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An Experimental Study for Municipal Organic Waste and Sludge Treated by Hydrothermal Carbonization

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The use of firewood and other biomass-based fuels have generated severe environmental pollution problems due high particulate matter emissions. Additionally, developing countries face considerable challenges in aspects related to the final disposal of organic waste in sanitary landfills that are already overflowed, and that constitutes a serious problem. In the last years, the search for alternative energy sources based on organic waste valorization has gained popularity. For waste biomass conversion, Hydrothermal Carbonization (HTC) has some advantages: low process temperatures required and the ability to work with biomass of different compositions and high moisture. Two groups of urban waste were considered in this investigation: 1) organic fraction of municipal solid waste (OFMSW) and 2) digested sludge (DS) from a water treatment plant. An Experimental Design was developed to study the effect of the blend composition with different OFMSW:DS ratios, reaction time (0.5 and 1 h) and temperature (190 and 220 ºC) on the Mass Yield (MY), the Higher Heating Value (HHV), Energy Densification Ratio (EDR) and Energy Yield (EY). The response equations had an average determination coefficient (R^2) of 0.95 with an RMSE of 5.9 %. The results showed that temperature was the most significant variable on the MY (-9.8 %) and the HHV (+8.7 %). Blend 2, with a greater amount of pruning waste, had higher MY and HHV. Blend 1 had the highest percentage of food waste and sludge, and, therefore, the highest MY values. The energy yield determined for the three mixtures was about 80 %, indicating that HTC is a feasible technology for the recovery of municipal waste biomass and sludge.

1. Introduction

One-third of the world food produced for human consumption annually, about 1.3 Gt, is lost or wasted throughout the supply chain, from production to consumption (Saqib et al., 2019). The organic fraction of municipal solid waste (OFMSW) represents 30 – 40 % of total waste has high humidity content and biodegradability properties due to the high proportion of food waste, kitchen waste, and leftovers from residences, restaurants, cafes, factory canteens, and markets (Alibardi and Cossu, 2015). A considerable amount of OFMSW is incinerated or goes to landfills, which are low-cost but polluting processes. Other options like biological treatments such as composting or anaerobic digestion are considered more environmentally technologies but are often not economically viable due to lengthy process times (20 – 30 d). Sludge is the most critical by-product in wastewater treatment. Its stabilization and the water content reduction is necessary to inhibit the generation of bad odors and minimize the volume occupied in its final disposal (Tsapekos et al., 2019). The sewage sludge contains large amounts of organic matter, micro-organisms, heavy metals, and refractory pollutants, which raises severe environmental damage. After mechanical drainage, the sludge moisture content remains at 80 % (Wang et al., 2019). However, sludge can be used as an energy source during the anaerobic digestion stage in which biogas is a by-product of the process. Biogas can be fed to a cogeneration machine to produce electricity and heat energy (Pfluger et al., 2019). In Chile, the constant increase in OFMSW is a challenge in terms of management and valorization. The waste

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generation in 2015 – 2017 had an increase of 8 %, going from 21.2 to 23 Mt. In 2017, 97.3 % of the total waste generated corresponded to non-hazardous waste, of which 1.6 % corresponds to sludge, 35.3 % to municipal waste and 60.4 % of industrial origin (Donoso-Bravo et al., 2019).

The valorization of these organic wastes for bioenergy generation is a challenge today. Still, it is an urgent need in developing countries, where the current situation of the final disposal of household waste indicates that landfills are close to reaching their useful life. The development of large-scale and efficient industrial processes is limited by the main composition of OFMSW and sludge, especially the high water content (Pham et al., 2014). One of the alternatives that have been successfully evaluated in waste biomass with high-moistures levels and that does not require previous drying processes is Hydrothermal Carbonization (HTC). Indeed, this process occurs at low temperatures (180 - 250 ºC) and reaction times of a few hours (Heidari et al., 2018). Therefore, there is an opportunity to implement circular economy systems through the use of valorization technologies to recover the energy content of different agricultural and urban wastes and generate a biofuel with better energy and storage characteristics. HTC has been studied in some agroforestry biomass such as sawdust (Zhang et al., 2017), olive (Mäkelä et al., 2016), among others; and in non-lignocellulosic biomass as food waste (Tradler et al., 2018) and sewage sludge (Danso-Boateng et al. 2015). Likewise, some studies have evaluated the behavior of mixtures between food waste and sewage sludge (e. g. Zheng et al., 2019). However, the influence of operational conditions and the amount of sewage sludge used in this blends has not been determined, which directly affects the possibility of increasing the HHV and the energy yield, as recent studies have shown (Vallejo et al., 2020b). Zheng et al. (2019) reported an average close to 40 % in the ash content for a temperature range of 180 – 280 ºC and Wilk et al. (2019) obtained 55 % of ash for hydrochar from sludge, in both cases the value that is too high for combustion pellets. Consequently, the main goal of this work was to study the optimal condition of the HTC process for the energy valorization of organic wastes analyzing the influence of time, temperature, and OFMSW/sludge ratio in blends on the performance and characteristics of the final product.

2. Materials and methods

2.1 Samples

The organic waste was collected in a sanitary landfill and the digested sludge was recovered from a Wastewater Treatment Plant, both located in the southwestern area of the Metropolitan Region of Santiago, Chile. The samples were kept at 4 ºC until its use. The moisture content was determined by the gravimetric method, and Higher Heating Value (HHV) was carried out in a Parr 6200 calorimeter. Finally, the raw biomass was analyzed for ultimate composition (dry basis). The raw samples characteristics are shown in Table 1.

Table 1: Raw sample characterization.

Raw biomass	C (%)	H (%)	N (%)	O (%)	Ash (%)	Moisture (%)	HHV (MJ/kg)
Leftover food	43.74±0.08	6.47±0.05	2.31±0.02	40.53±1.02	6.94±1.02	52.08±0.29	18.25±0.10
Fruits and vegetables	42.70±0.27	5.72±0.05	1.71±0.09	35.63±0.48	14.24±0.48	83.23±0.36	17.45±0.06
Garden waste	36.67±0.07	4.76±0.04	1.22±0.02	30.52±0.93	26.84±0.93	52.93±0.16	15.13±0.08
Sludge	35.26±0.06	4.96±0.05	4.52±0.07	17.19±0.13	38.07±0.13	75.42±0.23	16.06±0.04

As shown in Table 1, leftover food, fruits and vegetable fraction had the highest carbon content. On the other hand, sludge and garden waste showed the lowest carbon and oxygen content, which generated a lower calorific value. Although the composition and HHV of food waste, fruits and vegetables vary according to the season and geography, similar values have been obtained in the literature. (e. g. (Chen et al., 2018) HHV of potato: 19.08 MJ/kg). Finally, the HHV of the sludge was within the range reported by previous studies, with values of 16.5 and 16.33 MJ/kg for anaerobic and primary sludge, respectively (Tasca et al., 2019). The analysis presented in Table 1 was performed to obtain three raw mix samples that represent the different areas of the country according to their geographical characteristics. The Northern zone (Blend 1), Central zone (Blend 2) and Southern zone (Blend 3) were considered. The specific composition in a dry basis of organic waste is indicated in Table 2. The food waste fraction was assumed as the sum of leftover food, fruits and vegetables.

2.2 Experimental Design

The HTC experiments were carried out in high pressure and temperature reactor, model HiPR-SF5L with a capacity of 5 L. The reactor was loaded with 1,000 g of the OFMSW-sludge mixture in each run, with the composition indicated in Table 2. All runs were made with biomass:water ratio of 1:10. The variables considered in the Experimental Design (2^3) were the temperature and the time reaction process and the relation between the sludge and the OFMSW. The effects were analyzed in the solid product (hydrochar) according to the mass yield (MY), HHV, energy yield (EY), and ash content (ASH). The levels of the factors used are shown in table 3. The Normal Probability Graph was used to obtain the factor significances.

Table 2: Composition and characterization of raw sample blends

Blend	Food waste (%)	Garden waste (%)	Sludge (%)	HHV (MJ/kg)	Ash (%)	OFMSW: Sludge ratio
Blend 1	54.73	3.29	41.98	17.42	22.36±0.03	1.382
Blend 2	45.28	29.81	24.91	16.86	25.77±0.04	3.014
Blend 3	50.70	14.59	34.71	16.54	23.43±0.03	1.881

Table 3: Factors and levels in experimental design

Factors	Units	Levels	
Temperature	ºC	-1: 190	+1: 220
Time	min	-1: 30	+1: 60
OFMSW/sludge ratio	g / g	-1: 1.38	+1: 3.01

In each experiment, MY, energy densification ratio (EDR), and EY were determined, as shown in Eq(1) to Eq(3).

$$MY=\frac{Dry\ hydrochar\ mass}{Dry\ raw\ sample\ mass} \tag{1}$$

$$EDR=\frac{Hydrochar\ HHV}{Raw\ sample/blend\ HHV} \tag{2}$$

$$EY=MY \cdot EDR \tag{3}$$

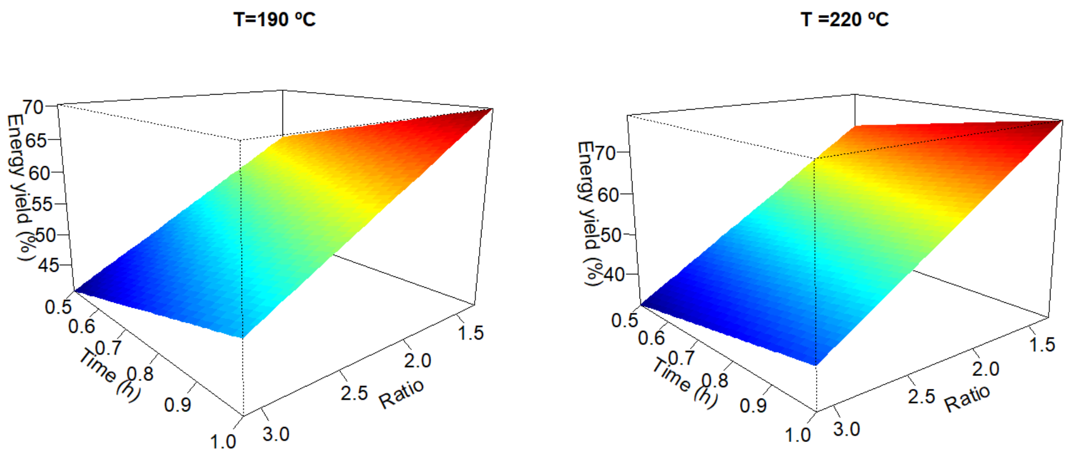
3. Results and discussion

The HTC runs for the blends showed MY between 30 % (Blend 3) and 57 % (Blend 1), as reported in Table4. The low values of the mass yield obtained are explained by the transfer of the proteins and lipids content, to the liquid phase at temperatures below 180 ºC throughout the process (Vallejo et al., 2020a). Results for MY at the same time reaction at 220 °C was lower or similar than at 190 °C. The difference in residence time was not significant, as indicated in Table 4, similar to what was reported in previous studies (Vallejo et al., 2019).

Table 4: Experimental results for HTC runs of blends

Blends	Blend 1				Blend 2				Blend 3			
T (ºC)	190		220		190		220		190		220	
time (h)	0.5	1.0	0.5	1.0	0.5	1.0	0.5	1.0	0.5	1.0	0.5	1.0
MY	53.5	56.9	57.0	56.8	39.4	41.4	32.1	30.7	30.5	31.8	31.9	30.6
HHV	18.1	22.3	20.7	24.0	18.4	19.5	19.6	22.4	17.4	18.9	18.6	19.7
EDR	1.04	1.28	1.19	1.38	1.09	1.16	1.16	1.33	1.05	1.15	1.13	1.19
EY	55.5	72.8	67.6	78.4	43.0	48.0	37.3	40.7	32.1	36.4	35.9	36.5
ASH	15.4	16.3	25.0	19.6	27.2	24.0	24.6	24.8	28.5	20.7	30.1	22.9

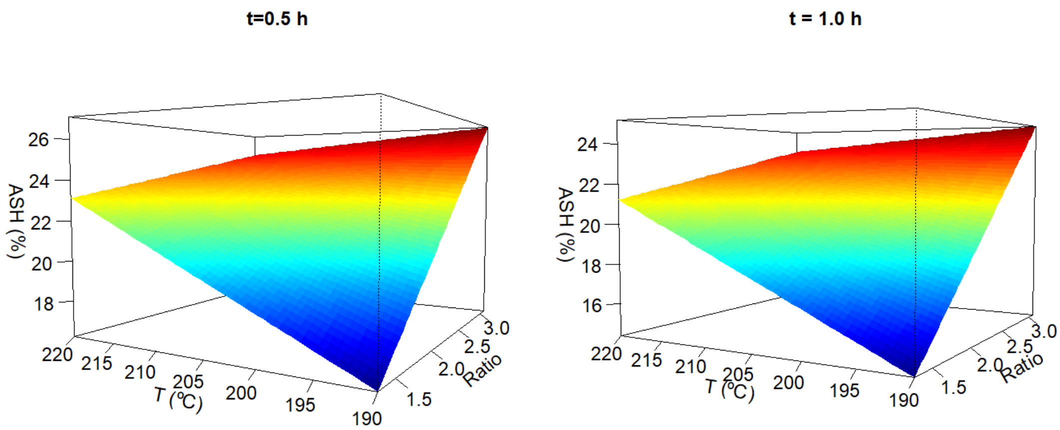
The EDR for Blends 1 and 2 at 220 ºC and 1 h was 1.38 and 1.33, with HHV values of 24.0 and 22.4 MJ / kg, respectively. These results were more significant than those reported by Reza et al. (2016), among others, that achieve HHV greater than 30 MJ/kg with temperatures above 250 ºC, but with ash contents of over 40 % (Reza et al., 2016). For the mixtures in this study, the percentage of ash was less than 30 %, and an average of 22.1 %. For HTC runs at 190 ºC, Blends 1 and 2 showed values less than 20 % of this variable. Some studies have been developed to evaluate the characteristics of the hydrochar obtained from OFMSW and sludge. Berge et al. (2011) reported an HHV of 20 MJ/kg, with EDR of 25 % and EY of 77 %. The operational conditions were extremely high at 250 ºC and 20 h compared to this study. Subsequent studies showed that the change in biomass subjected to subcritical conditions occurs in the first hours (Peterson et al., 2008), and it depends on the operational conditions and the initial composition of the biomass (Vallejo et al., 2020b). The increase in temperature and time leads to an increase in the ash content, showing an indirect effect in most of the experiments in severe conditions. Berge et al. (2011) reported a rise in ashes from 26 % to 45 % at the end of the process. Danso-Boateng et al. (2015) obtained hydrochar with an average of ash greater than 55 %, in a range of 220 ºC to 260 ºC. The existence of optimal conditions that allow a decrease in this value must be evaluated for each biomass, which directly affects the quality of the hydrochar as a biofuel (Li et al., 2018). Few studies have found general equations for predicting mass yield and calorific value for hydrochar (Vallejo et al., 2020b). However, its application in mixtures is a more significant challenge due to the presence of synergistic effects (Shen et al., 2017). The statistical analysis was developed, and the response equations were obtained with an average determination coefficient (R^2) of 0.95 and an RMSE of 5.9 %, values that were similar to those reported in Multiple Linear Regression equations (Vallejo et al., 2020b). The adequate fit indicates that the chosen variables and their ranges allow explaining the variation in the results. The response surfaces for EY show that mixtures with a lower value in the OFMSW / sludge ratio are desirable to achieve better energy retention values in the hydrochar. The increase in temperature generated a positive effect, increasing the maximum value from 70 % to 75 %, as shown in Figure 1.



$$EY = 55.4 + 4.6\,X_t - 13.2\,X_R - 3.9\,X_T\,X_R$$

Figure 1. Response surface for Energy Yield (a) 190 ºC and (b) 220 ºC.

Ash content is an essential variable since its value indicates the level of compliance with regulations in different countries. The mixtures with a lower value in the OFMSW / sludge ratio generated hydrochar with lower ash content due to the loss of mass (sugars, lipids, and carbohydrates) in the OFMSW. The response surfaces showed in Figure 2 indicate that the operating point that allowed the ash to decrease was at 190 ºC with times of 0.5 and 1.0 h. The recommended composition was Blend 1, and considering the ratio in Blend 2, the ash content would not increase significantly.



$$ASH = 22.1 + 1.4\,X_T - 0.95\,X_t - 3.1\,X_R - 1.9X_T\,X_R$$

Figure 2. Response surface for the hydrochar ash content (a) 0.5 h and (b) 1 h.

Figure 3 indicates the response surface for EDR. It can be seen that the most significant variables for this response are temperature and time, as noted in previous studies (Baratieri et al., 2015), an increase in time and temperature produce more severe conditions in the process (Vallejo et al., 2020b) increasing the carbon content and calorific value. Similar to that indicated for the other answers, with a lower value in the OFSMW / sludge ratio, higher energy densification is achieved. In this case, the variable with the most significant effect was time, increasing from 0.5 to 1 h increases the EDR value by 10 %. For 220 ºC and 1 h, the HHV increase was close to 40 %.

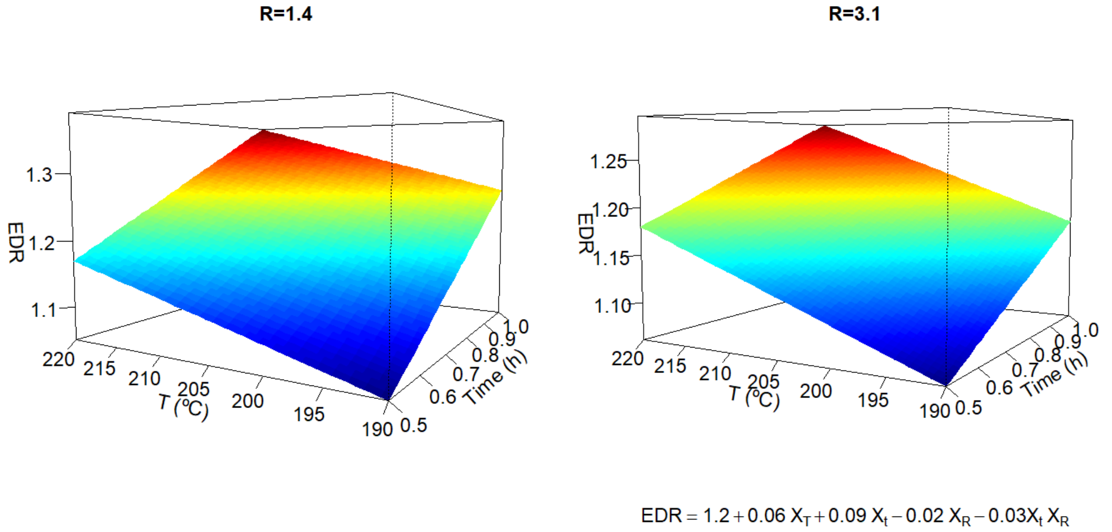


Figure 3. Response surface for Energy Densification Ratio (a) R=1.4 and (b) R=3.1.

4. Conclusions

The evaluation of several variables in the HTC process by Experimental Design allowed predicting the values of MY, HHV, EDR, EY, and ASH for several biomass blends. The analysis performed indicates that the temperature was the most significant variable on the MY and the HHV. The most significant variables for EDR are temperature and time, where an increase in time and temperature increases the calorific value. A lower value in the OFSMW / sludge ratio allows achieving the higher Energy Densification Ratio. For this variable, time changing from 0.5 to 1 h increases the value by 10 %.

The response surfaces for EY indicate that mixtures with a lower value in the OFMSW / sludge ratio are desirable to achieve better energy retention values in the hydrochar. The mixtures with a lower value in the OFMSW / sludge ratio generated hydrochar with lower ash content. The response surfaces indicate that the operating point that allowed the ash to decrease was at 190 ºC with times of 0.5 and 1.0 h, and the recommended composition was Blend 1.

HTC is a feasible technology for the recovery of municipal waste biomass and sludge. Further studies must be carried out to optimize the blend composition, considering country raw biomass availability, economic aspects and environmental impacts.

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