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Energy Transformation: Assessment of Urban Lignocellulosic Biomass Biofuels via Hydrothermal Carbonization: a Review

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Urban lignocellulosic biomass (ULB), derived from the maintenance of public and private green spaces, such as parks, gardens, sports facilities, and areas along roads, emerges as a solid alternative to replace fossil sources. These resources stand out due to their abundance, low cost, and availability. Hydrothermal carbonization (HTC) emerged as an efficient technology to process this biomass that has a high moisture and ash content, eliminating the need for prior drying. This article reviews the conversion of ULB to solid biofuels through the HTC process, addressing the physicochemical and energetic properties of ULB and the resulting hydrochar, as well as the HTC process and the influence of the operating variables that affect the energetic quality of hydrochar. The carbonization temperature (180 to 280 °C) has a very significant influence more than the residence time (0.5 to 24 h) on the increase in the higher heating value (17.83 to 30.12 MJ kg^{-1}) and the fuel ratio, the decrease in the H/C and O/C ratios, the decrease in the combustibility index, the increase in the ignitability index (> 14.5 MJ kg^{-1}), and the carbon range. However, high carbonization temperatures and long residence times in the reactor are unsuitable because they require large amounts of energy to perform HTC. Therefore, biofuel prices, biomass logistics, optimization of HTC process variables, and reduction of energy consumption are challenges for the HTC process to generate a solid biofuel with excellent energy properties.

1. Introduction

Industrialization and anthropogenic activities have caused an increase in global energy demand, due to the consumption of fossil fuels, which has generated various global environmental problems, such as climate change (Lee et al., 2023). Urban lignocellulosic biomass (ULB), a significant fraction of urban biomass, includes mainly urban green waste such as branches and leaves, and waste wood products, including sawdust, wood furniture, and manufacturing by-products (Li et al., 2017). Traditionally, this waste has been deposited in landfills, which occupy large areas due to its low density, typically ranging from 50 to 75 kg m^{-3} (Gupta et al., 2018). It has also been left to decompose outdoors, incinerated in open-air settings (Sharma & Dubey, 2020), or partially composted (Shao et al., 2020). The urban pruning waste has not been valorized due to limitations such as complex composition, transportation, and logistics (Maccarini et al., 2020). However, a significant fraction of ULB could be used and converted into suitable energy for the daily activities of urban residents (Li et al., 2017). Urban lignocellulosic biomass (ULB) stands out as a valuable raw material, due to its abundance and availability, low cost, and favorable ecological characteristics that make it ideal for producing biofuels (Awasthi et al., 2023). Hydrothermal carbonization (HTC) is a process that converts wet biomass into carbon-rich solids known as hydrochars (Merzari et al., 2018). Unlike pyrolysis or gasification, the HTC process does not require pre-drying to produce carbon-grade solid fuels (Wilk et al., 2020).

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Therefore, HTC has become a promising technology for producing carbonaceous materials with negative carbon emissions from dry and wet ULB (Yu et al., 2023). HTC is performed in water under autogenous pressure and at temperatures between 160 and 280 °C (Merzari et al., 2018), with residence times of up to 24 h (Sharma et al., 2019). HTC produces a solid fraction with a high carbon content (hydrochars), a liquid fraction with many organic compounds, including valuable carbon, and a gaseous fraction, mainly CO_2 (Wilk et al., 2020). Studies on the operation conditions of the HTC process such as temperature (T), residence time (t), biomass/water ratio (B/W), and pressure (P) have shown that they influence both the physicochemical and energetic properties of hydrochars (Merzari et al., 2018; Sharma et al., 2019).
This review focuses on the conversion of urban lignocellulosic biomass (ULB) through HTC, as well as the influence of the operation conditions of the HTC reactor on the production of hydrochars for energy purposes. For this reason, much research has prioritized agricultural and forest biomass; however, ULB poses unique challenges due to its heterogeneous composition, species diversity, lack of separation, and recycling.

2. Urban lignocellulosic biomass

2.1 Chemical composition

The chemical composition plays a fundamental role in its suitability for biofuel production. Yard waste composed of grass (33 %), dry leaves (65 %) and wood chips (2 %) contain 38.01 % of cellulose, 25.05 % of hemicellulose, and 19.77 % of lignin (Panigrahi et al., 2019). On the contrary, furniture wood waste contain 44.7 % of cellulose, 18.0 % of hemicellulose, and 33.0 % of lignin (Moreno & Font, 2015).

2.2 Physical, chemical and energetic characteristics

Urban lignocellulosic biomass (ULB) is characterized by its high moisture content, which varies depending on the source. *Acacia* pruning (*Robinia pseudoacacia*) contains approximately 50 % moisture (Wilk et al., 2020), while pruning *Ficus benjamina* (Fb), its moisture content varies between 6.4 and 9.44 % on a dry basis (Llanos et al., 2023). The high moisture content not only influences aspects such as transport, storage, and processing of biomass, but also affects the properties of hydrochars and their energy quality (Yan et al., 2020). Table 1 shows significant variability in volatile matter content (VM) in ULB, ranging from 69.66 and 87.96 %. ULB typically contains a high VM content, leading to increased reactivity and lower enthalpies of combustion. In contrast, coals have low VM content, resulting in lower reactivity and higher combustion enthalpies (Sharma et al., 2019).
The ash content present in the ULB varies widely, between 0.16 and 11 %, as shown in Table 1. Garden pruning, Fb branches, furniture wood waste, and *Acacia* trees have a low ash content, below 5 %, and are suitable for energy conversion. This variability is due to the influence of biomass species, and climatic, agricultural, and agronomic factors. Regarding elemental composition, it is crucial to maintain N and S levels below 0.98 % and 0.24 %, respectively, to avoid negative environmental impacts (Ahmad et al., 2017). ULB presents significant potential for use in energy facilities, enabling energy production. For instance, the city of Pato Branco, Brazil, aims to generate between 1 and 1.5 MW of electricity daily using 4.84 to 6.77 ton of ULB (Maccarini et al., 2020).

Table 1: Proximate, ultimate, and calorific analyses of urban lignocellulosic biomass (ULB), % wt., dry basis

ULB	Proximate analysis			Elemental analysis				HHV,	Ii*,	CI**,	Reference
	VM	Ash	FC	C	H	N	S	MJ kg^{-1}	MJ kg^{-1}	MJ kg^{-1}	
Grass clippings	73.8	11.0	15.2	44.2	6.9	3.9	---	18.2	17.70	87.5	(Brown et al., 2022)
Waste furniture	87.38	1.37	11.25	49.13	6.19	0.12	0.01	19.99	18.53	168.03	(Dang et al., 2023)
Lawn cuttings	72.66	10.47	16.87	48.03	6.08	2.18	0.18	17.08	15.66	73.2	(Hansen et al., 2022)
GPW	76.5	5.1	18.4	46.9	6.1	0.9	0.4	19.7	17.62	85.73	(Ipiales et al., 2022)
Ficus b. branches	77.59	3.39	12.62	42.36	6.15	0.64	0.04	17.34	15.55	113.3	(Llanos et al., 2023)
Furniture wood waste	77.30	1.8	20.9	47.9	6.0	2.9	0.05	15.8	11.30	63.0	(Moreno & Font, 2015)
Green waste	87.96	10.59	1.45	44.10	5.59	1.12	---	18.15	20.08	1094.8	(Shao et al., 2020)
Acacia tree	76.58	0.16	16.04	50.60	5.62	0.21	1.23	20.56	19.77	107.4	(Wilk et al., 2020)

Where: FC: Fixed Carbon. Ii and CI: Combustion indices. (*, **): Estimated results (Conag et al., 2017)

The higher heating value (HHV) of ULB varies between 15.8 MJ kg^{-1} (Moreno & Font, 2015) and 20.56 MJ kg^{-1} (Wilk et al., 2020), as detailed in Table 1. Combustion indices such as fuel ratio (FR), combustibility index (CI), and ignitability index (Ii) are used to evaluate solid fuel before its use in power generation systems or its blending with coal (Ohm et al., 2015). The ULB have a very low FR (FC/VM) because they contain a high content of volatile matter, so they are not suitable for direct energy generation. The HTC process as pretreatment could improve the fuel ratio of the ULB. Subbituminous coal used in thermal power plants usually has a FR ranging from 0.5 and 1. Furthermore, for coal-fired power plants, the recommended FR falls between 0.5 and 3 (Ohm et al., 2015). The ULB ignitability index varies between 11.30 and 20.08 MJ kg^{-1}, as detailed in Table 1, recommending a minimum of 14.5 MJ kg^{-1} (Conag et al., 2017). The combustibility index used to evaluate the compatibility of biomass with the coal mixture suggests a range of 12.5 MJ kg^{-1} to 23 MJ kg^{-1} (Ohm et al., 2015).

3. Hydrothermal carbonization process

Hydrothermal carbonization (HTC) emerges as an effective alternative to process biomass with a high moisture and ash content, eliminating prior drying. This leads to the obtaining of a biofuel with a 20 % to 30 % increase in calorific value compared to the original biomass (Vallejo et al., 2022). This process also improves biomass dehydration, storage, and logistics (Hansen et al., 2022). During HTC, hydrolysis, dehydration, decarboxylation, condensation, and aromatization reactions occur sequentially and simultaneously (Merzari et al., 2018). The main product derived from HTC is an energetic and dense solid known as hydrochar, biochar, or HTC coal (Mendoza Martinez et al., 2021) that has properties similar to coals such as lignite (Sharma et al., 2019), peat, and bituminous (Dang et al., 2023). Typically, HTC coal contains between 55 and 90 % of original biomass and retains between 80 and 95 % of its energy content (Mendoza Martinez et al., 2021). Hydrocarbons exhibit a higher carbon content, in a range of 50 % and 67 %, thus enhancing both the calorific value and the FC content compared to initial biomass (Wilk et al., 2020). The HTC process could achieve self-sufficiency using only 43 % to 51 % of the energy generated from hydrocarbon combustion, with surplus energy directed toward electricity generation (Ipiales et al., 2022). Furthermore, integration of the HTC process with a Rankine cycle power plant would streamline the process design and concurrently improve its efficiency (Mendoza Martinez et al., 2021).

4. Effect of operating parameters on the fuel characteristics

4.1 Temperature

Temperature is the most influential variable affecting fuel properties, significantly enhancing its energy characteristics (Hansen et al., 2022). As shown in Table 2, a gradual temperature increase leads to higher fuel ratio (FR), HHV, energy density (ED) and energy yield (EY) of hydrochar. This thermal rise increases FC content while reducing VM content, thereby contributing to an increase in HHV and C content (Sharma et al., 2019). The lignocellulosic structure fragments due to hemicelluloses, with observable thermal cellular changes in resulting hydrocarbons (Mendoza Martinez et al., 2021). These observations indicate improved hydrocarbon classification (lignite, sub-bituminous, bituminous) due to dehydration and decarboxylation reactions in the HTC reactor (Gao et al., 2016), decreasing H and O content and improving H/C and O/C ratios with rising HTC process temperatures (Śliz & Wilk, 2020). Fuels with low O/C and H/C ratios are preferred for reduced smoke emissions, water vapor and energy loss. However, higher temperatures decrease both HHV and EY due to the loss of energy caused by the natural degradation of lignocellulose, generating smaller particles that are released from the hydrocarbon and are carried away by the inert gas of the reaction system (Yao et al., 2016). The EY values presented in Table 2 are within the range observed in hydrocarbons derived from forest biomass (61 - 79 %) and agricultural residues (54 - 69 %) (Mendoza Martinez et al., 2021). Hydrocarbons exhibit properties similar to mineral coals, are suitable for industrial applications as fuel and comply with the regulations of the ISO 17225-8 standard (Ipiales et al., 2022). Additionally, hydrocarbons that meet the FR and ignitability index (Ii > 14.5 MJ kg^{-1}) requirements are suitable for domestic and residential heating applications (Conag et al., 2017).

4.2 Residence time

Several studies have explored the effect of residence time on the HTC process of different biomasses, covering periods from 0.5 to 24 h, as detailed in Table 2. Long carbonization periods, approximately 8 h, have been observed to result in a significant increase in the content of C, HHV, FR, and ED. However, during these long periods, the VM content, the H/C and O/C ratios, as well as the EY, decrease significantly, with no apparent influence on the ignitability index. It should be noted that the influence of longer periods, starting from 12 h, is significant in ED and Ii, while the H/C and O/C ratios do not present significant changes (Sharma et al., 2019). Similar results have been reported on H/C and O/C ratios, with constant values at various time intervals (Gao et al., 2016). Other studies highlight that residence times of 4, 6, 8, and 10 h do not generate a significant effect on performance, properties, and thermal behavior (Wilk et al., 2020). In the HTC of garden waste, it has been identified that after 3 h the maximum ED is reached, but the yield decreases due to the slow degradation of cellulose, resulting in a reduction in EY. However, according to other studies, the impact of residence time on fuel properties is considered less significant compared to reaction temperature (Hansen et al., 2022).

4.3 Biomass/Water ratio

In the HTC process, the most commonly used biomass/water (B/W) ratio, is around 1/10, as detailed in Table 2. It has been observed that this ratio barely significantly influences the HHV, ED, and EY in hydrochar derived from pine sawdust (Vallejo et al., 2022). In specific studies on HTC of *Acaci*a trees, an increase in VM content was observed from 70.85 to 71.85 % and an increase in the ignitability index from 17.66 to 18.30 MJ kg^{-1}. Furthermore, there was also a significant decrease in FC content and HHV. During HTC at 200 °C for 4 h with B/W ratios of 1/5, 1/8, and 1/10, no significant differences were observed in the EY, ED, FR, and the H/C and O/C ratios (Wilk et al., 2020). On the other hand, in the HTC of the waste furniture, detailed in Table 2, an increase in FC content was observed, while the FR, HHV, ED, EY, H/C and O/C ratios and the ignitability index decreased when operating at 240 °C for 0.5 h with B/W ratios of 1/1 and 1/2 (Dang et al., 2023). Another study highlighted that temperature and the B/W ratio are the primary influential variables in optimizing the energy properties of sawdust-derived hydrocarbons. They achieved an EY of 77 % and an HHV of 28.6 MJ kg^{-1} at 280 °C for 100 min, with a B/W ratio of 14 % (Vallejo et al., 2022). However, other studies suggest that the B/W ratio does not yield significant variations in the energetic properties of hydrocarbons (Gallant et al., 2022; Wilk et al., 2020).

4.4 Pressure

In the HTC process, as the temperature increases, the pressure increases until it reaches subcritical water conditions (Yu et al., 2023). A slight increase in HHV is observed with increasing pressure, while the rest of the properties did not show significant differences when the pressure in the HTC reactor is about 30 MPa. However, a significant increase in HHV, and a notable improvement in hydrochar graphitization (Heidari et al., 2019), are observed when the HTC reactor operates at elevated pressures, up to 100 MPa. However, higher pressures in the reactor could generate issues in your design, and increase manufacturing costs (Marzbali et al., 2021).

Table 2: Energetic properties of hydrochars from urban lignocellulosic biomass

ULB	Hydrochar T t W/B	VM	FC	FR	H/C	O/C	HHV MJ kg^{-1}	EY %	ED	Ii MJ kg^{-1}	References
Grass clippings	200-1-10	69.7	20.70	0.30	1.53	0.49	22.0	68.90	1.21	21.53	(Brown et al., 2022)
	250-1-10	58.6	27.40	0.47	1.28	0.25	25.80	52.90	1.42	28.22	
Waste furniture	240-0.5-1	59.23	23.27	0.67	1.23	0.48	26.73	82.70	1.34	22.47	(Dang et al., 2023)
	240-0.5-2	62.28	36.48	0.59	1.03	0.34	25.64	79.55	1.28	21.37	
	260-0.5-2	55.05	44.06	0.80	0.92	0.27	27.78	79.07	1.39	23.41	
	280-0.5-2	46.18	52.67	1.14	0.78	0.18	30.12	65.08	1.51	26.67	
Garden and park waste	210-1-4	67.6	28.60	0.42	1.21	0.51	22.3	86.19	1.16	18.69	(Ipiales et al., 2022)
	230-1-4	60.9	34.10	0.56	1.11	0.41	24.5	84.69	1.24	21.30	
Yard waste	180-2-10	77.50	17.18	0.22	1.30	0.69	17.83	86.43	1.15	15.51	(Sharma et al., 2019)
	200-2-10	68.99	24.95	0.36	1.22	0.64	19.60	78.52	1.26	16.19	
	200-8-10	64.90	28.48	0.43	1.08	0.50	20.16	66.15	1.31	16.23	
	200-12-10	64.01	28.45	0.44	1.02	0.41	23.33	77.81	1.51	21.42	
	200-24-10	63.24	27.80	0.43	1.03	0.40	24.59	72.50	1.60	24.03	
Acacia tree	200-2-10	74.94	23.50	0.31	1.22	0.49	21.63	86	1.20	18.26	(Wilk et al., 2020)
	200-4-10	71.85	25.63	0.36	1.16	0.41	21.81	86	1.17	18.30	
	200-4-8	71.13	26.47	0.37	1.15	0.39	21.51	84	1.19	17.66	
	180-4-10	77.82	21.09	0.27	1.23	0.46	20.92	87	1.16	17.72	
	220-4-10	61.49	36.80	0.60	0.99	0.29	23.89	77	1.32	18.62	
Native kenaf stem	250-0.5-8	68.76	27.44	0.40	1.12	0.48	21.33	45.55	1.29	17.53	(Youn et al., 2023)

5. Conclusions and future perspectives

The use of urban lignocellulosic biomass (ULB) faces significant technical, logistical, environmental, and economic challenges. These challenges range from the collection, handling, and segregation of biomass to logistical problems such as transportation and regular supply of this resource. Overcoming these challenges is essential, as it directly contributes to the reduction of ULB, the reduction of carbon emissions, and the reduction of dependence on nonrenewable resources. Overcoming these challenges motivates the circular economy and promotes sustainable development. The complex composition of the ULB stimulates its exploration and potential transformation through the HTC process, in order to optimize the utilization of these renewable, economical and sustainable resources in energy applications. With respect to the HTC process, the main challenge is to reduce the energy consumption required by the process so that it can be consolidated as a competitive technology compared to fossil fuels. To do this, an effective reduction of the carbonization temperature is required, therefore avoiding the loss of biomass components. Strategies such as decoupling temperature from pressure to reduce temperature (Yu et al., 2023), minimizing residence time, or exploring catalysts to enhance both process efficiency and product quality (Heidari et al., 2019) are potential pathways. Conducting an exhaustive scientific and technical analysis of the HTC reactor becomes essential. This comprehensive analysis covers the complex composition of ULB and optimizes operating conditions, such as temperature, residence time, and B/W ratio, to maximize the energetic quality of the resulting hydrochar. Hydrochars derived from ULB, by present combustion properties similar to peat, bituminous, subbituminous, and lignite, show favorable qualities, including high HHV (17.83 MJ kg^{-1} to 30.12 MJ kg^{-1}), ED (1.16 to 1.61), and ignitability index (15.51 to 28.22 MJ kg^{-1}), making them suitable for solid biofuel applications. Integration with other processes, such as anaerobic digestion and energy cogeneration cycles, holds promise in substantially improving HTC efficiency. Future research is required to quantify improvements in energetic fuel properties, considering aspects such as fuel price, reactivity, and logistical aspects.

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