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Research Article

The Impact of Hydrothermal Carbonization Treatment on Anaerobic Digestion of Organic Fraction of Municipal Solid Waste

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Abstract
A significant portion of the produced Municipal Solid Waste (MSW) is organic materials, especially in developing countries. Most MSW management problems are pertinent to the Organic Fraction of the Municipal Solid Waste (OFMSW). In this experimental investigation, the impact of the hydrochar produced by Hydrothermal Carbonization (HTC) at different temperatures on Anaerobic Digestion (AD) of Tehran's OFMSW has been investigated. The parameters including the amount of Volatile Matter (VM), Fixed Carbon (FC), ash content, hydrochar yield, heating value, and energy yield, elemental analysis, proximate analysis, and biomethane production results were employed to examine how and why hydrochars are effective. The impact of the hydrochars produced at 150, 190, and 230°C on AD was analyzed for the OFMSW. In the hydrothermal carbonization process, the hydrochar yield declined as temperature increased while the energy yield in hydrochar-190 reached its maximum thanks to increased heating value. The impact of hydrochar on biomethane production content varied. In the hydrochars produced at 150 and 190 °C, biomethane production was increased 35.88% and 47.33%, respectively, which was due to the destruction of the hard structure of the OFMSW. However, due to the production of the inhibitors, such as phenol and furfural, in the HTC process, the biomethane production of hydrochar-230 declined by 29%. The effect of the hydrothermal carbonization on AD under the optimum condition included an increase in biomethane production and a reduction in the retention time in biomethane production.
Keywords: Hydrothermal Carbonization, Anaerobic Digestion, Organic Fraction of the Municipal Solid Waste (OFMSW), Biomethane, Hydrochar.

Introduction

Management of the municipal solid waste (MSW) is one of the major concerns in urban communities. Because of population growth, solid waste production increases, consequently threatening the health of the community and contaminating the environment. In 2018, the solid waste production rate was estimated equal to 2 billion tons per year, which will reach 4.3 billion tons by 2025 (Tyagi et al., 2018). Thereby, its management is essential than ever.

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The common MSW disposal methods include sanitary landfilling, thermal methods (in high-income countries), composting, dumping (in low-income countries). If sanitary landfilling is not carried out appropriately, it may contaminate the water resources in the vicinity of the disposal area and the surrounding soil by leachate containing heavy metals, persistent organic pollutants, and microbial pathogens. Because of poor management, the landfilled areas contaminate the air by emitting odors, greenhouse gases, and volatile organic compounds (Pham et al., 2015; Vergara & Tchobanoglous, 2015; Wilson et al., 2015; Liu et al., 2017). As a thermal method, waste incineration requires high investment and exploitation costs, leading to the production of ash, waste materials, gaseous pollutants, such as NO_X, SO_2, CO_2, dioxin, and furan. Pyrolysis and gasification that are technically making progress have not been employed at an industrial scale. The composting method has not captured public attention because of producing low-value waste products and limited waste volume reduction (Ferrari et al., 2020).

One of the MSW management challenges in developing countries is its high organic fraction and high moisture content. Organic fraction accounts for 50 to 70% of total MSW in low-income countries and 20 to 40% in high-income countries (Pham et al., 2015). If the management of the OFMSW is carried out properly, it can be a valuable renewable energy source.

Because it consists of many organic compounds, the MSW is a feedstock source for anaerobic microorganisms. OFMSW can be decomposed into simpler compounds in AD (Abudi et al., 2016). AD is a process by which microorganisms break down organic materials in the absence of oxygen. AD is an energy source and a suitable method for reducing the pollutions caused by improper management of organic wastes and preventing the emission of greenhouse gases (Abudi et al., 2016; Zamri et al., 2021; Delarestaghi et al., 2018).

Anaerobic digestion breaks down organic materials into two valuable products. 1- Biomethane: it has heating value as a renewable fuel used for heating and electricity generation or as the consumed fuel in equipment and vehicles. 2- Fertilizer: digestate materials that are employed in agriculture in a direct or combined way (Bolzonella et al., 2006).

Over the past years, AD has been widely employed as the management method of the OFMSW and other organic wastes. The produced energy has been recovered in the form of biogas. Various researchers have carried out chemical pretreatments, such as ozonation, adding acid and alkali, and physical pretreatment, such as crushing, thermal, microwave, ultrasonic, and biological pretreatments to improve and increase the biomethane content (Zeynali et al., 2017; Cesaro et al., 2019; Cesaro et al., 2014; Liu et al., 2020; Yu et al., 2019).

Hydrochar is a solid Carbonaceous absorbent obtained from the HTC as a novel method of thermal conversion of organic material (Libra et al., 2011). HTC generally takes place at the temperature range of 150 to 220 °C and the maximum pressure of 20 bar in an aquatic environment (Libra et al., 2011). According to the previous studies, adding hydrochar to the AD process increases the buffer capacity, reduces the prohibitor factors of the process, and increases the biogas production level. In a study, the maximum biogas production was 450 mL/g-VS, and methane production increased from 5/57% to 8/69%. 25 mg ammonium and 50 mg volatile fatty acids were removed per gram of hydrochar (Xu et al., 2018). In another study, the impact of hydrochar on the AD process was investigated. The highest biogas and methane content production at 140 °C were equal to 288 L/Kg VS increased by 24%, and 207 L/Kg VS increased by 37%, receptively. Besides, at the period of 10 to 13 days, biogas yield increased by 95%. Also, in this study, the methane production yield declined as the temperature increased (Choe et al., 2021).

Dusgupta and Chandel used hydrothermal pretreatment to increase biogas yield during AD OFMSW in India. OFMSW was treated at 80, 100, 120, 140 and 160 ° C at 0, 15, 30, 60 and 120 minutes. OFMSW and hydrochars were used for AD with cow residue as inoculum.

biodegradability improving and thus methane yield increasing and digestion time reducing have been the results of their study. The cumulative methane production increasing was between 3 and 32% (Dasgupta & Chandel, 2019). Another study investigated the effect of adding hydrocard and HTC temperature on methane production in AD of fish processing waste. HTC HTC temperature in the range of 200-280 ° C had significant effects on performance and methane content, but hydrocard had little effect (Pazoki & Ghasemzadeh, 2020; Heidary et al., 2017).

Therefore, the hydrothermal carbonization temperature were selected for this study to obtain HTC pretreatment effects on AD to increase biomethane yield. In this study, OFMSW was firstly hydrothermally treated, and obtained hydrochar was added to the AD of raw OFMSW for investigating the effect of the effect of HTC pretreatment on AD. This study mainly investigates the impact of hydrochar produced at different temperatures in the HTC treatment on AD of the OFMSW.

Material and Methods

Substrate and inoculum preparation

The samples of OFMSW were gathered from the waste disposal area of Arad-Kouh, located in the south of Tehran in the vicinity of Kahrizak town on the old Tehran-Qom road. They were employed as substrates in this research. After the trommel screen and manual separation of inorganic materials, the produced organic waste was selected as the OFMSW. After crushing OFMSW into 1 to 1.5 mm pieces, they were preserved in a plastic bag in a refrigerator at 4 °C until experiments were carried out. The waste materials were allowed to reach the ambient temperature before the test (Choe et al., 2021).

As the inoculum material, the activated sludge was gathered from the anaerobic sludge of the municipal wastewater treatment plant of South Tehran. Given that the gathered sludge contained solid particles of different sizes, it was filtered using a filter with hole sizes from 1 to 1.5 mm to separate its solid particles. The inoculum material was stored at 4°C and was allowed to reach the ambient temperature (Choe et al., 2021).

Hydrothermal Carbonization

The hydrochar was produced at a 3-liter reactor (working capacity of 2 liters) made of stainless steel equipped with a temperature controller and barometer. Based on the design, 200 g MSW was added to 1.8 liters of distilled water. After mixing for 10 minutes, it was isolated at the reactor. The reactor with the temperature increase rate of 10 °C per minute reached HTC process temperature (according to the design). After the reactor temperature reached the designed temperature, the HTC process was kept at this temperature as the residence time for 40 minutes. Each experiment was carried out three-time to reach sufficient accuracy. The barometer indicated a pressure of 4-10 for temperature variation. When the residence time of the reaction is over, the reactor was cooled at room temperature. After taking out the reactor's content, hydrochar and the produced liquid were separated by a filter paper with hole sizes of 6 µm. Besides, to dry the produced hydrochar, it was kept at 105 °C for 24 hours. Afterward, for preservation and further use in the AD process, all produced hydrocahrs were kept in a plastic bag at 4°C. The yield of produced hydrochar and energy are calculated according to the following equations:

$$Hydrochar\ Yield\ (\%) = \left(\frac{Hydrochar\ Dry\ Mass\ (g)}{OFMSW\ Powder\ Dry\ Mass\ (g)}\right) \times 100 \quad (1)$$

$$Energy\ Yield = hydrochar\ yield \times \left(\frac{HHV\ of\ hydrochar}{HHV\ of\ OFMSW}\right) \quad (2)$$

Anaerobic digestion

For obtaining the potential of biomethane production from OFMSW and hydrocahrs, the 118 ml glass bottles were employed as batch reactors. 20 ml microbial mixture consisting of activated sludge and 5 ml distilled water along with 0.25 g (WT% dry basis) substrate (the hydrochar produced in the HTC process at the designed temperature) was added to each reactor. The bottles caps were fastened using plastic rubber and aluminum caps. The properties of the microbial content are presented in Tables 1 and 2. Afterward, a gas consisting of 80% Nitrogen and 20% carbon dioxide was injected into these bottles, and simultaneously, to obtain an anaerobic environment, the air inside the bottles was vacuumed using another syringe. This batch system is known as the Hansen method and has been employed by various researchers as the AD batch reactor in various experiments (Hansen et al., 2004).

Sampling and gas analysis

Analysis of the produced biogas and determining the methane and carbon dioxide percentage (sum of methane, carbon dioxide, and nitrogen gases are considered as the produced biogas) were carried out employing a GC device. The device was equipped with a peak ABC software, which was employed to analyze the different gas forms resulting from sample injection. Helium gas was used as the carrier gas with a flow rate of 20 ml/min.

The temperatures of the column, the injector, and the indicator are adjusted to 50, 90, 140 °C, respectively. First, the pure gas peaks of methane, carbon dioxide, nitrogen, and air were determined for the Gas Chromatography (GC) device. Also, the gas appearance time was specified by the indicator (Ebrahimian et al., 2020).

Sampling from reactors and produced gas was carried out every three days. In sampling from discontinuous anaerobic digestion reactors, 250 µl of the produced gas sample was injected into the GC device via a vacuum locking syringe. The injection results to the GC device were saved and used to calculate the biogas percent composition and methane and carbon dioxide gas volume (Ebrahimian et al., 2020).

Analytical methods

In this study, the volatile solids (VS) and Total Solids (TS) percentage were obtained by APHA standard method to get acquainted with the feed, consisting of the OFMSW, inoculum, and many types of hydrochars (Federation & APH Association, 2005). The samples' ash content was achieved at 575 ±5 °C temperature according to ASTM D-1102 (ASTM, 2021). Elemental or terminal analysis was performed by the analyzer "Elementary Trading Shanghai Co., Ltd. China" to identify the Carbon, Nitrogen, Hydrogen, and Sulfur content. The oxygen content was also obtained as a result of the mentioned elements' decline from 100% (Volpe et al., 2018). The pH value was obtained by a testo 250 pH meter. Also, the Volatile Materials (VM) and Fixed Carbon (FC) amount was obtained according to ASTM D-3175-89 standard method (Volpe et al., 2018).

Results and Discussion

The physical and chemical properties of the feedstock and produced hydrochars

The physical and chemical properties of the inoculum and OFMSW were provided in Table 1 to get familiar with the used feedstock in the digester and HTC. As the temperature of the hydrothermal carbonization process rose, the carbon percentage in the approximate analysis increased. The carbon of hydrochar 150, 190, 230 increased by 7.5, 19.3, and 35.2%, respectively. As a result of these changes, the produced hydrochar had a higher C/N compared to the initial feed or the OFMSW. Concerning the fact that the intensity of the HTC process increased as a result of temperature enhancement, more organic material would be turned into carbon materials. These observations also took place in other studies (Basso et al., 2016; Mäkelä et al., 2015; Benavente et al., 2015).

Table 1. Characterization of OFMSW and inoculum

Parameter	OFMSW	Inoculum
TS (wt% wet basis)	25.72	3.3
VS (wt% wet basis)	20.61	1.9
VS/TS (wt% dry basis)	80.13	57.58
C (wt% dry basis)	49.22	25.74
N (wt% dry basis)	3.82	2.81
H (wt% dry basis)	6.1	3.54
S (wt% dry basis)	0.4	1.22
O (wt% dry basis)	40.46	66.69
C/N	12.88	9.16
pH	5.2	6.6
Ash content	4.98	-

The final analysis results, including the percentage of carbon, hydrogen, nitrogen, sulfur, and oxygen, were indicated in Table 2. Besides, the approximate analysis, including the percentage of the FC, the VS, and ashes for the anaerobic digester feed, such as the OFMSW and produced hydrochars in different temperatures of hydrothermal carbonization process, at 160, 190, and 230 °C, were provided there. According to the approximate analysis in Table 2, the VM declined due to the temperature increase in the HTC process. Also, the fixed carbon content was enhanced. This was due to more charring as a result of temperature enhancement in the HTC (Volpe & Fiori, 2015). According to the elemental analysis, the sulfur content was very insignificant (lower than 0.5% weight percent). As the temperature of the hydrothermal carbonization process increased, the H/C and O/C ratios were decreased due to the dehydration reaction in this process (Volpe & Fiori, 2015).

Table 2. Physical and chemical parameters of raw OFMSW and hydrochars at different temperature

	Proximate analysis (wt% dry basis)			Ultimate analysis (wt% dry basis)				
	VM	FC	Ash	C	H	N	S	O
OFMSW	71.54	24.03	4.43	49.22	6.1	3.82	0.4	40.46
Hydrchar-150	68.97	25.78	5.25	52.91	6.1	5.12	0.44	35.43
Hydrchar-190	66.01	27.02	6.97	58.74	7.08	4.24	0.46	29.48
Hydrchar-1230	61.14	30.96	7.9	66.57	6.34	3.94	0.46	22.69

Impact of hydrochar on biomethane production in AD

The related results to the produced hydrochar yield (%), heating value (MJ/kg), and energy yield (%) of the produced hydrochars in the hydrothermal carbonization process was indicated in Table 3. The hydrochar yield percentage for hydrochar 150, 190, and 230 were 61.54, 56.74, and 47.53, respectively. Also, the heating value for the organic fraction of the MSW and hydrochars 150, 190, and 230 were equal to 20.37, 24.84, and 27.64, respectively. According to the yield of the hydrochar and the heating value, and also Equation 1, the energy yield for each mentioned hydrochars was equal to 68.84, 77.4, and 72.14%, respectively.

As indicated in Table 3, the hydrochar yield declined as a result of temperature increase. Therefore, a lower extent of hydrochar would be produced during the process. The reduction percentage in the produced hydrochars was 7.8 and 22.8% at 190 and 230, compared to 150 °C temperatures, respectively. The other studies demonstrated similar results (Basso et al., 2015). These observations were due to dehydration and decarboxylation reactions more effective in higher temperatures and led to more gas production and solid material reduction (Basso et al., 2015). However, according to another study, in higher temperatures, e.g., 260 and 280 °C, the hydrochar yield was incremental due to a back-polymerization reaction from liquid to a solid phase in HTC, overcoming the amount of initial organic materials decomposition (Coronella et al., 2014). Unlike the hydrochar yield, the heating value increased by the temperature enhancement of HTC. The variation level of the heating value of produced hydrochars at 150, 190, and 230 °C to the feedstock inputted in the HTC reactor were 11.9, 36.4, and 51.8%, respectively. Concerning the carbon percentage increase, it was predicted that the heating value would rise. Some investigations on other feeds prove this issue, and the main reason was due to the enhancement of output material charring following the temperature increase in the hydrothermal carbonization process (Volpe & Fiori, 2017; Jain et al., 2016). As a result of the mentioned points and Equation 2, as the temperature of HTC increased, the energy yield enhanced at the first stage and then declined. As indicated in Table 2, the energy yield percentage for the hydrochar 190 was equal to 77.4%.

Table 3. HTC process yields and total biomethane production

	Hydrochar yield (%)	HHV (MJ/kg)	Energy yield	Total biomethane production
OFMSW	-	18.21	-	131
Hydrochar-150	61.54	20.37	68.84	178
Hydrochar-190	56.74	24.84	77.4	193
Hydrochar-1230	47.53	27.64	72.14	93

Impact of hydrochar on biomethane production in AD

Figure 1 indicated cumulative biomethane yield in each digester during 45 days. The produced biomethane content in the control sample (the mixture of OFMSWand inoculum) was 131 mL/gVS at the end of day 45. The control sample was for identifying the variation level before and after the HTC treatment. Also, the produced biomethane content in the AD samples of hydrochar 150, 190, 230 °C were 178, 193, and 93 mL/g VS, respectively. It has shown an increase for hydrochars 150 and 190 and a decrease for hydrochar 230.

According to Figure 1, the produced biomethane content was equal to 131 mL/gVS in the control sample. After the HTC process, that value increased by 35.88 and 47.33% in hydrochar 150 and 190, respectively. However, it had a decrease by 29% in hydrochar 230. The biomethane content production increase was due to the breakdown of complex organic compounds to the heavy molecular compounds in the HTC process. The compounds with heavy molecular mass included carbohydrates, proteins, and other hydrolyzed products. Accordingly, these compounds had a higher potential to turn into lighter molecular mass, and accordingly, biomethane production (Phuttaro et al., 2019). The biomethane content production decline in hydrochar 230 was due to the volatile materials decline in hydrochars from 68.97 to 61.14%. Another reason was pertinent to the production of phenol and furfural and other compounds with higher molecular mass, non-decomposable at higher temperatures in the HTC process. Phenol and furfural were recognized as a barrier against the complete reaction of the AD process. Other researchers achieved similar results in different temperature ranges (Choe et al., 2019; He et al., 2014; Aragón-Briceño et al., 2017).

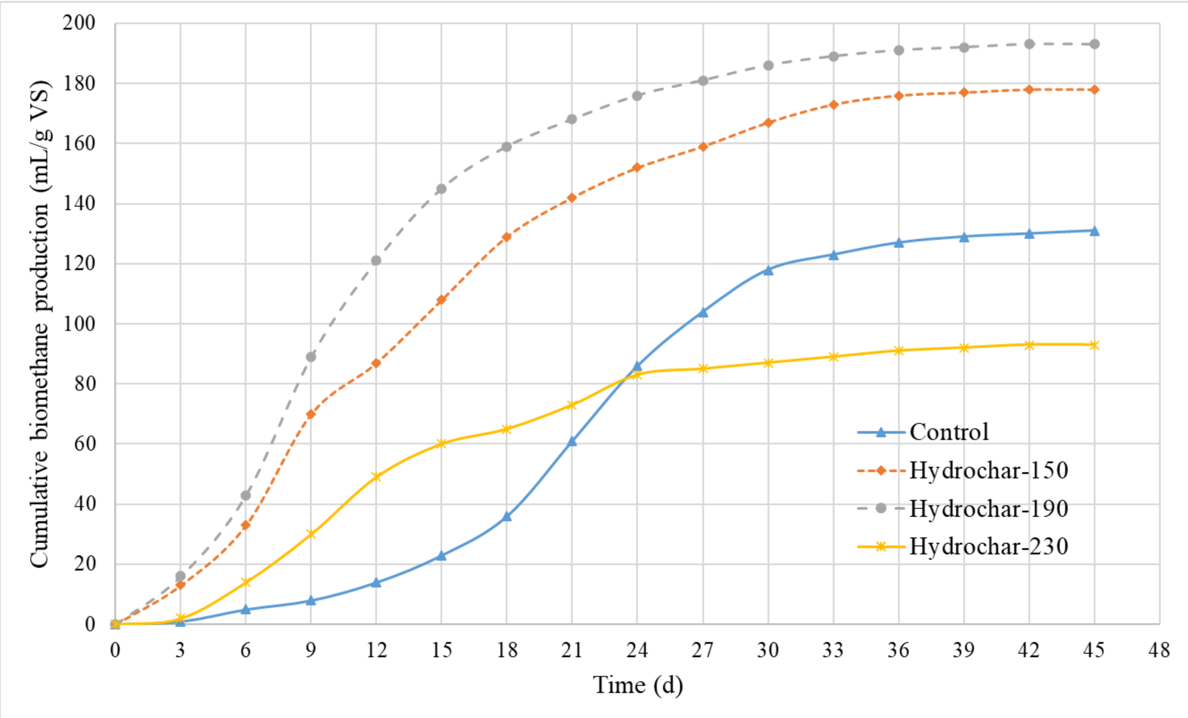

Figure 1. cumulative biomethane yield during 45 days AD of mixed OFMSW and hydrochar

Daily biomethane yield production (once every three days) was displayed in Figure 2, in which the anaerobic digester behavior is demonstrated. The maximum produced biomethane content was in digesters having hydrochar 150 and 190 on day 9, in the digester having hydrochar 230 on day 12, and in the control sample on day 22 since the production start point. The biomethane production rate was also increased compared to the control sample. In order words, the maximum point of biomethane production in Figure 2 occurred in a lower time than the control sample.

According to Figure 2, the peak of each biomethane production graph took place in a lower period. The maximum level of biomethane production in the control sample, hydrochars 150, 190, and 230, occurred on days 22, 9, 9, and 12, respectively. This issue indicates that the digesters with lower capacity (volume) will be required, and therefore, the operational costs will be declined (Rani et al., 2012).

Conclusions

Among the examined samples, the best performance was related to the produced hydrochars in 190 °C, increasing the biomethane content compared to the raw sample. In this case, the biomethane production took place at a lower time with no delay. According to the results, the pretreatment by the HTC process directly affected the biomethane production level and its time. The biomethane production depends on the condition of the hydrothermal process; according to the results, as the temperature of the hydrochar production increased, the biomethane production in the AD reactor enhanced at the first stage and quickly declined in higher temperatures. Therefore, before using the HTC pretreatment, the optimal temperature should be obtained. Regarding the time duration, concerning the breakdown in materials' hard structure in the hydrothermal process, the delay would not occur in biomethane production, and the time duration of the biomethane production decreased in all hydrochars. However, there was the possibility of producing compounds as barriers against the anaerobic digestion process completion in higher temperatures.

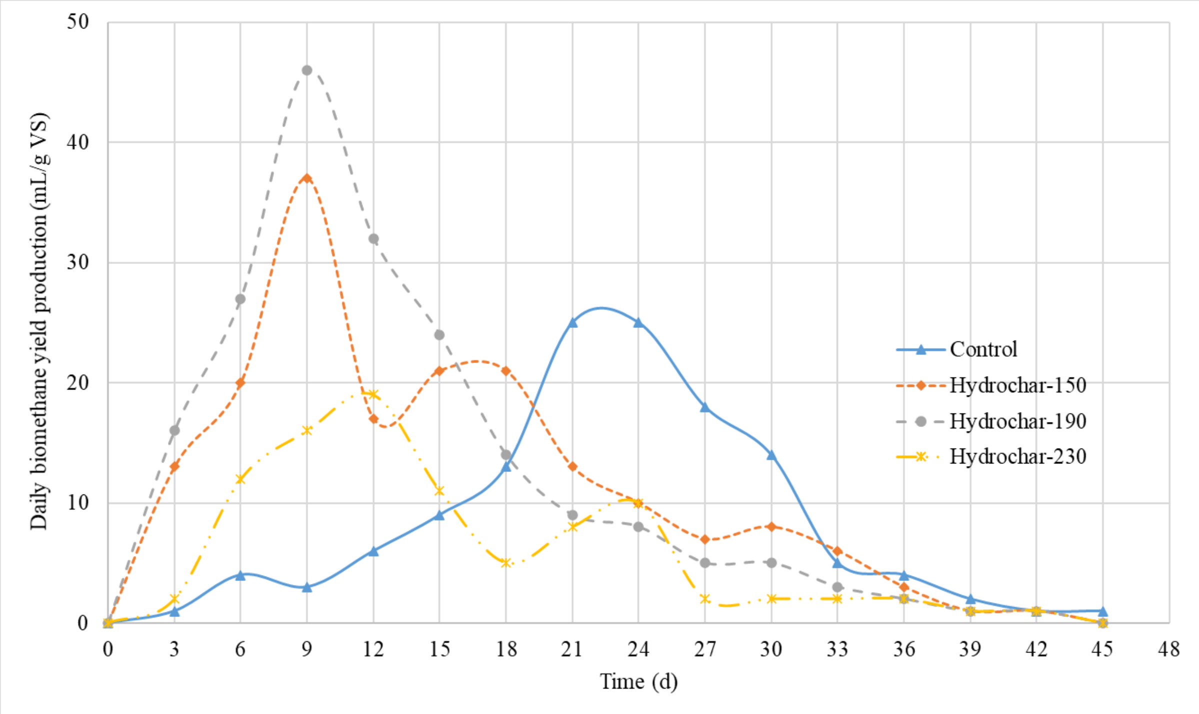

Figure 2. daily biomethane yield production in AD during 45 days

Concerning the obtained results, if used optimally, the HTC process could significantly affect the AD of the OFMSW when 1. The biomethane production increases, and 2. The delay is decreased in biomethane production. Also, it is suggested that other contributing factors, such as retention time, pH, feed concentration of hydrothermal carbonization process, and the effectiveness of these parameters on each other, could be studied by other researchers.

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