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Valorization of Municipal Solid Waste using Hydrothermal Carbonization and Gasification: A review

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In developing countries, there is a relevant problem related to air pollution caused by the indiscriminate use of residual biomass and the disposal of Municipal Solid Waste. Currently, various technologies (biological, thermal, thermochemical, among others) are available to valorize them. This review shows a current summary of two technologies: hydrothermal carbonization and gasification. The first one has been considered as a Waste to Energy technology capable of providing a solid, that can be used as a fuel, with higher calorific value and lower moisture and ash content than raw residual biomass. On the other hand, gasification generates syngas used as fuel or in the generation of electricity. During the last decade, most of the studies focused on hydrothermal carbonization of Municipal Solid Waste in contrast to gasification. However, the integration of those technologies has not even had the same interest. This study analyzed in-depth the product characteristics and the associated costs for both processes, including from transportation to obtaining the final product. Both promising Waste to Energy alternatives can result in Municipal Solid Waste disposal cost savings and environmental impact reductions. Nevertheless, research combining technologies must be enhanced in order to develop sustainable systems.

1. Introduction

The generation of Municipal Solid Waste (MSW) is currently considered one of the significant environmental challenges facing our society, especially in developing countries, due to the lack of technological development (Kumar and Samadder, 2017). MSW is usually disposed of in landfills, which regular operation requires large spaces, and the problems caused by this treatment have increased over the years, such as bad odors and filtered liquids that can compromise the aquifer. This situation has generated more demanding and stringent policies (Moya et al., 2017). However, the heterogeneous characteristics of the MSW, together with the large volumes issued daily, generate the possibility of establishing large-scale valorization processes. Although incineration has been used in recent years due to its high efficiency, energy requirements and the need to avoid greenhouse gases have made it possible for new technologies to be considered for the sustainable elimination of MSW. In the last 5 y, an average of 1.3×10^9 t/y of MSW has been generated worldwide, with an expected increase to 2.2×10^9 t/y in 2025 (Gutiérrez Ortiz et al., 2019). Usually, the organic fraction is between 50 and 65 %, and it is feasible to valorize with hydrothermal carbonization (HTC) or gasification. It should be noted that waste materials are low-cost resources, with the most significant proportion of the value comes from transportation (Thompson et al., 2019). Indeed, the waste flow that is currently observed and the opportunity for its recovery can be seen in Figure 1. In summary, the biomass conversion techniques goal is to increase the amount of carbon existing in it, since the energy contained in the carbon-oxygen and carbon-hydrogen bonds is less than in the carbon-carbon bonds (Wei et al., 2017a). Available thermochemical processes include gasification and HTC, which have been studied as alternatives for the energetic valorization of MSW and obtain biofuels. In this way, the MSW is considered as a viable renewable resource for the use of environmentally friendly energy. This review presents the current state of the art of MSW valorization using

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HTC and gasification. An analysis of the latest results available in the literature was carried out, with emphasis on the operational variables of the process and its effect on the product obtained.

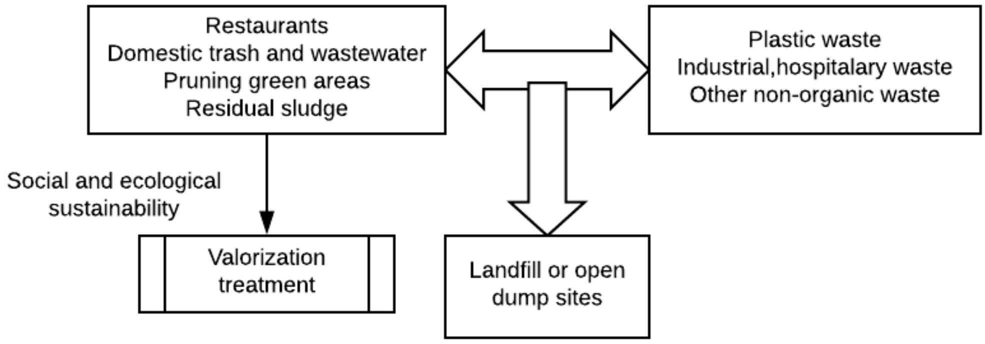

Figure 1. MSW recovery opportunity by the thermochemical valorization

2. MSW valorization by hydrothermal carbonization

One of the main advantages of HTC is that it does not require removing moisture from the raw material to be processed. Since the action of water at high pressures (around 20 MPa) and temperatures between 180 and 300 ºC is to release the hydrogen and oxygen atoms from the carbon molecules. Indeed, this process could improve the fuel quality of MSW in terms of water content reduction, chlorine removal, and energy densification (Lin et al., 2020). As a result, a solid carbon enriched, a highly concentrated aqueous solution and a mixture of gases are obtained, with carbon dioxide being the majority substance. The dehydration and decarboxylation reactions reduce the mass of the product concerning the raw material used (Vallejo et al., 2020). The HTC process alters the ash content and the calorific value of the processed material, highly relevant variables for hydrochar energy use, and complying with the regulations of any nation. Table 1 shows the characteristics of hydrochars produced from municipal solid waste (H-MSW) studied and reported. The studies shown in Table 1 reported many differences in the ash content, which depends on the original composition of MSW.

Table 1 Characteristics of H-MSW at different conditions

Reference	Ultimate analyses (% db)				Ash	HHV	Temperature	Time
	C	H	N	O	(% db)	(kJ/kg)	(ºC)	(min)
(Peng et al., 2016)	49.19	6.36	2.06	42.06	1.61	NR	200	30
(Jin et al., 2013)	56.85	7.33	0.97	32.59	1.09	NR	190	30
(Berge et al., 2011)	33.50	2.70	0.63	14.20	46.00	20.00	250	1,200
(Reza et al., 2016)	35.50	4.57	1.50	34.60	26.40	23.60	200	30
	45.40	5.01	1.50	35.10	24.10	22.10	200	120
	39.90	4.76	1.90	25.50	36.60	28.40	250	30
	42.60	4.38	2.20	19.00	39.10	31.00	250	120
	42.50	4.54	2.30	15.70	46.10	38.40	300	30
	43.90	4.00	2.30	15.70	39.50	34.70	300	120
(Maqhuzu et al., 2019)	58.60	5.96	0.93	16.41	17.67	22.47	NR	60
(Puccini et al., 2017)	23.63	1.07	2.60	35.04	37.66	NR	NR	NR
(Triyono et al., 2019)	60.87	8.35	0.79	29.88	5.99	30.17	150	30
	63.81	8.81	0.72	26.55	6.15	30.77	175	30
	66.55	8.78	0.70	23.87	5.45	33.01	200	30
	70.54	8.97	0.60	19.81	5.33	36.69	225	30
(Lucian et al., 2018)	67.10	6.60	4.30	16.20	5.80	29.40	220	180
	71.60	7.20	4.10	12.40	4.70	31.50	240	180
	73.50	7.80	4.00	11.80	2.90	31.90	260	180
	73.70	7.00	4.00	11.00	4.30	32.50	280	180
(Wei et al., 2017a)	47.36	8.56	1.78	23.01	18.91	NR	NR	NR

NR: Not reported. db: Dry basis

Substances of inorganic origin such as metals and glass are not altered in HTC. They are retained in the hydrochar, resulting in high percentages of ash, which limits the energy recovery of MSW by HTC. In those cases, in which this property exceeds the permitted values for energy use, the hydrochar can be used as an adsorbent material. For example, H-MSW was used as activated carbon, improving the surface properties by KOH chemical activation, being a promising application in the future (Puccini et al., 2017).

There are some studies analyzing feed material of typical individual components of MSW like food waste (Akarsu et al., 2019), waste textile (Lin et al., 2016a), waste paper (Lin et al., 2017), among others. The study by Berge et al. (2011) was one of the first to show the usefulness of HTC for the energy recovery of MSW. A synthetic blend of MSW with paper, glass, plastic, food waste, and metal (aluminum) was used. A 64 % reduction of volatile carbon in the hydrochar was obtained, which is relevant for use as a low emission fuel for atmospheric pollutants.

The hydrochar combustion has been another focus of research, specifically in the measurement of substances during this process. Recently, lower emissions of polycyclic aromatic hydrocarbons (PAHs) were obtained by combusting treated H-MSW at 200 ºC for 30 min, compared to the same residue without this heat treatment (Peng et al., 2016). It is associated with the migration of some metals into the liquid medium, which means they do not interfere with the hydrochar combustion, achieving excellent results in this area. Even a mixture of hydrochar obtained with coal further reduces emissions of these atmospheric pollutants. It is relevant to mention that the hydrochar and coal blend improved the emissions of the burned coal individually. It indicates that the energy recovery of MSW should not only be limited to HTC.

An interesting study was carried out by analyzing the implementation of the H-MSW treatment for Zimbabwe (Maqhuzu et al., 2019). An energy production plant that uses H-MSW in the combustion process generates positive results. The replacement of traditional MSW treatment alternatives for storage, incineration, and associated transport processes makes reductions in CH_4, CO_2, and sulfur oxides (SO_x). One of the considerations made for these results is the recirculation of process water or a suitable treatment in its absence to reduce environmental impacts. On the other hand, the presence of toxic substances such as trace metals, which must be removed, either from MSW or H-MSW to avoid toxic emissions of these compounds into the air, was not considered in this study.

3. MSW valorization by gasification

The gasification process converts a relatively dry solid biomass mainly into two products: syngas and char. In some cases, e.g., updraft gasification, a non-negligible amount of aqueous organic liquid can be produced (Pecchi and Baratieri, 2019). The process occurs through a sequence of thermo-chemical reactions under an oxygen-deficient environment (Sunil et al., 2019). Syngas from biomass gasification is widely discussed as an intermediate feedstock to produce electricity and second-generation biofuels.

Currently, there is an increasing interest in its use for bioenergy production with carbon capture and storage (Tu et al., 2019). This process has been improved by adding pure O_2 rather than air, when high ash content of waste is present, reaching higher temperature (> 1,600 ºC) and producing a vitrified and inert granular material (Borgogna et al., 2019). Other authors suggest gasification at low temperature (800 ºC) to feasible the syngas production for fermentation and biochemical added-value compounds generation (Pinto et al. 2019). The tar formed during gasification is one of the significant issues, catalytic cracking is recognized as the most efficient method to diminish the tar formation in the gas mixture and can operate at relatively lower temperatures and generate high tar removal efficiency (Li et al., 2008). By using a catalyst, it lowers the gasification temperature, improves the steam reform, and water-gas exchange reactions to produce hydrogen-rich gas and more product gas (He et al., 2009). Some studies about MSW gasification, as shown in Table 2, reported the influence of temperature, moisture content, the presence of catalyst and steam ratio on gas yield, syngas composition, and carbon conversion efficiency (CCE). The reactions in this process are endothermic, the reactor temperature is the most critical operating variable for MSW gasification (Luo et al., 2012). The gasification of organic fraction of MSW and MWS/coal blend has been reported recently (Tokmurzin et al., 2019). CH_4 between 35 – 37 % vol was obtained at 800 ºC when 0.5/0.5 mix was used. This yield is higher compared to the values in Table 2. A similar approach needs to be addressed in future research. To the best of the authors' knowledge, there are only three research related to H-MSW gasification. Lin et al. (2016b) studied the effect of HTC temperature (210, 230, 250, and 280 °C) and residence times (30, 60, and 90 min) on the H-MSW combustion, pyrolysis and char CO_2-gasification characteristics by the thermogravimetric analyzer. The findings suggested that combining hydrothermal carbonization treatment with the subsequent thermochemical process could be a soundly positive way for both energy generation and waste remediation. Wei et al. (2017b) studied the impact of gasification temperature, a blended ratio on co-gasification reactivity, and synergy of Shenfu bituminous coal (SF) and H-MSW.

Table 2. Temperature effect on product results

Reference	Gas composition (mol % db)						CCE	Gas yield	Temperature
	H_2	CO	CO_2	CH_4	C_2H_4	C_2H_6	(%)	($N\cdot m^3$/kg)	(°C)
(Luo et al., 2012)	34.01	11.34	38.25	10.30	3.31	2.79	65.60	0.88	700
	40.47	13.26	36.76	5.89	2.02	1.6	62.03	0.95	750
	46.54	14.65	32.61	2.87	2.85	0.48	75.16	1.28	800
	51.16	15.66	28.23	2.14	1.86	0.95	70.50	1.32	850
	54.22	22.72	20.61	1.33	0.13	0.99	84.87	1.75	900
(He et al., 2009)	27.01	9.34	35.25	20.30	6.31	1.79	62.05	0.74	700
	34.70	11.16	30.86	17.89	4.02	1.37	62.13	0.85	750
	40.64	12.65	26.61	15.57	3.85	0.68	64.74	0.98	800
	45.60	13.46	24.23	12.24	3.60	0.87	68.18	1.12	850
	48.63	14.85	23.59	9.62	2.38	0.93	72.37	1.28	900
	53.29	16.92	22.05	5.76	1.01	0.97	74.51	1.48	950
(Luo et al., 2012)	31.03	27.27	28.20	12.25				0.54	900 *
(Guan et al., 2009)	50.20	18.80	20.60	6.30				1.47	800 **

NR: Not reported. * Catalyst: NiO/Ɣ-Al_2O_3 ** Catalyst: Calcined dolomite

The results showed that higher char gasification reactivity occurred at higher HTC char proportion and gasification temperature. The main synergy behaviour on co-gasification reactivity was performed as a synergistic effect. Recently, (Lin et al., 2020) focused on the gasification of real H-MSW, under various temperatures (600–1,000 °C) and atmospheres conditions (Air, CO_2/O_2, and steam/O_2 fractions). The results showed that the syngas quality was improved whereas the tar yield was generally reduced with the reaction temperature, independent on the atmosphere. These findings suggested that the gasification, coupled with HTC, was a practical approach to produce hydrogen-rich gas from MSW.

4. Profitable and sustainable on an industrial scale

Studies on the economic evaluation to determine if the process is feasible in MSW processing has been carried out in the last decade. Yassin et al. (2009) studied the technical and economic performance of the fluidized bed combustion and gasification processes for obtaining electrical energy from MSW. In economic terms, a decrease of 7 % was reported between capital costs of 449 – 576 €/t for the gasification process, and 481 – 603 €/t for waste combustion. It was concluded that fluidized bed gasification with a combined cycle gas turbine (CCGT) was the optimal treatment option in terms of cost and efficiency, especially for electricity generation in plants with a capacity higher than 50,000 t/y in the United Kingdom. Another relevant aspect is to identify the costs concerning the complete system that includes gasification, furnace, and construction expenses. Mohammed et al. (2011) estimated the production of H_2 gas from fruit residues (empty fruit cluster, EFB) using a fluidized bed gasifier and air as gasifying agents, with a total cost of 1,941 €/t. On the other hand, a study by (Inayat et al., 2011) designed an integrated heat flow diagram for the production of H_2 gas from palm oil residues, MSW fraction, using a fluidized bed gasifier and steam as a gasifying agent with *in situ* CO_2 capture. The minimum cost of H_2 production was calculated at 1,757 €/t. On the other hand, conventional incineration has been compared to a plasma gasification process to produce electrical energy from MSW. An exhaustive economic analysis was carried out for the implementation of this plant in Portugal. From this study, it was found that selling the energy produced by the gasification plant, as well as one of its by-products (vitrified slag), generated revenues of more than 113 €/t of MSW treated, reducing the cost of final treatment from € 23 to 74 €/t. A net present value (NPV) of approximately € 20 x 10^6 and a recovery period (PBP) of 18 y were achieved (Ramos et al., 2020). A capital return period similar to that reported in pyrolysis recovery studies, which was estimated at 17 y for a 50 t/h MSW plant (Gutiérrez Ortiz et al., 2019). The main cost of treating directly MSW was the removal of moisture to which it must previously be subjected (Gai et al., 2016). This reason considers an extra cost of treating organic waste through HTC. This value has been estimated at around 150 – 200 €/t (Lucian and Fiori, 2017). In the case of using the HTC - gasification system, a study by Steurer and Ardissone (2015) determined a return of an investment horizon of 15 y, for 4,000 t/y treatment.

5. Conclusions

The literature review related to MSW valorization by hydrothermal carbonization and gasification identifies different MSW composition, process parameters, and the variation in the final product properties. All reports about HTC showed hydrochar composition dependence on the type of raw material used and the operational parameters used. In the case of MSW and H-MSW gasification, fewer studies were found that will provide detailed data on the composition of the syngas produced. Some studies showed graphs of the effect on the syngas composition obtained by varying some parameters. Considering that the process has several variables to control, and additional possible combinations, the number of studies is expected to increase. The economic evaluations of gasification and HTC at the industrial scale indicate about 18 y for the recovery investment. Besides, both technologies contributing to energy saving by the generation of energy or electricity. These promising Waste to Energy alternatives can result in MSW disposal cost savings and environmental impact reductions.

Regarding future work, research on by-products is required. In the case of hydrothermal carbonization, the gaseous and liquid fractions resulting from the process should be assessed. In the case of gasification, a more significant foray into catalysts with higher activity and stability is necessary. Besides, the tar that is generated as a by-product can be converted into valuable synthesis gas through tar reforming. The studies using data processing methods increase is expected to optimize and predict product characteristics. Since, in both technologies, it depends on specific parameters that can be controlled and modeled. Finally, it is expected that further studies assess the integration of both techniques, seeking sustainable development, and optimize the raw material composition for its efficient transformation.

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