccharides, amino acids, and volatile fatty acids) that can be easily metabolized by anaerobic bacteria.

Finally, the use of digestate in agriculture obtained from both SS and bio-waste is not acceptable in many cases, as feedstock is often not suitable in terms of content in heavy metals (Cd, Ni, Pb, Cu, Zn, Hg, As, Cr), polycyclic aromatic hydrocarbons (PAH16), polybrominated diphenyl ethers (PBDE), di-isononyl-phthalate (DINP), di(2-ethylhexyl)phthalate (DEHP), short- and long-chain perfluorinated surfactants (PFAS), hexabromocyclododecane (HBCD), dioxins, furans and dioxin-like PCB's, bisphenol A, pesticides, hormones, siloxanes, drugs and pharma, and microplastics [8]. Therefore, other disposal routes are chosen, such as incineration and landfilling. Interestingly, digestate has a high residual content of non-biodegradable organic compounds that can be further energetically valorized.

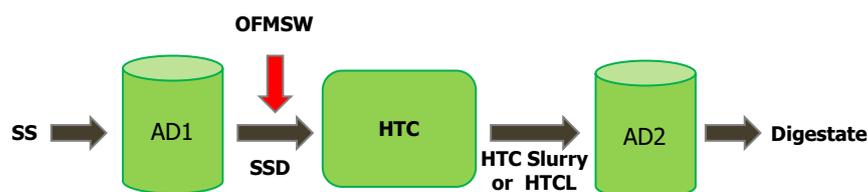
If either AD is not (technically) feasible or land application of digestate is not allowed, other system configurations for the treatment of SS and bio-waste should be explored, also including a co-treatment of these wastes.

Hydrothermal treatments, in particular hydrothermal carbonization (HTC), have gained increasing attention in recent years for treatment of biomasses with high moisture content [9], including SS [10–12], the organic fraction of MSW (OFMSW) [13,14], and digestate [15]. HTC is a promising process to exploit the potential of biomass for cleaner production, in line with the strategy for a circular economy-based society implemented by the European Union [9,16]. It is a thermo-chemical process that can convert organic feedstocks with high-moisture content into a slurry consisting of a carbonaceous product, called hydrochar, and a liquid (HTCL) with a high load of organic and inorganic compounds. Hydrochar is characterized by higher carbon content and lower hydrogen and oxygen to carbon ratios than the initial substrate [17,18], with surface functionalization patterns that permit hydrochar utilization in numerous end-use applications [19], such as solid fuel [17], microbial carriers in soil [20] and AD [21], adsorbent for pollutants [22,23], and soil amendment [22]. HTCL is characterized by high concentrations of organic matter, and a promising recovery route is AD [24] or recycling in the activated sludge process [25]. In HTC, biomass is treated in the presence of liquid water under saturated pressure at temperatures between 180 and 250 °C for a residence time of several minutes to several hours.

Due to HTC by-product characteristics, the coupling of HTC and AD is being investigated for different waste biomasses, including SS and OFMSW, as recently reviewed by Ischia and Fiori [26]. The coupling of HTC and AD has some advantages, as it allows one to reduce sludge production, improve sludge dewaterability, and increase energy and material recovery. Although HTC is a process that requires an energy source, the recovery of energy from AD of the HTC by-products can possibly enable an energy self-sustaining process [16].

Different system configurations have been proposed, including pre-treatment of biomass by HTC before AD (HTC-AD) [27] or post-treatment of digestate by HTC and recycling HTCL to AD (AD + HTC) [28]. However, some studies confirm the potential of an inter-stage treatment configuration with thermal [29] and hydrothermal [30] treatment between two anaerobic digesters (the first being fed with untreated biomass) in terms of overall biogas increase and volatile solid destruction. The scope of the inter-stage treatment configuration is that the first AD (AD1) mainly converts most of the bioavailable and readily biodegradable organic matter into biomethane, leaving the poorly biodegradable organic matter mainly unaltered. Some of the poorly biodegradable organic matter can then be solubilized by the inter-stage HTC treatment and possibly converted to biomethane by the second AD (AD2).

To the best of the authors' knowledge, no study has yet been conducted to investigate both the treatment of digestate from SS (SSD) and OFMSW using HTC in an inter-stage treatment configuration (i.e., AD1-HTC-AD2) (Figure 1), with the aim of increasing biogas production.



**Figure 1.** System configuration integrating the HTC of SS digestate and OFMSW with a post-treatment with AD in an inter-stage HTC configuration.

Given this background, this study aims to investigate the effects of different percentages of OFMSW added to SSD in the HTC treatment in terms of characteristics and methane yields of both HTC slurry and the HTCL produced, as well as the dewaterability of HTC slurry. At the same time, the research focuses on the evaluation of energy recovery resulting from the combined treatment of SSD and OFMSW in an inter-stage HTC configuration.

## 2. Materials and Methods

### 2.1. Feedstocks Characterization

The experimental study was conducted using two different biomasses as feedstock for the HTC reactor: anaerobic sewage sludge digestate (SSD) from a municipal WWTP and the organic fraction of municipal solid waste (OFMSW). SSD used in this study was a mixture of the sludge leaving the anaerobic digester (digestate), with a dry matter content of  $2.8 \pm 0.1\%$ , and the same digestate after the addition of polyelectrolyte and passage in a centrifuge (dewatered sludge), with a dry matter content of  $21.1 \pm 0.2\%$ , collected from the municipal WWTP of Trento, Italy. Both digestate and dewatered sludge were mixed to obtain the first feedstock for the HTC reactor with a dry matter content of  $10.2 \pm 0.3\%$ , which could be easily pumped into an industrial-scale HTC reactor [28]. The use of SSD as feedstock for the HTC tests allowed the AD1-HTC step of the AD1-HTC-AD2 inter-stage configuration to be simulated.

The OFMSW used in this study, called OW (organic waste), was collected from the storage tank of the OFMSW treatment plant located in Lana (Bolzano, Italy), after a shredding pretreatment.

Different percentages of OW were mixed with SSD to obtain three different feedstocks for the HTC reactor: designated 90SSD + 10OW, 80SSD + 20OW, and 50SSD + 50OW, corresponding, respectively, to 10%, 20%, and 50% of OW addition to the total mass of the inlet feedstock to the HTC reactor.

The characteristics of SSD, OW, and the mixed raw materials used in the HTC tests are listed in Table 1.

**Table 1.** Characteristics of DIG, OFMSW, 90SSD + 10OW, 80SSD + 20OW, and 50SSD + 50OW.

Parameter	SSD	OW	90SSD + 10OW	80SSD + 20OW	50SSD + 50OW
TS [%]	10.2 ± 0.3	16.4 ± 0.6	10.8 ± 0.5	11.2 ± 0.1	11.8 ± 0.3
TKN [g TKN L <sup>-1</sup> ]	7.4 ± 2.5	5.5 ± 2.2	6.8 ± 2.5	6.6 ± 0.5	6.4 ± 1.0
NH <sub>4</sub> -N [mg NH <sub>4</sub> -N L <sup>-1</sup> ]	2.0 ± 0.5	0.7 ± 0.3	1.6 ± 0.5	1.5 ± 0.3	1.3 ± 0.4
PO <sub>4</sub> -P [mg PO <sub>4</sub> -P L <sup>-1</sup> ]	700 ± 10	120 ± 7	600 ± 6	480 ± 5	350 ± 5
TP [g TP L <sup>-1</sup> ]	3.5 ± 0.3	0.8 ± 0.2	2.7 ± 0.4	2.4 ± 0.2	2.1 ± 0.4
sCOD [g sCOD L <sup>-1</sup> ]	13 ± 2	78 ± 9	27 ± 5	32 ± 3	40 ± 5
COD [g COD L <sup>-1</sup> ]	116 ± 15	227 ± 12	120 ± 12	134 ± 10	175 ± 17

TS: total solid; TKN: total Kjeldahl nitrogen; NH<sub>4</sub>-N: ammonium nitrogen; PO<sub>4</sub>-P: soluble phosphorus; TP: total phosphorus; sCOD: soluble chemical oxygen demand; COD: total chemical oxygen demand.

## 2.2. Hydrothermal Carbonization Tests

Hydrothermal carbonization tests were carried out in duplicate in a 2 L batch reactor at a temperature of 190 °C and with a 1 h treatment time. The HTC reactor was loaded with (i) 1.20 ± 0.10 kg SSD with a TS concentration of 10.2 ± 0.3%; (ii) 1.08 ± 0.10 kg SSD and 0.12 ± 0.01 kg OW for testing 90SSD + 10OW to obtain a mixture with a TS content equal to 10.8 ± 0.2%; (iii) 0.96 ± 0.10 kg SSD and 0.24 ± 0.01 kg OW for testing 80SSD + 20OW to obtain a mixture with a TS content of 11.2 ± 0.1%; and (iv) 0.60 ± 0.10 kg SSD and 0.60 ± 0.10 kg OW for testing 50D + 50OW to obtain a mixture with a TS content of 11.8 ± 0.1%.

The procedure for the HTC tests has already been described in detail [31]. At the end of the HTC reaction, part of the slurry, consisting of liquid and solid (hydrochar) fractions, was collected and kept at 4 °C for further analyses, and another part was filtered with a qualitative filter paper to separately obtain the liquid fraction (HTCL). The solid yield was calculated as the ratio between the mass of hydrochar after the HTC process and the mass of the feedstock before the treatment, both expressed on a dry basis. Gas yield was calculated as the mass of gas produced, consisting only of CO<sub>2</sub>, per unit of mass of the initial feedstock, both expressed on a dry basis. The liquid yield was determined by difference.

## 2.3. Anaerobic Digestion Tests

Anaerobic digestion tests were carried out to assess the biochemical methane potential (BMP) of the SS, HTC slurry, and HTCL generated by the HTC tests. The tests were conducted under mesophilic conditions over a period of 30 days using serum bottles with a volume of 135 mL inoculated with anaerobic digested sludge from Trento WWTP, Italy. The main characteristics of the inoculum were: TS 2.8 ± 0.1%, vs. 77 ± 0.3%, COD 35 ± 2 g COD L<sup>-1</sup>, sCOD 20 ± 1 g sCOD L<sup>-1</sup>, TKN 2.5 ± 0.1 g TKN L<sup>-1</sup>, NH<sub>4</sub>-N 0.5 ± 0.1 g NH<sub>4</sub>-N L<sup>-1</sup>, PO<sub>4</sub>-P 210 ± 7 mg PO<sub>4</sub>-P L<sup>-1</sup>, TP 740 ± 40 mg TP L<sup>-1</sup>, and a pH of 7.5. The inoculum was pre-incubated at 35 ± 0.1 °C for 14 days to completely biodegrade any organic substance eventually present in the digestate. The anaerobic digestion tests were carried out in triplicate, using a feeding/inoculum ratio (F/I) of 0.5 g COD<sub>substrate</sub> g<sup>-1</sup> COD<sub>inoculum</sub>, as suggested by Angelidaki et al. [32], with a serum

bottle filling volume fixed at 90 mL. Biogas and biomethane production were measured according to the procedure described by Ferrentino et al. [33]. The collected data allowed the determination of specific biogas production (SBP) and specific methane production (SMP), which are expressed as  $\text{mL}_{\text{biogas}} \text{g}^{-1} \text{COD}_{\text{added}}$  and  $\text{mL CH}_4 \text{g}^{-1} \text{COD}_{\text{added}}$ , respectively.

#### 2.4. Dewaterability Tests

To measure the filterability of the HTC slurry compared to untreated samples, capillary suction time (CST) tests were performed in triplicate. CST is the time required for water separated from sludge to travel a certain distance through a filtration paper (Whatman No. 17). The test was carried out using a capillary suction timer type 304B manufactured by Triton Electronics Ltd. CST tests were performed according to Standard Methods [34] on 10 mL of distilled water and HTC slurry. The CST value of the distilled water (6.4 s) was subtracted from each measured value, and these CST values were normalized to the initial TS concentration of the sample.

#### 2.5. Analytical Methods

Chemical analyses were carried out in triplicate on the untreated SSD, OW, and their mixtures before HTC treatment. Characterization of the HTC slurries and HTCLs was also carried out after all HTC treatments. Volatile solids (VS), total solids (TS), total chemical oxygen demand (TCOD), soluble COD (sCOD), ammonium nitrogen ( $\text{NH}_4\text{-N}$ ), total Kjeldahl nitrogen (TKN), total phosphorus (TP), and soluble phosphorus ( $\text{PO}_4\text{-P}$ ) concentrations were quantified according to Standard Methods [34]. sCOD,  $\text{NH}_4\text{-N}$ , and  $\text{PO}_4\text{-P}$  were measured after filtration of the samples on a  $0.45 \mu\text{m}$  paper filter.

### 3. Results and Discussion

#### 3.1. Yields of the HTC Tests

Figure 2 shows the yield of solid, liquid, and gas assessed for each feedstock used, resulting from the HTC tests.

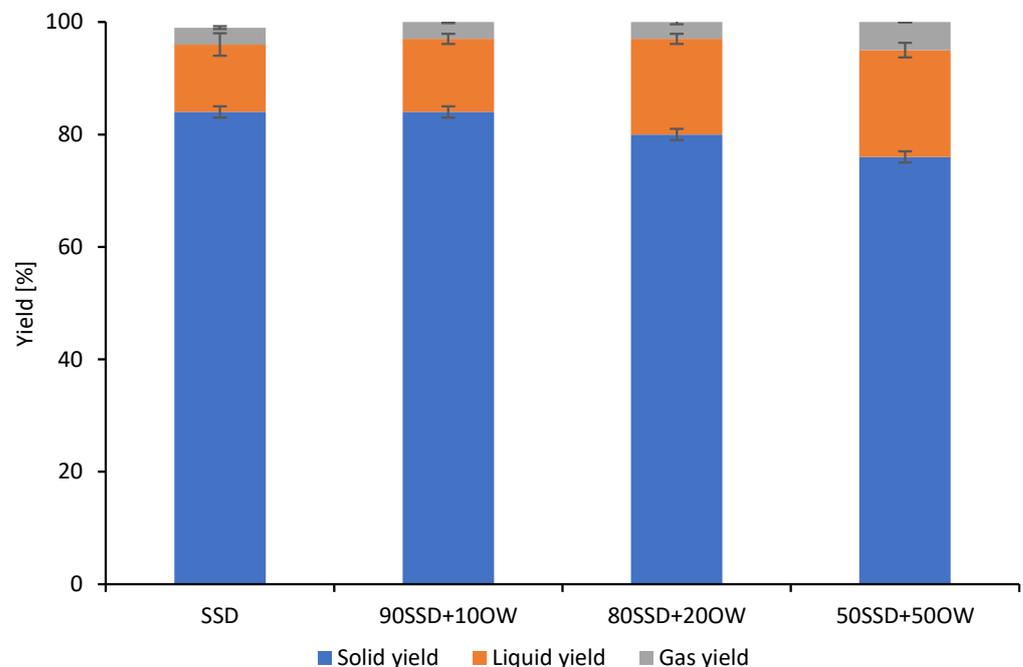


Figure 2. Solid, liquid, and gas yields.

The results show that the solid yield for the SSD sample was  $84 \pm 1\%$ , the same as that of the 90SSD + 10OW sample, also accounting for  $84 \pm 1\%$ , which indicates that the addition of 10% OW to the total mass of feedstock did not cause any significant difference

from the SSD test. On the other hand, when the percentage of OW added to the SSD was increased, the solid yield decreased to  $80 \pm 1\%$  and  $76 \pm 1\%$  for the 80SSD + 20OW and 50SSD + 50OW samples, respectively. The gas yield was  $3.0 \pm 0.3\%$  for the SSD, 90SSD + 10OW, and 80SSD + 20OW samples, whereas a slight increase of  $5.0 \pm 0.7\%$  was observed for the 50SSD + 50OW sample. As is apparent, the latter showed the highest liquid yield, equal to  $19 \pm 1\%$ , among all the feedstock tested, corresponding to the sample that showed the highest volume reduction in the solid phase and, thus, the lowest amount of solid waste to be disposed of. These data are consistent with those reported in the literature for SS but contrast those of OFSMW, which have a lower solid yield. Liu et al. [35] in their review reported an average solid yield of 60.2% for HTC from SS, varying from 22.6% and 84.9% for all studies considered. High variability in solid yield was attributed to different severities of the HTC reaction in terms of temperature and treatment time, which had a major impact on the performance of the HTC process [35]. Recently, Lucian et al. [14] reported that the yield of the hydrochar, the HTCL, and the gas for the HTC of OFMSW, performed at a reaction time of 1 h and with a temperature of  $180\text{ }^{\circ}\text{C}$ , were 67%, 32% and 1%, respectively.

### 3.2. Slurry and HTCL Samples Characterization

After the HTC reaction, the HTCL and HTC slurry was characterized by chemical analyses (Table 2).

**Table 2.** Chemical characterization of HTCLs (L) and HTC slurries (S).

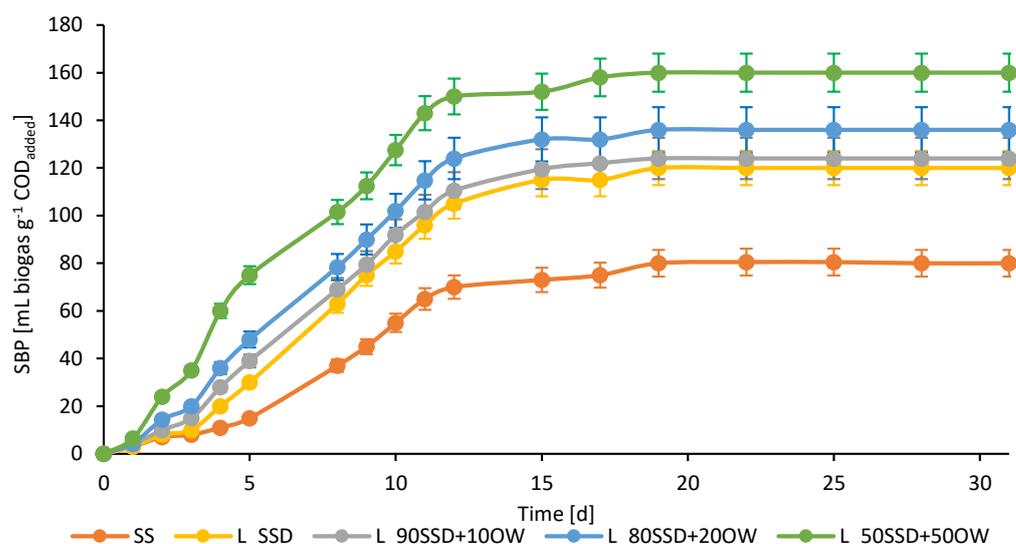
Parameter	L_SSD	L_90SSD + 10OW	L_80SSD + 20OW	L_50SSD + 50OW	S_SSD	S_90SSD + 10OW	S_80SSD + 20OW	S_50SSD + 50OW
TCOD [g TCOD L <sup>-1</sup> ]	41 ± 3	50 ± 7	72 ± 5	105 ± 3	96 ± 10	115 ± 7	125 ± 15	170 ± 10
TKN [g TKN L <sup>-1</sup> ]	5.8 ± 0.1	6.1 ± 0.2	6.0 ± 0.5	6.0 ± 0.1	4.8 ± 0.3	5.7 ± 0.2	5.8 ± 0.2	6.2 ± 0.2
NH <sub>4</sub> -N [g NH <sub>4</sub> -N L <sup>-1</sup> ]	1.4 ± 0.2	1.4 ± 0.2	1.3 ± 0.3	0.9 ± 0.3	1.4 ± 0.2	1.4 ± 0.2	1.3 ± 0.3	0.9 ± 0.3
TP [g TP L <sup>-1</sup> ]	0.15 ± 0.01	0.18 ± 0.01	0.13 ± 0.01	0.25 ± 0.01	3.4 ± 0.3	2.4 ± 0.2	2.2 ± 0.1	2.3 ± 0.3
PO <sub>4</sub> -P [g PO <sub>4</sub> -P L <sup>-1</sup> ]	0.14 ± 0.01	0.16 ± 0.01	0.15 ± 0.01	0.22 ± 0.01	0.14 ± 0.01	0.16 ± 0.01	0.15 ± 0.01	0.22 ± 0.01
TS [%]	3.1 ± 0.1	3.4 ± 0.5	3.5 ± 0.2	4.1 ± 0.1	8.6 ± 0.1	9.0 ± 0.1	9.0 ± 0.1	8.9 ± 0.2
VS [% d.b.]	78.5 ± 0.5	87.1 ± 0.1	86.7 ± 0.2	88.7 ± 0.5	70.1 ± 1.9	74.3 ± 1.2	77.6 ± 1.8	86.0 ± 1.1

The TCOD concentrations of both the HTCL and the HTC slurries confirmed that the HTC treatment hydrolyzed the feedstock, transferring low-weight molecular organic compounds from the biomass to the liquid phase. Therefore, the TCOD concentration of the HTC slurries remained unchanged compared to that of the initial biomass, whereas the TCOD concentration of the HTCL, which corresponds to the soluble fraction of the COD of the HTC slurry, increased compared to the untreated samples. Specifically, the ratio between TCOD<sub>HTCL</sub> and TCOD<sub>SLURRY</sub> of the untreated feedstocks was 13%, 22%, 24% and 23% for the SSD, 90SSD + 10OW, 80SSD + 20OW, and 50SSD + 50OW samples, respectively. On the other hand, the TCOD<sub>HTCL</sub> and TCOD<sub>SLURRY</sub> ratio, after the HTC process, increased up to 43% for the SSD and 90SSD + 10OW samples, up to 48% for the 80SSD + 20OW sample, and up to 62% for the 50SSD + 50OW sample. This shows that the TCOD solubilization increased with the increase in the OW fraction added to the initial feedstock. Considering the nitrogen compounds, both the TKN and the NH<sub>4</sub>-N concentrations slightly decreased after HTC treatment compared to the initial feedstocks in all tests, which can be attributed to some loss of ammonia in a gaseous form. Regarding TP, it accumulated in the solid phase, i.e., in the hydrochar, as reported in numerous literature studies [36]. For this reason, the concentration of TP in the HTCLs was very low and similar to the concentration of PO<sub>4</sub>-P, whereas all TP present in the untreated feedstocks was found in the HTC slurries, confirming that P segregated into the solid phase (hydrochar). Finally, an analysis of the TS concentration confirmed that the HTC process also allowed for a reduction in sludge amount, variable from 16% (for SSD and 90SSD + 10OW samples) up to 24% (for 50SSD + 50OW sample).

### 3.3. Biogas and Biomethane Production

Anaerobic digestion tests were conducted to evaluate potential biogas and biomethane production (BMP) of the SS, HTCL, and HTC slurry of each feedstock. Considering the AD1-HTC-AD2 inter-stage configuration, the tests conducted with the SS samples simulated biogas production in AD1, whereas the tests conducted with the HTCL and HTC slurry simulated the AD2 stage.

At the beginning of the test, each sample of SS, HTCL, and HTC slurry was mixed with the inoculum. The pH of the mixture ranged from 7.0 to 7.5 for all BMP tests, which are optimal values for the establishment of the anaerobic digestion process. Figure 3 depicts the trend of specific biogas production of SS and HTCL of the SSD, 90SSD + 10OW, 80SSD + 20OW and 50SSD + 50OW samples.



**Figure 3.** Specific biogas production of SS and HTCL of each HTC feedstock.

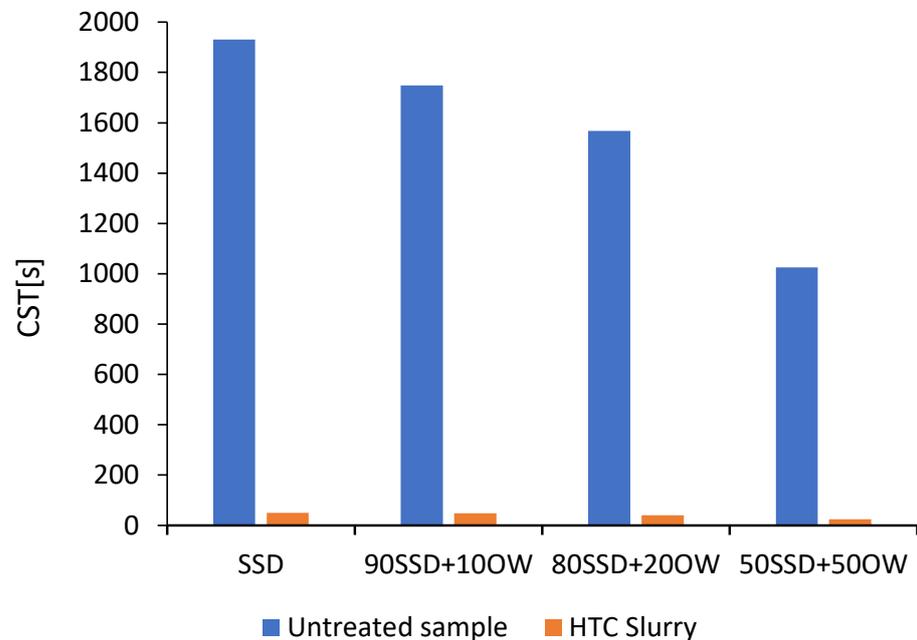
The specific biogas production deriving from the anaerobic digestion of HTCL was  $120 \pm 12$ ,  $124 \pm 10$ ,  $136 \pm 15$  and  $160 \pm 10$  mL biogas  $\text{g}^{-1}$   $\text{COD}_{\text{added}}$  for SSD, 90SSD + 10OW, 80SSD + 20OW and 50SSD + 50OW, respectively. The specific biogas production of the untreated SS, when used as feed of the AD process, was  $80 \pm 8$  mL biogas  $\text{g}^{-1}$   $\text{COD}_{\text{added}}$ . Thus, compared to the specific biogas productions of SS, the results show that the HTCL derived from the HTC of SSD had 50% higher biogas production. This result is in agreement with that of Aragón-briceño et al. [37]. They found that HTCL from SSD at low HTC temperatures (160–220 °C) had a specific biogas production of 260–277 mL $\text{CH}_4$   $\text{g}^{-1}$   $\text{COD}_{\text{added}}$ , which is higher than that of the untreated samples.

Moreover, biogas production increased when the percentage of OW added to the total volume of the feedstock increased. More specifically, the HTCL obtained from the addition of 10% OW to 90% of the digestate did not significantly increase biogas production compared to the test using only HTCL from SSD. However, increasing the mass of OW by 20% (compared to the total mass of HTC feedstock) resulted in a 70% increase in biogas production compared to SS and a 13% increase compared to HTCL from SSD. The best result was obtained when the AD test was conducted with HTCL from HTC feedstock consisting of half OW and half SSD. In this case, a 100% increase in specific biogas production was observed compared to SS. The advantages of pre-treated OFMSW with the HTC process were recently reported by Lucian et al. [14]. The authors showed that the SMP of HTCL increased by about 37% (at 180 °C, 1 h) compared to the SMP of untreated OFMSW, which accounted for 149 mL $\text{CH}_4$   $\text{g}^{-1}$   $\text{COD}_{\text{added}}$ . Recently, experimental studies have shown that the presence of hydrochar can affect biogas production. In fact, controversial results have been reported. Aragón-Briceno et al. [37] reported that hydrochar did not significantly affect the anaerobic process. On the other hand, several researchers proved that hydrochar



### 3.4. Dewaterability Improvement

To assess the dewaterability of the HTC treated samples, measurements of CST were performed and compared with those of the untreated feedstock. The results are reported in Figure 5.



**Figure 5.** The trend of CST measurements for untreated and HTC treated samples.

As expected, the results show that the CST values decreased with increasing the percentage of OW added to the SSD both in the untreated samples and in the HTC slurries. However, in all slurries obtained after HTC treatment, there was a significant decrease in the CST value by up to 98%. Since the HTC slurries had a different concentration of TS than the untreated samples, the CST value was also reported in relation to TS to neutralize the effect of the total solid concentration. A slight decrease of  $0.58 \pm 0.08 \text{ s g}^{-1} \text{ L}$  in the CST/TS value was observed for the HTC slurry corresponding to the SSD sample, up to  $0.53 \pm 0.12$ ,  $0.45 \pm 0.06$ , and  $0.27 \pm 0.05 \text{ s g}^{-1} \text{ L}$  for the HTC slurries of the 90SSD + 10OW, 80SSD + 20OW and 50SSD + 50OW samples, respectively. The results of the CST measurements are reported in Table 4.

**Table 4.** Results of Capillary Suction Time (CST) measurements referring to TS concentration.

	Sample	CST [s]	TS [g L]	CST/TS [ $\text{s g}^{-1} \text{ L}$ ]
Untreated sample	SSD	1930 ± 15	102	18.92 ± 0.15
	90SSD + 10OW	1750 ± 10	108	16.16 ± 0.21
	80SSD + 20OW	1570 ± 20	112	14.00 ± 0.16
	50SSD + 50OW	1030 ± 10	118	8.69 ± 0.11
HTC slurry	SSD	50 ± 4	86	0.58 ± 0.08
	90SSD + 10OW	48 ± 4	90	0.53 ± 0.12
	80SSD + 20OW	41 ± 4	90	0.45 ± 0.06
	50SSD + 50OW	24 ± 5	89	0.27 ± 0.05

In general, the results in Table 4 confirm that HTC treatment at 190 °C with a reaction time of 1 h significantly improves the dewaterability of the treated biomass, thus contributing to reductions in volume of solid sludge to be disposed. Furthermore, these results confirm that HTC treatment leads to the breaking of sludge flocks and thus to increases in free water. Thus, the HTC process is able to convert water chemically bound to the particles and the stored water in free water, thus improving dewaterability [40]. These results are

consistent with other literature studies. Kim et al. [41], who treated SSD with the HTC process at 180, 220 and 250 °C for 30 min, confirmed a reduction in the CST value from 72 s (raw sludge) to 15.3, 10.9, and 10.1 at 180, 220 and 250 °C, respectively. Furthermore, Ahmed et al. [42], found an increase in dewaterability by a factor of seven for HTC of SSD at 190 °C and a reaction time of 1 h.

### 3.5. Energy Evaluation

This paragraph reports the energy assessment of an HTC system coupled with AD in an inter-stage treatment configuration (i.e., AD1-HTC-AD2).

In the AD1-HTC-AD2<sub>HTCL</sub> scenario (Figure 6a), the HTCL derived from HTC of SSD is used as feedstock for an AD2 process, and the hydrochar can be recovered and used for further applications. Conversely, in the AD1-HTC-AD2<sub>S</sub> scenario (Figure 6b), all the slurry derived from the HTC of SSD is fed to the AD2 unit. The mass and energy balances are performed based on the experimental HTC yields obtained in this study and are based on 100 m<sup>3</sup> SS at 2.8% TS treated per day in AD1. SS is characterized by a concentration of 13 g sCOD L<sup>-1</sup> with an SMP of 45 mL g<sup>-1</sup> COD. The energy demand for the AD process can be estimated to be 25% of the energy recovered from the biogas produced [43]. Therefore, the treatment of 100 m<sup>3</sup> of SS in AD1 results in methane production of 59 m<sup>3</sup> CH<sub>4</sub>, which corresponds to 590 kWh considering that 1 m<sup>3</sup> CH<sub>4</sub> can provide 10 kWh [44]. According to the proposed schemes (Figure 6), the digestate is dewatered or thickened to obtain a 10% TS slurry and then treated by HTC at 190 °C for 1 h. The energy demand for solid/liquid separation with a screw press can be estimated at 10 to 30 kWh per ton of treated sludge [45]. Assuming an average power consumption of 20 kWh per ton of treated sludge, the energy requirement for treating 100 m<sup>3</sup> of SSD with a screw press is 2000 kWh.

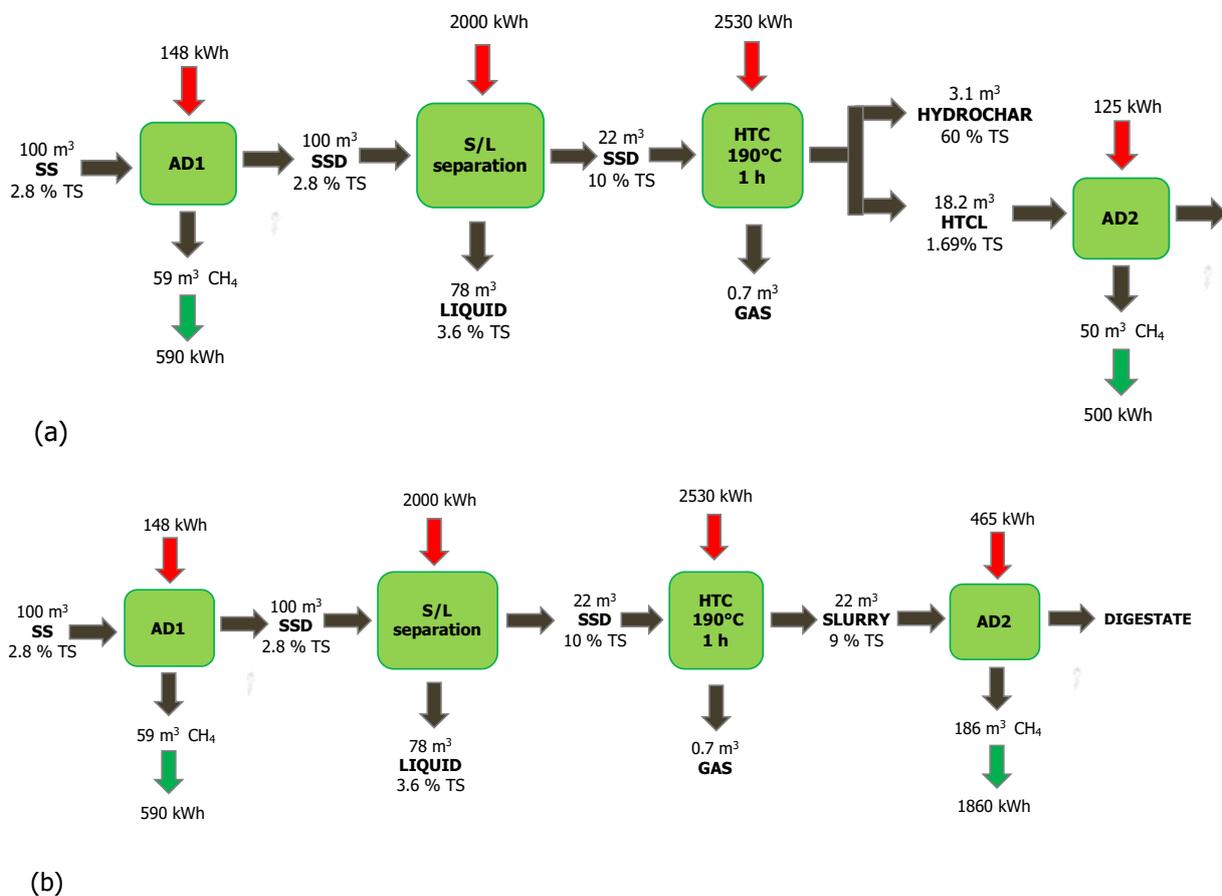
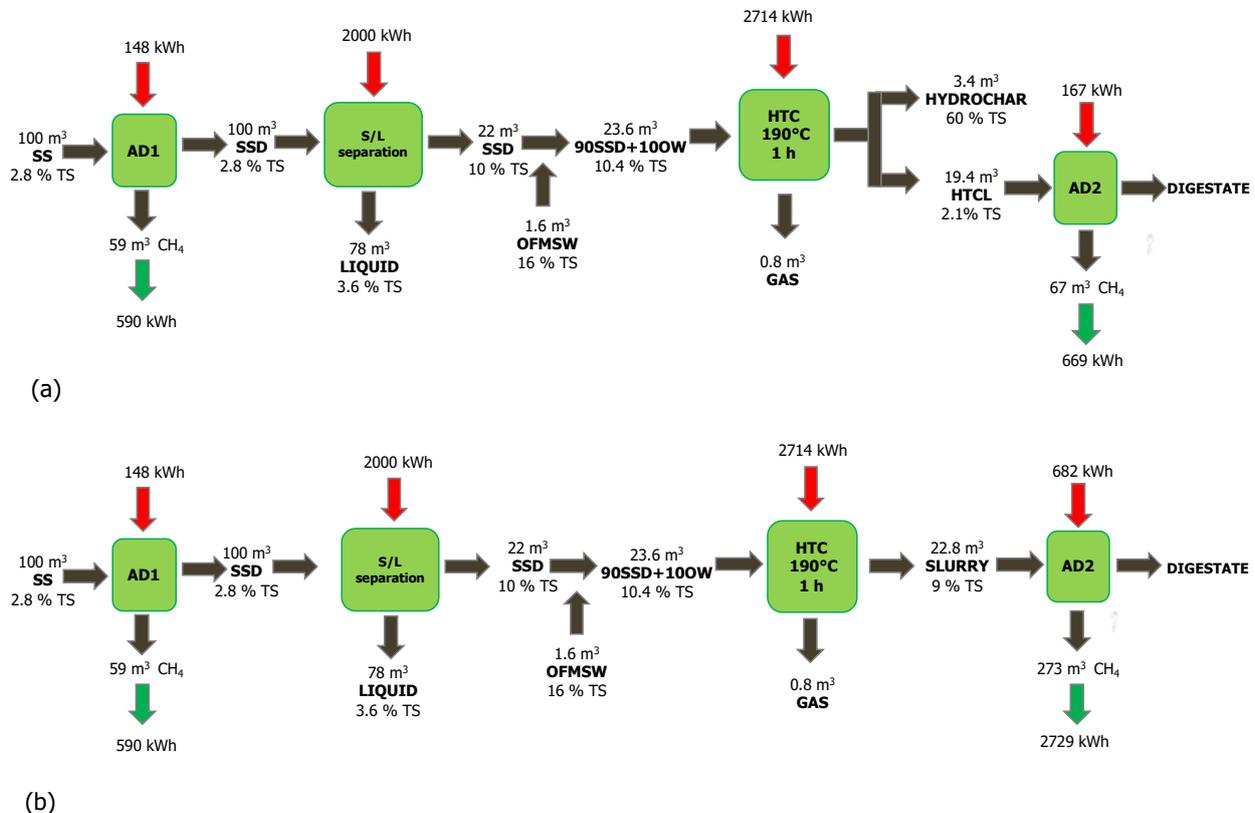


Figure 6. Energy and mass balances of (a) AD1-HTC-AD2<sub>HTCL</sub> and (b) AD1-HTC-AD2<sub>S</sub> scenarios.

After HTC treatment, hydrochar is dewatered to produce  $3.1 \text{ m}^3$  of wet hydrochar with 60% TS [12] and  $18.2 \text{ m}^3$  of HTCL with 1.7% TS per day. HTCL is characterized by a concentration of  $41 \text{ g COD L}^{-1}$  with an SMP of  $67 \text{ mL CH}_4 \text{ g}^{-1} \text{ COD}$ . Thus, treating the HTCL in AD2<sub>HTCL</sub> leads to methane production of  $50 \text{ m}^3 \text{ CH}_4$  corresponding to 500 kWh. The thermal and electrical energy demands for the HTC process are estimated to be about 100 and 15 kWh per ton of sludge treated, respectively [12,42]. In this analysis, the energy input for the combined AD1-HTC-AD2<sub>HTCL</sub> process is 4800 kWh, whereas the energy output, due to the enhancement of  $\text{CH}_4$  from the SS and HTCL fed to AD1 and AD2, respectively, is 1090 kWh. The energy ratio, defined as the ratio between energy output and energy input, is thus 23%. Hence, feeding HTCL from the SSD to the AD2 unit can only cover 23% of the energy required for the combined process. Ahmed et al. [42], considering an HTC-AD configuration, reported that the AD of HTCL can only cover one quarter of HTC's energy needs.

In the AD1-HTC-AD2<sub>S</sub> scenario, the amount of energy generated by treating  $100 \text{ m}^3$  SS in AD1 is 590 kWh, whereas the solid/liquid separation energy requirement is 2000 kWh, as in the previous scenario. Afterward, the 10% thickened sludge is fed into the HTC reactor at  $190^\circ \text{C}$  for 1 h. All the derived slurry,  $22 \text{ m}^3$  at 9% TS, is fed to the AD2 unit, resulting in an increase in the production of  $186 \text{ m}^3$  of  $\text{CH}_4$ , corresponding to 1860 kWh. In this case, energy input for the combined AD1-HTC-AD2<sub>S</sub> is 5150 kWh, whereas energy output, due to the enhancement of  $\text{CH}_4$  derived from the SS and whole HTC slurry feeding to AD1 and AD2, respectively, is 2450 kWh. Thus, the inter-stage configuration AD1-HTC-AD2<sub>S</sub> enables up to 48% of the energy needs of the combined process to be met.

Then, the contribution of OFMSW to energy balance is considered. Similarly, in the AD1-HTC<sub>SSD+OW</sub>-AD2<sub>HTCL</sub> (Figure 7a) and AD1-HTC<sub>SSD+OW</sub>-AD2<sub>S</sub> (Figure 7b) scenarios, the HTCL and the slurry, derived from HTC of 90% SSD and 10% OW, are fed into the AD2.



**Figure 7.** Energy and mass balances of (a) AD1-HTC<sub>DIG+OW</sub>-AD2<sub>HTCL</sub>- and (b) AD1-HTC<sub>DIG+OW</sub>-AD2<sub>S</sub>-scenarios.

Table 5 summarizes the data for the energy and mass balances of each scenario. Feeding HTCL back to AD2 only partially contributes to covering the energy required for the inter-stage AD1-HTC-AD2 process. Conversely, feeding HTC slurry into the AD2 compartment allows for a significant increase in biogas production, and, in the case of feeding the mixture of 50SSD + 50OW, it can completely cover the internal energy consumption of the AD1-HTC-AD2 treatment. In the case of feeding the slurry from the 50SSD + 50OW sample, not only can the entire energy consumption be covered, but also energy can be recovered to be used within the WWTP.

**Table 5.** Energy and mass balance data considering, for each scenario, (a) HTCL fed to the AD2, and (b) HTC slurry fed to AD2.

	SSD		90SSD + 10OW		80SSD + 20OW		50SSD + 50OW	
	(a) HTCL	(b) Slurry	(a) HTCL	(b) Slurry	(a) HTCL	(b) Slurry	(a) HTCL	(b) Slurry
Flow rate to AD1 [m <sup>3</sup> d <sup>-1</sup> ]	100	100	100	100	100	100	100	100
Biogas production from AD1 [m <sup>3</sup> ]	59	59	59	59	59	59	59	59
Flow rate to AD2 [m <sup>3</sup> d <sup>-1</sup> ]	18.2	22	19.4	22.8	21	24.6	28.3	34.3
Biogas production from AD2 [m <sup>3</sup> ]	50	186	67	273	99	366	291	880
Energy input for AD1 [kWh]	148	148	148	148	148	148	148	148
Energy input for S/L separation [kWh]	2000	2000	2000	2000	2000	2000	2000	2000
Energy input for HTC [kWh]	2530	2530	2714	2714	2921	2921	4140	4140
Energy input for AD2 [kWh]	125	465	167	682	248	916	729	2200
Total energy output (from biogas) [kWh]	1090	2450	1260	3320	1580	4250	3500	9400
Output/Input Ratio	23%	48%	25%	60%	30%	71%	50%	111%

The use of OFMSW alone in AD can result in considerable biogas production and thus energy recovery. Treating 100 m<sup>3</sup> OFMSW per day with an sCOD concentration equal to 32 g sCOD L<sup>-1</sup> and an SMP concentration of 149 mL CH<sub>4</sub> g<sup>-1</sup> COD [14] in AD can lead to energy recovery of 3500 kWh. These data confirm the high suitability of OFMSW as a booster for the AD process of substrates with lower SMP values. However, OFMSW lacks certain characteristics that may limit its efficacy as such a resource, and bio-resources such as sewage sludge and animal wastes can be used as co-substrates [46].

#### 4. Conclusions

This study shows the experimental results of co-treatment of digestate from SS and OFMSW using HTC in an inter-stage treatment configuration (i.e., AD1-HTC-AD2). Four different feedstocks were used as the feedstock for the HTC tests: digestate and three different mixtures of SSD and OFMSW. The performances of the combined AD1-HTC-AD2 process were assessed in terms of increases in biogas and methane production and in terms of increases in dewaterability and thus reduction in wastes to be disposed. The data show an increase in biogas and biomethane production as the percentage of OFMSW added to the digestate increases. When a mixture consisting of half SSD and half OFMSW was fed to the HTC process, the highest biogas production was achieved, reaching 160 ± 10 and 240 ± 15 mL biogas g<sup>-1</sup> COD<sub>added</sub>, corresponding to the treatment of HTCL and HTC slurry in AD, respectively. These configurations can allow significant energy recovery, which, in the case of feeding only HTCL to AD2, was about 70% of the energy consumption of the combined AD1-HTC-AD2 process. Furthermore, in the case of feeding all the slurry produced by the HTC process (hydrochar and HTCL) to AD, the produced biogas can cover 100% of energy consumption and have an energy surplus (11%) that can be used for other energy needs within the WWTP. Moreover, the addition of a percentage of OFMSW to the digestate, when both are used as feedstock for the HTC process, was found to be beneficial also in terms of drastically improving the dewaterability of the biomass mixture.

Future research should focus on improved integration processes aimed at combining environmental and economic sustainable development in the management of sewage sludge and organic waste. In particular, the integration of HTC with other technologies, beyond AD, can be of interest to improve the overall performance of organic sludge waste treatment while looking at material and energy recovery from the HTC products.

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