REVIEW

Hydrochar Pelletization towards Solid Biofuel from Biowaste Hydrothermal Carbonization

Ao Li, Kai Jin, Jinrui Qin, Zhaowei Huang, Yu Liu, Rui Chen, Tengfei Wang[*] and Junmin Chen[*]

Faculty of Geosciences and Environmental Engineering, Southwest Jiao Tong University, Chengdu, 611756, China
[*]Corresponding Authors: Tengfei Wang. Email: wtfgxp@163.com; Junmin Chen. Email: cjm@swjtu.edu.cn

ABSTRACT

Hydrothermal carbonization is highly applicable to high moisture biomass upgrading due to fact that moisture involved can be directly used as reaction media under subcritical-water region. With this, value-added utilization of hydrochar as solid fuel with high carbon and energy density is one of the important pathways for biomass conversion. In this review, the dewatering properties of hydrochar after the hydrothermal carbonization of biowaste, coalification degree with elemental composition and evolution, pelletization of hydrochar to enhance the mechanical properties and density, coupled with the combustion properties of hydrochar biofuel were discussed with various biomass and carbonization parameters. Potential applications for the co-combustion with coal, cleaner properties and energy balance for biowaste hydrothermal carbonization were presented as well as the challenges.

KEYWORDS

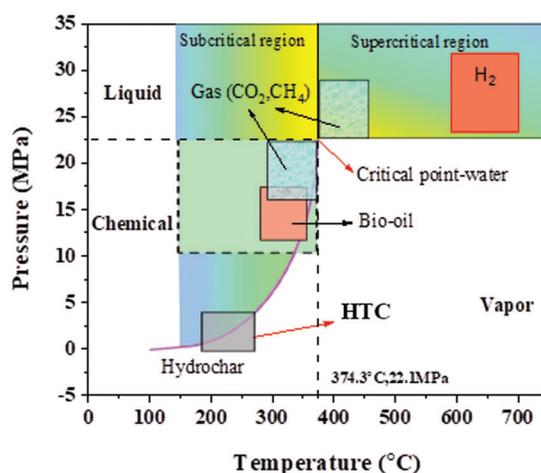Biomass; hydrothermal carbonization; hydrochar; pelletization

1 Introduction

Biomass represents the non-fossil energy and biodegradable organic material derived from plants, agriculture and forestry waste [1]. In addition to traditional lignocellulosic materials, scientists focus more on biowaste valorization like sludge and microalgae in recent years due to the access to recover carbon sources and the reduction of environment burden simultaneously [2]. Generally, bottleneck of biomass resource utilization is the high moisture, low mass and carbon density, and low energy content. Considering the high cost of separation and treatment of water involved in biomass, new techniques are required to develop to meet reaction with both biowaste and water, and hydrothermal carbonization (HTC), liquefaction (HTL), and gasification (HG) have got enough attention to replace the traditional pyrolysis pathways [3]. Compared with traditional thermal conversion pathways like high temperature or low temperature pyrolysis, microwave pyrolysis, flash pyrolysis, HTC conversion pathways have showed several advantages like the flexible regulation, high efficiency, and short cycle in biomass pretreatment, upgrading, and materials preparation [4,5]. Another difference is that solid-liquid separation is required for the hydrochar and liquid phase, and this is also problem for industrialization application of HTC.

Under high temperature and pressure, water can be used as reactants, catalyst, and energy of the media during subcritical region. As shown in Fig. 1, the dielectric constant water decreased significantly below the

critical point [6]. Thus, varying hydrothermal parameters or severity can efficiently convert biomass into desired mass yield of hydrochar, biocrude-oil and gas products. Especially, a series of reactions such as hydrolysis, dehydration, ploymerization reaction is the domain reaction for biomass carbonization [7,8]. For HTC of biowaste, feedstock undergos hydrolysis, dehydration, decarboxylation reaction, after which intermediate products such as HMF undergo re-dehydration or isomerization, and the ploymerization of these chemicals formed typical hydrochar with core-shell structure [9]. The carbonization mechanism is basically concluded from the chemical structure and surface characteristics until now which is generally regarded as the mimic process of natural coalification [9]. One of the most typical tendency is that the dehydration and decarboxylation reaction can significantly reduce the O and H content in hydrochar, and aromatic structure dominates at high hydrothermal severity [10]. The increased carbon density and unique structure stimulate researchers to apply hydrochar as solid biofuel or carbon materials. However, HHVs depended much on feedstock and hydrothermal parameters, and low energy content can not support a stable combustion and co-combustion with coal [11]. In addition, calorific value increases to some extent after HTC, and part of nitrogen, sulfur, chlorine can be removed by water-washing release or cracked in liquid phase, so cleaner performance of hydrochar can be achieved [12,13]. The differences between hydrochar and coal are the energy density, physical-chemical structure, and fixed carbon content, so pelletization process is expected to enhance the physical properties and energy density of hydrochar [14]. However, since works related to hydrochar as pellet fuel is still in initial stage, investigation on energy consumption and pelletization mechanism involved in pellet structure are facing challenges [15,16,17].

Figure 1: Temperature range of subcritical and supercritical water properties for hydrothermal conversion of biomass

In this review, we focused on the hydrothermal carbonization of biomass for solid fuel pelletization, and the fuel properties for dewaterability, element composition and evolution tendency, and corresponding pelletization process and combustion characteristics were discussed. Additionally, future perspectives and challenges were presented about HTC and hydrochar fuel applications.

2 Unique Properties of Hydrochar towards Solid Biofuel

2.1 The Dewatering Properties of Hydrochar

Direct combustion of biomass requires more energy consumption for moisture drying and pollutant gas like SO_2 and NO_x can be released by reaction between water volatilization during combustion [18].

Therefore, hydrothermal carbonization shows advantage for water removal due to the dehydration reaction and removal of hydroxyl groups. Moisture in the biomass is generally divided into free water and bound water [19,20]. Former is easy to be physically removed by heating, while the combined water is aroused by hydroxyl and carboxyl groups in organic matter to form hydrogen bonds, which is difficult to be removed directly [21]. As shown in Fig. 2, the physical structure of biomass is cracked in subcritical water by hydrolysis reaction, and a series of small molecular substances are formed, and the channel in hydrochar became bigger, which resulted in high diffusivity. After HTC, hydrophilic functional groups like hydroxyl groups on hydrochar surface can be significantly weakened compared to feedstock and it can be decreased with the increase of carbonization degree. At the same time, solid-phase condensation reaction occurs during hydrothermal carbonization, since hydrochar is composed of components and spherical structures formed by solid-solid pyrolysis, its pore size is more abundant than that of feedstock, so HTC can be used as one pathway to release water. Coupled with the increased hydrophobic properties, more bound water is converted into free form that can be easily removed by physical separation. Additionally, water produced can not only serve as a solvent, but also participates in hydrothermal carbonization as catalyst and medium [22]. Typical application of dewatering is the hydrothermal conversion of sludge, which showed that hydrothermal carbonization can hydrolyze the viscous organic matter in sludge and destroy its colloidal structure, and release macromolecular organic matter from cell, thereby enhancing the dewatering performance of sludge [23]. The dewatering performance depended much on the feedstock and HTC parameters, as presented in Table 1. For instance, in Peng et al.'s work [24], sludge with woody biomass was mixed as feedstock for HTC, and the dewatering performance is followed by the biomass species, mixer ratio and carbonization temperature (Fig. 3).
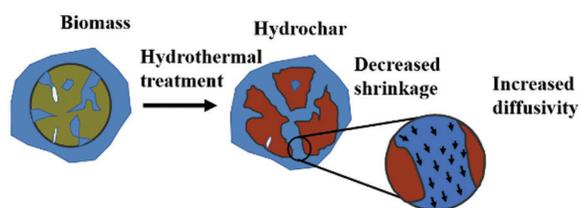
Figure 2: Schematic diagram of channel in hydrochar after hydrothermal carbonization of biomass

Table 1: Comparation of moisture content in feedstock and hydrochar

Feedstock	Temperature (°C)	Residence time (h)	Moisture before HTC (%)	Moisture after HTC (%)	Reference
Food waste	200–230	1	93.0	40.0	[25]
Oil palm waste	150–200	8	15.5	7.0	[26]
Oil palm waste	200–250	8	15.5	5.9	[26]
Oil palm waste	250–300	8	15.5	4.2	[26]
Sludge	240	0.5	90.32–95.51	48.32	[27]
Sludge	240	2	90.32–95.51	41.03	[27]
Watermelon peel	260	3	8.30	0.9	[28]
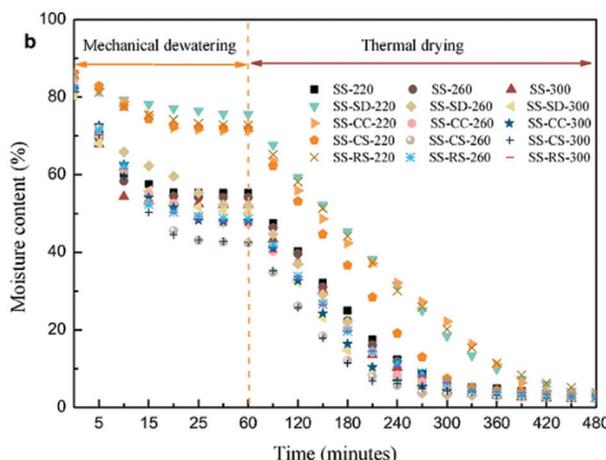
Figure 3: Tendency of the moisture ratio in sludge and lignocellulose mixture derived hydrochar with time of mechanical dewatering and thermal drying [24]

2.2 Elemental Composition and Coalification of Hydrochar

For mimic the coalification process of biomass hydrothermal carbonization, researchers used elemental composition to trace the atomic ratio change including C, H, N, O, and the information can provide details about the degree of dehydration and decarboxylation [29]. Thus, calorific value can be evaluated for hydrochar, and the value ranged from 15–35 MJ/kg generally, which is severally depended on feedstock and hydrothermal severity, since oxygen content and ash have obvious effect on the fuel quality [30]. Even this, the coalification degree of hydrochar is lower compared to typical coal like anthracite and bituminous coal, and in most works hydrochar quality is close to the lignite [31]. As indicated by Table 2, both hydrothermal parameters and feedstocks are dominant factors during HTC. Food waste and woody biomass hydrochar have relatively higher C content than others due to the high sugers which can he easily carbonized. With temperature and residence time increased, both O/C and H/C decreased, along with higher HH *Vs*. Sludge and MSW derived hydrochar has low HHVs due to the high ash content. Even this, hydrochar fuel with dehydrogenation and deoxygenation is essential when high quality fuel like coal is prepared, but the balance of oxygen should be considered when oxygen-rich fuels is prepared.

Table 2: Elemental composition, atomic ratio of hydrochar from different biomass and parameters

Feedstock	Temperature (°C)	Time (Min)	C (%)	H (%)	N (%)	O (%)	O/C	H/C	HHVs (MJ/kg)	Reference
Micro algae	130	50	58.54	9.98	6.11	24.93	0.31	2.04	17.47	[32]
Sludge	240	60	27.87	2.50	1.91	5.48	0.15	1.08	11.57	[27]
Wheat straw	220	30	46.6	8.9	–	47.5	–	–	22.2	[33]
Digestive municipal solid waste	200	120	20.67	2.41	1.24	11.93	–	–	9.24	[34]
Forest waste	280	60	70.75	5.27	1.24	19.95	0.21	0.86	28.78	[35]
Wood biomass	220	240	66.07	5.5	0.223	27.28	0.29	0.99	23.89	[36]
Yard waste	220	60	44.2	6.24	0.84	48.21	0.81	1.61	15.43	[37]
Food waste	220	60	35.16	4.9	1.01	56.8	1.15	1.60	9.48	[37]

(Continued)

Table 2 (continued)

| Feedstock	Temperature (°C)	Time (Min)	C (%)	H (%)	N (%)	O (%)	O/C	H/C	HHVs (MJ/kg)	Reference
Chuanxiong herbal residue	240	–	42.10	6.12	2.17	38.35	0.68	1.73	17.68	[38]
ShenMu lignite coal	240	–	60.31	5.26	0.88	26.76	0.33	1.04	24.39	[38]
Pearl millet	300	60	61.3	2	3	13.4	0.22	0.02	25	[39]
Sewage sludge	200	60	34.32	4.09	3.22	18.42	0.40	1.42	14.21	[40]

The evolution of the elements is highly related to the carbonization mechanism. Feedstocks firstly hydrolysis with temperature increased, and the degradation of hemicellulose is favored than that of lignin and cellulose. After the broken of the body, two pathways of the compositions in liquid products are generally accepted. One is the pyrolysis-like process, which means a directly solid-solid conversion from biomass to hydrochar during HTC, and another is the ploymerization of the chemicals like HMF and the derives to form secondary hydrochar [9]. The pathways depend on parameters. From the view of basic structure of carbon, with the increase of carbonization degree of hydrochar, furan structure transitions to aromatic-rich structure, and carbon density, calorific value of hydrochar increases accordingly [9]. For example, Wang et al. [41] used food waste as feedstock for HTC, and It can be clearly seen that the increase of temperature increased the aromatic structure in hydrochar, and the furan-atomic ratio decreased, which is the directly evidence of the loss of H and O at high hydrothermal severity [42]. Therefore, the atomic number ratio of O/C and H/C also decreases, and calorific value of hydrochar also increased accordingly, which is the most powerful evidence to simulate coalification in the natural process, and is widely used to evaluate the degree of coalification of hydrothermal coals compared with lignite, bituminous and anthracite coal [43]. In addition to reaction temperature, residence time is another factor for varying the elemental composition. However, when reaction time is not enough to support the ploymerization of hydrolyzed intermediates like furans and the derived chemicals in liquid phase, low yield with high oxygen content is obtained [7]. Nevertheless, long residence time will also require high energy consumption. Therefore, reasonable control of residence time is critical for industrial application of hydrothermal carbonization technology.

As observed from Table 2, the elemental composition and calorific value of hydrochar not only depend on the parameters of hydrothermal carbonization, but also can be affected by composition of feedstock. For example, biowaste contained high organic content is prefer to be carbonized than that of high-ash species. In our previous work [41], it suggested that carbonization degree of N-containing biowaste defers from that of N-free biowaste, since Mallard reaction happened and N can be incorporated into hydrochar, which resulted in high N ratio and low C ratio in hydrochar [44]. Until now, the conversion of biomass for solid fuel preparation still faces challenges on fuel upgrading by varying elemental content, and balance of energy consumption of hydrothermal carbonization, which is also the bottleneck for the sustainable development of its industrial application in future.

### 2.3 Pelletization of Hydrochar as Solid Biofuel
Pelletization of hydrochar is essential for solid biofuel preparation and shows great advantage than biomass pelletization like low ash and high mass density, proper moisture, and the enhanced mechanical properties [45]. However, mechanical properties of hydrochar pellets depend on temperature, pressure, particle size, moisture content, feedstock and binders [46]. Generally, A wide range of temperatures from room condition to 140°C in pelletization process is conducted, and physical modification after

pelletization can help the hydrochar particles densification by formation ordered structure. After pelletization, the energy density, mass density and compressive strength have been improved to varying degrees, and the detail pelletization process is depicted in Fig. 4.
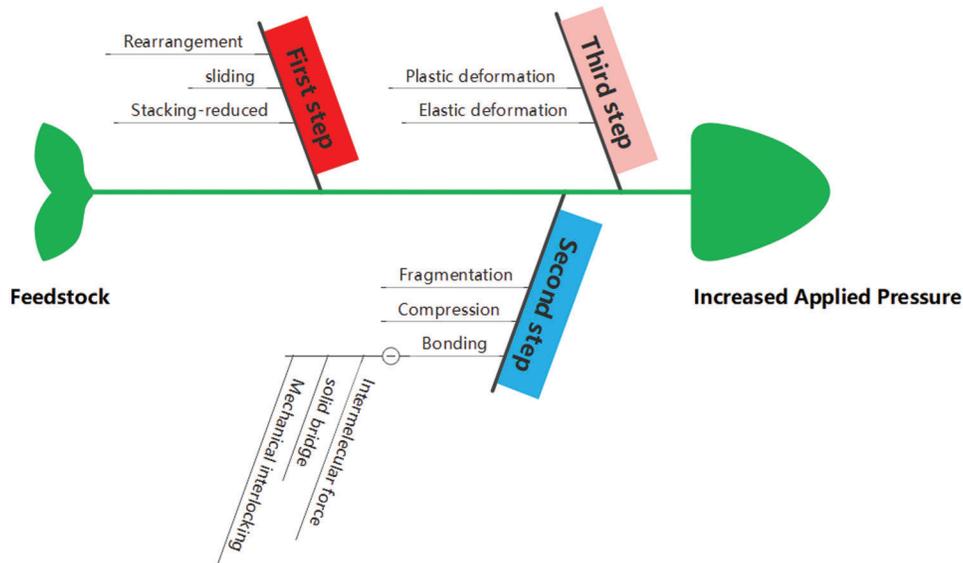
Figure 4: Detail pelletization process of feedstock and changes involved in pellet

In the absence of solid bridges as studied, particles are close enough to bind forces such as intermolecular (valence, hydrogen, and van der Waals), electrostatic, and can make solid particles stick to each other attached. Previous works sudied adhesion theories and proposed a microscopic explanation for particle bonding, and driving force for adhesion from the electronic interaction between molecules, and maximum gravitational force is close to the minimum potential energy by forming chemical bonds [47]. Due to the action of high pressure and high temperature, solid bridges can be formed by molecular diffusion from one particle to another. Solid bridges can be crystallization of certain components, chemical reactions, hardening of binders (such as molasses and tar), solidification of molten components, or linkages between particles, as shown in Fig. 5 [45,48,49]. For lignocellulose derived hydrochar, solid bridges formed from carbonized cellulose is critical considering the content and thermal stable structure than that of hemicellulose. Thus, temperature should be well controlled during HTC to make sure that cellulose and lignin maintains the main physical properties. Meanwhile, the HTC temperature may also have obvious effect on the energy consumption during pelletization of hydrochar. In Wang et al.'s work [22], energy consumption of hydrochar pelletizaiton increased when hydrochar is prepared from 220°C to 260°C, while the high food waste ratio in woody waste blend can decrease approximately 15–20 J/g for the compression process. This suggested that the properties of hydrochar is dominant for the pelletization. Liu et al. [50] reported that liquid bridge can be formed by the local melting of binding components at particle junctions under high densification pressure, and the gaps between hydrochar particles can be reduced. In some works, binders like starch, molasses, and calcium oxide were used to enhance the solid or liquid bridge structure in hydrochar pellet. Binder typically softens at the glass transition temperature or melts at the melting temperature to provide a liquid bridge connecting adjacent particles. For example, at glass transition temperature, lignin softens and flows under pressure, contacting surrounding particles and forming covalent bonds [51].
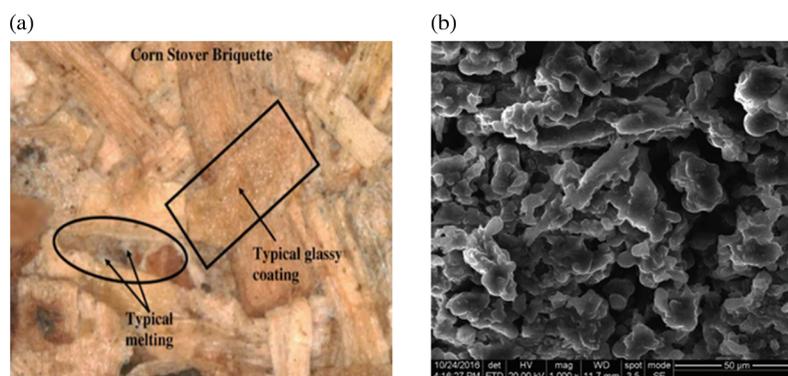
Figure 5: The surface and structure of typical pellet: (a) pelletization with the natural binders and forms coating and melting structure in corn stover pellet; (b) hydrochar with molasses as binder forming solid bridge structure between particles [16,47]

The role of moisture during pelletization is important, since moisture can act as both a binder and a lubricant, so energy consumption of friction can be reduced [52]. Additionally, under the action of heat and pressure, water-soluble components in biomass, such as starch, protein, sugar, soda ash and salt, can undergo starch gelatinization, protein denaturation or salt dissolution in the presence of moisture, thereby improving the degree of binding [16,45]. Water can also form a thin film around the particles, and Van der Waals forces can be further promoted for the binding, as contact area between particles increases. Generally, water content is added with 5%–15% mass ratio during pelletization which is used for lubrication, while high water content can cause pellet fracture due to the higher hydrophobic properties of hydrochar than coal. In addition to feedstock, the pelletization process is related to the holding time, relaxation time, pressure, and compression rate, and systematically optimize these parameters in works is critical to obtain pellet with excellent durability and strength. The energy consumption should also be concerned at the same time [53].

2.4 Combustion Properties of Hydrochar Biofuel

An intuitive advantage is that hydrochar has better combustion performance after HTC. Compared to biomass, a higher ignition point for hydrochar can be achieved, which means a save environment for the hydrochar storage is excepted [38]. It reported that the ignition temperature shifts to a high temperature range around 100°C [54]. As a result, the temperature at maximum peak moves into the high temperature range for hydrochar combustion. In addition, the combustion rate was largely improved, representing about 2~3 times higher than that of biowaste, regardless of hydrothermal severity. Since volatile content is removed during the HTC stage, more fixed carbon can be recovered in hydrochar. For instance, lignin mainly degrades at temperature above 260°C, and the high carbon density and matrix makes it thermal stable [55]. With this regard, HTC parameters have significant influence on the combustion properties of hydrochar. Chen et al. [43] showed HTC temperature had a significant impact on combustion behavior and activation energy of hydrochars. The first combustion decomposition peak of hydrochars were much higher than feedstock, leading to a better combustion performance [5].

Additionally, application of binders also optimizes the combustion properties of hydrochar. For example, Wang et al. [15] studied the effect of four organic binders (protein, starch, lignin and molasses) on woody biomass pellets. Regarding combustion performance, a reduction in ignition temperature was observed for all binder hydrochar. Thus, ratio of fixed carbon and volatile content in hydrochar is highly related to combustion properties. Considering these, co-combustion of hydrochar with coals is investigated in many works towards industrial application. Low ignition temperature and high combustion

reactivity of hydrochar make the co-combustion attractive. However, binders will affect the burnout temperature of hydrochar pellet, and this depends on binder species. For example, molasses enhances the combustion process can delay combustion with a higher final temperature. Thus, binder should be used based on the combustion requirement. Muthuraman et al. [56] compared the co-combustion characteristics of Indian coal with MSW derived hydrochar and stated that the devolatilization properties of coal could be improved by blending hydrochar, and the ignition temperature was significantly reduced, which became more prominent with increasing the blending ratio of hydrochar resulting from its high volatile matter content. Additionally, single biomass hydrothermal carbonization may not be able to achieve satisfactory combustion results. Mixture of different biomass for HTC can often improve the combustion performance of hydrochar. For example, Lang et al. [57] found that the combustion weight loss in the second stage was significantly higher than that of pig manure due to the presence of more volatile substances in straw, and the complete combustion time was shorter. However, pig manure has a higher combustion temperature with metal oxides and inorganic minerals. After mixing the two biomasses for HTC, the combustion performance and stability of the mixture were improved.

Meanwhile, hydrochar has showed the cleaner combustion properties than that of feedstock. As it proved, inorganic elements can be released into liquid product and low ash can be achieved [58]. As for N, S, and Cl, these elements also undergo same trend but the forms in hydrochar which is directly to gas release during combustion. In He et al.'s work [59], denitrification is considered to be main reaction happened in HTC of sludge, and it showed the nitrogen was mainly released in liquid products as ammonia nitrogen, while nitrogen in hydrochar changed to stable form like pyridines-N. For sulfur, many works have showed that more inorganic forms of S in hydrochar was observed compared to feedstock, which will also reduce the SO_2 during combustion [2]. Since Cl has smaller atomic radius than that of N and S, most works reported that approximately 90% can be released into liquid phase, and washing process can also help for the reduction of Cl in hydrochar [11]. Based on these, HTC has showed the advantage for cleaner biofuel production, but the transformation and balance of these elements in HTC products still need further works.
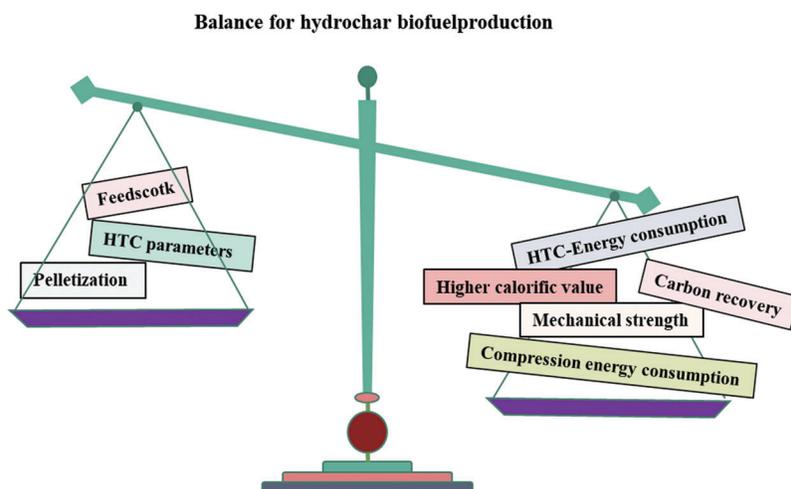
Figure 6: A simple balance for pelletization of hydrochar as solid biofuel production

3 Conclusion and Future Perspectives

In this paper, hydrothermal carbonization of biowaste for solid biofuel was reviewed about the dewatering properties, elemental composition, pelletization and combustion. The investigation on

hydorchar as biofuel is limited to the optimization of hydrothermal carbonization parameters, the control of physical characteristics of fuel after pelletization, and the study of optimal combustion characteristics. Therefore, various balance controls like mass and energy balance need to be noted, which are common to researchers working on hydrothermal carbonization technologies. For preparation of solid fuel from hydrochar, future works should also pay attention to the goal of high calorific value, easy pelletization, stability of combustion, cleaner production of hydrochar. A simple balance for hydrochar fuel production is presented in Fig. 6 and future challenges and possible research hotspots are as follows:

(1) Instu-analysis of the carbonization process of biomass with time and temperature should be cared instead of solo analysis of the hydrochar;

(2) The mechanism involved in hydrochar pelletization like solid-physics and models should be got more attention because this benefits the decrease of energy consumption during pelletization;

(3) The inter-effect of various feedstock derived hydrochar during pelletization should be investigated in future with the binders since single biowaste may not conducive to pelletization with high mechanical strength;

(4) Recycle of liquid products for HTC is essential, and it should also be considered for recycle optimization and coalification of hydrochar.

Conflicts of Interest: The authors declare that they have no conflicts of interest to report regarding the present study.

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