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Biomass Wastes Hydrothermal Carbonization: A Mini-Review on Hydrochar Properties and Combustion Performance

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Hydrothermal carbonization (HTC) is one of the thermochemical processes commonly used to convert raw biomass into hydrochar, a valuable solid carbonaceous product. Waste biomasses such as forestry residues, agricultural wastes and sewage sludges have drawn considerable interests as hydrochar feedstocks. These materials are regarded as economically sustainable material and their usage provides an effective solution to waste management concerns. The carbonization procedure is accomplished using water as a reaction medium at temperatures between 150 and 300 °C and pressure up to 20 bar. Typically, the biomass transformation process is completed using a carbonization time ranging from several minutes to several hours. In addition to the HTC temperature and carbonization time, the impact of biomass feedstocks utilized on the hydrochar properties and fuel/combustion performance dictated their potential applications as solid fuel and carbon materials. This paper provides a brief commentary on recent studies involving the main biomass wastes transformation using the HTC approach. The impact of raw biomass quality on the resulting hydrochar properties is highlighted and combustion behaviour/performance are compared and discussed.

1. Introduction

The global demand for water, food, and energy supplies rises in tandem with increases in the world's population and urbanization process. In this regard, biomass wastes such as forestry residues, agricultural wastes and biosolids from industrial effluent and sewage treatment plants offers an enormous potential for energy production. These wastes can be cleanly and efficiently valorized via an emerging thermochemical conversion process known as hydrothermal carbonization (HTC). The selection of feedstock for hydrochar production for the purpose of energy application via HTC should be made after thorough considerations on their local availability, feedstock characteristics, desired qualities of the hydrochar, requirements of the intended application and effects on the environment (Demirbas, 2004). Table 1 shows classifications of biomass wastes and compares their advantages and disadvantages as feedstocks for the HTC. Organic biomass constituents are transformed into a useful carbon-rich solid substance upon exposure to high pressure and temperature in an aqueous environment. In this regard, the HTC supports multiple reactions involving hydrolysis, dehydration and decarboxylation that initially degrade the biomass into reactive intermediates before eventually polymerized into hydrochar as a result of condensation and aromatization reactions. The exact hydrochar transformation reaction mechanisms may vary with different biomass waste feedstock used which signify the importance of the feedstock materials.

The solid product of the HTC process, known as hydrochar, is distinguished by its carbon-rich makeup mainly due to the liberation of hydroxyl and carboxylic groups. By having lower H and O atoms concentrations than the starting feedstock, the hydrochar exhibits higher energy content and porosity (Wilk et al., 2020). For solid biofuel

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applications, the hydrochar offers several advantages over the original biomass waste feedstock such as higher energy density, lower moisture content and more importantly it is stable under long storage and transportable. Hydrochar has also attracted interests beyond energy production, particularly for soil fertility enhancement (Venkatesh et al., 2022), adsorbent for wastewater treatment (Yao et al., 2011) and a precursor for the production of activated carbon (Mohan et al., 2014). Currently, there is a lack of research addressing the fuel properties and energetic performance of hydrochars derived from various sources. Hence, this paper aims to assess and compare the fuel properties of hydrochars with respect to their raw feedstocks. This comparative analysis will provide valuable insights in understanding the changes in properties after HTC for potential applications in energy generation or other relevant fields.

Table 1: Comparison of various hydrochar feedstocks.

Type of feedstocks	Advantages	Disadvantages	Reference(s)
Forestry residues (chips, sawdust and pallets of various woody biomass species)	Sustainable, low carbon footprint, high volatile and carbon content, low ash content, homogenenous	quality dependent on wood species and processing conditions	(Demirbas, 2004) (Wilk et al., 2021)
Agricultural residues (annual and perennial crops - sugar cane bagasse, oil palm wastes, straws, cornstalks)	Sustainable waste materials, high volatile and carbon content, homogeneous, eliminate open field burning and land dumping	Ash content may vary can influence soil pH, high moisture, Competition for soil organic matter leads to reducing the availability of nutrients for plants	(Venkatesh et al., 2022) (Wang et al., 2020) (Parshetti et al., 2013)
Biosolids (industrial, sewage and pulp and paper sludge)	Waste management solution, recovers energy and nutrients	Heavy metal content requires monitoring, potential for odor during processing	(Danso-Boateng et al., 2013) (Hämäläinen et al., 2022) (Paiboonudomkarn et al., 2022)

2. Fuel properties

Over the years, various types of biomass feedstocks have been converted into eco-friendly hydrochar via the HTC and several examples representing the forestry, agriculture and biosolids residues are shown in Table 2. Overall, a notable change in the elemental compositions can be observed when these feedstocks were subjected to the HTC treatment. Significant concentration of an elemental C in the hydrochar were recorded for forestry and agriculture waste. By contrast hydrochars derived from biosolids showed lower concentration of C especially for sewage sludge and industrial wastewater sludge. Among the biomass feedstocks reviewed, the maize straw hydrochar contains the highest C content, and hence the high HHV. Meanwhile, the H content of the hydrochar samples were not significantly affected by the HTC process. Concomitantly, the O content of all feedstocks experienced a significant reduction with hydrothermal treatment. Generally, it was found that an enrichment in the C content is accompanied by a decline in the O content as the reaction temperature was elevated. This may due to the decomposition of lignin component and simultaneously removing the oxygen atoms via dehydration and decarboxylation reactions to form a lignite-like materials (Wang et al., 2020). Although it may result in a considerable soot formation, solid-fuel containing oxygen can be useful in boosting the reactivity and combustibility of a fuel (Johansson et al., 2021). The N content across the hydrochar range was observed to be around 0.7 to 5.5 % with sewage sludge from industrial WWT having the highest value. According to Corrado et al. (2020), the high N content is normally associated with high protein compounds in the sewage sludge. However, a high N content in the solid fuel is undesirable as it will result in the formation of nitrogen oxides (NO_x) during combustion which causes air pollution. The advantage of HTC process shows that the N content is able to be reduced by converting the nitrogen in the feedstocks into gas or liquid and separating it from the hydrochar (Zhao et al., 2013).

The HTC process is capable of enhancing the fuel properties of the hydrochar by enriching the FC content and reducing the VM content as evident in all hydrochar samples studied. This isa desirable when considering hydrochar as a solid biofuel since high amount of VM is commonly associated with emission problems upon combustion. The FC in the hydrochar samples ranges from 7.5 - 47.3 % compared to that of the initial feedstocks (3.1 - 20.5 %). Despite the low fixed carbon content, the HHV remains high. The high C content can be attributed to the presence of C in the volatile matter. The utilisation of hydrochar as a solid fuel in combustion processes is not recommended due to certain limitations. However, hydrochar exhibits advantageous properties such as high energy density and reactivity, making it suitable for many applications. It is clear from the tabulated data that the HTC is favourable for the treatment of agriculture wastes more than the other type of biomass. As expected, due to the C concentration and liberation of O atoms, all hydrochar samples exhibited higher HHV (12.9 – 29.9 MJ/kg) compared to their starting feedstocks (10.7 – 18.2 MJ/kg). According to Z. Liu et al. (2013), the increase in the HHV after HTC treatment is attributed to the breakdown of low-energy chemical bonds and the formation of high-energy chemical bonds. In this regard, lignocellulosic rich forestry residues and agriculture wastes showed higher HHV than those of the biosolids. This confirms that the HHV of the hydrochar depends strongly on the composition of the starting feedstocks. The trend is contributed by the considerable presence of combustible constituents such as C and H atoms in these samples and non-combustible contents such as ash in the starting materials. Biosolid feedstocks experienced a rise in the ash content after the HTC treatment, which may result from the buildup of inorganic fraction in the hydrochar and the breakdown of organic matter into the process liquid (Roslan et al., 2023). Solid fuel with low ash content is favourable in order to prevent from fouling, slagging and corrosion problems which lowers the efficiency of the combustion process and contributes to a higher maintenance cost (Yao et al., 2017).

Table 2: Fuel properties of hydrochar produced via HTC from various feedstock.

Feedstock	Ultimate analysis, wt % (hydrochar)					Proximate analysis, wt % (hydrochar)			HHV (MJ/kg)	Reference(s)
	C	H	N	O	S	FC	VM	Ash	Feedstock (hydrochar)	
Forestry residue										
• Beet pulp	43.4	5.9	1.5	46.0	0.1	15.6	81.2	1.0	17.3	(Wilk et al., 2020)
	(60.8)	(5.9)	(2.1)	(26.3)	(0)	(28.3)	(66.7)	(1.0)	(25.4)	
• Chinese palm fan	49.0	6.1	-	36.1	-	15.8	77.2	7.0	20.4	(Yao and Ma, 2019)
	(66.5)	(5.8)		(16.3)		(23.2)	(68.5)	(8.3)	(29.4)	
• Pinewood sawdust	46.5	6.0	0.1	47.4	-	-	-	-	-	(Zhang et al., 2019)
	(67.8)	(4.8)	(0.03)	(27.4)						
Agriculture wastes										
• Agriculture waste from corn field	46.2	6.5	0.6	43.3	0.4	18.4	78.6	3.0	18.1	(Wu et al., 2023)
	(71.3)	(5.6)	(1.1)	(20.3)	(0.2)	(46.7)	(51.8)	(1.5)	(28.6)	
• Oil palm EFB	48.8	6.1	0.6	40.7	-	3.0	32.6	1.0	19.98	(Jamari and Howse, 2012)
	(62.2)	(5.3)	(0.9)	(30.6)		(-)	(-)	(1.0)	(28±0.5)	
• Maize straw	45.3	5.9	0.8	42.2	0.2	16.2	78.4	5.5	18.2	(Wang et al., 2020)
	(73.0)	(5.4)	(1.4)	(18.1)	(0.1)	(47.3)	(50.7)	(2.1)	(29.9)	
Biosolids										
• Sewage sludge	46.9	6.1	4.2	50.9	-	5.3	70.3	16.0	18.0	(Danso-Boateng et al., 2013)
	(46.2)	(5.8)	(1.9)	(49.4)		(7.5)	(63.7)	(25.3)	(23.1)	
• Pulp and paper industry sludge	42.4	5.8	1.2	50.2	0.4	-	-	12.7	14.9	(Hämäläinen et al., 2022)
	(55.5)	(5.1)	(1.6)	(37.3)	(0.5)			(19.7)	(20.4)	
• Industrial wastewater treatment sludge	27.4	4.5	5.7	22.7	0.5	17.3	41.9	39.3	10.7	(Paiboonudomkarn et al., 2022)
	(28.0)	(3.9)	(5.5)	(3.9)	(0.6)	(20.7)	(25.1)	(58.2)	(12.9)	

3. Experimentally determined HHV vs calculated HHV

HHV is a measure of the energy content of a substance or fuel which can be determined experimentally using an adiabatic bomb calorimeter or calculated using the established Dulong equation with inputs from the ultimate and/or proximate analysis data (Krishnan et al., 2018). Quite recently, more correlations were established based on the original Dulong equation. In this article, four equations were considered to evaluate the extent of the hydrochars elemental composition on the HHV and their accuracy against the experimental measurement. The equations include the simplest (Eq(1)), the original Dulong equation (Eq(2)), equations which consider sulphur content (Eq(3)) and finally equation that include the influence of ash (Eq(4)). These equations were chosen based on the mean absolute error less than 7 % compared to experimental value, demonstrating their strong universal application. The calculated HHV data are shown in Table 3 together with their experimental HHV pairs retrieved from the respective references. The percentage difference between the two measurements were presented in bracket. Contrary to general believe, that the calculated HHV accuracy against the experimental

HHV measurement will increase with increasing complexity of the equation, the results showed no distinctive trend. According to Poomsawat and Poomsawat, (2021), the HHV of the biomass and the resulting hydrochar were directly related to the increasing carbon content and reduction in an oxygen content. Yang et al. (2022) observed that the HHV of the hydrochar increased with decreasing ash content. The inclusion of C and H variables in the equations are appropriate because it is widely recognized that both elements are central to the volatile organic matter and fixed carbon, hence contributed greatly to the biomass energy content (Sheng and Azevedo, 2005). It shows that the HHV value is dependent on the chemical composition of feedstock and hydrochar. The choice of the equation is based on type of feedstocks; Eq(1) suitable for biomass from agricultural residues, wood and plant materials, fruit and nut shells and other specific plant materials (Chun-Yang, 2011) , Eq(2) is a correlation model for calculating the heating value of coal samples, Eq(3) is derived for biomass material based on pertinent combustion of C, H and S to produce CO_2, water vapor and SO_2 and suitable for coconut shells, groundnut shells, types of wood and other biomass material with significant C, H and S content and relatively simple combustion reactions (Channiwala and Parikh, 2002) and Eq(4) can be used to calculate HHV of various fuel types, including gaseous fuels, liquid fuels, solid fuels like coal/coke, and various biomass materials, diverse agricultural residues, industrial by-products and waste materials (Channiwala and Parikh, 2002).

$$\mathrm{HHV1\ (MJ/kg)} = 0.2949\mathrm{C} + 0.8250\mathrm{H} \tag{1}$$

$$\mathrm{HHV2\ (MJ/kg)} = 0.3383\mathrm{C} + 1.422\ (\mathrm{H} - \mathrm{O}/8) \tag{2}$$

$$\mathrm{HHV3\ (MJ/kg)} = 0.328\mathrm{C} + 1.419\mathrm{H} + 0.0928\mathrm{S} \tag{3}$$

$$\mathrm{HHV4\ (MJ/kg)} = 0.3491\mathrm{C} + 1.1783\mathrm{H} + 0.1005\mathrm{S} - 0.1034\mathrm{O} - 0.0151\mathrm{N} - 0.0211\mathrm{Ash} \tag{4}$$

Table 3: HHV evaluated using correlation equation.

Hydrochar from feedstock	Experimental HHV (MJ/kg)	HHV1 (MJ/kg) (% diff.)	HHV2 (MJ/kg) (% diff.)	HHV3 (MJ/kg) (% diff.)	HHV4 (MJ/kg) (% diff.)	Reference(s)
Forestry residue						
• Pinewood sawdust	24.30	23.93 (1.52)	24.86 (2.30)	29.02 (19.42)	26.46 (8.89)	(Zhang et al., 2019)
Agriculture wastes						
• Agriculture waste from corn field	28.62	25.65 (10.38)	28.48 (0.49)	31.35 (9.54)	29.36 (2.59)	(Wu et al., 2023)
• Oil palm EFB	28±0.5	22.71 (18.89)	23.14 (17.36)	27.92 (0.29)	24.76 (11.57)	(Jamari and Howse, 2012)
Biosolids						
• Sewage sludge	23.13	18.41 (20.41)	15.10 (34.72)	23.39 (1.12)	17.29 (25.25)	(Danso-Boateng et al., 2013)
• Pulp and paper industry sludge	20.4	20.57 (0.83)	19.40 (4.90)	25.49 (24.95)	21.14 (3.63)	(Hämäläinen et al., 2022)
• Sewage sludge from industrial WWT	12.89	11.46 (11.09)	14.31 (11.01)	14.76 (14.51)	12.70 (1.47)	(Paiboonudomkarn et al., 2022)

*% diff = |(Experimental HHV – Calculated HHV)|/Experimental HHV

4. Combustion behaviour

The main purpose of performing the combustion behaviour analysis for hydrochar is to determine the combustion efficiency. Researchers can optimize the combustion and energy output by examining T_i, T_{bo}, and combustion rate where an efficient and sustainable combustion systems that use hydrochar as a renewable energy source can be implemented (Tang and Zhang, 2019). Three characteristic temperatures that define combustion behaviour are evaluated from a TG-DTG curves; ignition temperature (T_i), maximum combustion rate temperature (T_m) and burnout temperature (T_{bo}). In addition, comprehensive combustibility index (CCI) characterizes the comprehensive combustion performance of the sample and a larger CCI value indicates better combustion characteristics of the sample (Chen et al., 2020). Table 4 shows the combustion characteristics of three selected hydrochars derived from each of the biomass feedstocks category. As can be seen, the hydrochar starts burning when the temperature exceeds 250 ºC, except for the hydrochar derived from sewage sludge (Paiboonudomkarn et al., 2022). Most of the hydrochars recorded a T_m between 350 and 500 ºC indicating an effective region for combustion. Meanwhile for the T_{bo}, it represents the temperature at which all the volatile components of the fuel have been burned off and leaving behind only non-carbonaceous residues. Studying the burnout temperature helps in understanding the extent of combustion and the completeness of the combustion process (Sahu et al., 2014). A good solid biofuel exhibits high HHV, low T_i for quick start-up, high T_{bo} to give more complete combustion and leaving fewer unburned residues and high T_m to provide more intense and rapid heat release and get better combustion efficiency. Additionally, understanding of the kinetics and mechanisms involved in the combustion process will aids in modelling and predicting the behaviour of the hydrochar during combustion. Concurrently, analyzing combustion behaviour can determine the hydrochar's energy potential and viability as a sustainable alternative to fossil fuels.

Table 4: Combustion characteristic of some hydrochar from various feedstocks

Hydrochar from feedstock	T_i (ºC) (ignition)	T_m (ºC) (combustion)	T_{bo} (ºC) (burnout)	CCI (10^{-7}) (%/C^3 min^2)	Reference(s)
Forestry residue					
• Pinewood sawdust	256.48	432.14	441.23	0.768	(Zhang et al., 2019)
Agriculture wastes					
• Agriculture waste from corn field	319.50	336.57	478.04	11.0	(Wu et al., 2023)
• Oil palm EFB	340	477	538	-	(Parshetti et al., 2013)
Biosolids					
• Sewage sludge	191	350	488	1.09	(Paiboonudomkarn et al., 2022)

5. Conclusions

The feasibility of hydrochar as a suitable alternative solid fuel can be rigorously assessed based on its proximate analysis (fixed carbon (FC), volatile matter (VM), and ash content), ultimate analysis (C, H, N, O, S elemental compositions) and high heating value (HHV). The evaluation of these components is important to give a perspective on the applicability of the hydrochar produced via the HTC. Hydrochar fuel properties are strongly influenced by the conditions at which they are produced such as reaction temperature, residence time and pressure. Nonetheless, feedstock type also plays a significant role in the formation of the hydrochar and its qualities as solid fuel. Feedstock rich in lignocellulosic content, low in ash, and with high moisture content may be well-suited for the HTC process to produce hydrochar as a solid biofuel. Furthermore, investigations on parameters such as ignition temperature, burnout temperature, combustion rate are important in order to optimize combustion efficiency and ensure proper utilization and management of hydrochar as a renewable energy source.

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