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Improving the energy-related aspects of biowaste treatment in an experimental hydrothermal carbonization reactor

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Abstract
Hydrothermal carbonization (HTC) is a thermochemical conversion process with the potential to treat the prevalent wet urban biowaste in low- and middle-income countries. The generated hydrochar solids are a hygienic, homogenized, carbon rich and energy dense product with economic value that can be used as an alternative to wood-based charcoal or fossil fuel. Obtaining a satisfactory energy efficiency of the process is, however, one of the prerequisites for the possible breakthrough of this technology. In an experimental HTC reactor, a model kitchen/market waste feedstock (17.8 MJ/kgdb) was hydrothermally carbonized with varying loading rates (TS 20 and 25 %) under mild operational conditions with peak temperatures of 160–190 °C and process times of 2–10 h above 160 °C. The aim was to evaluate the energy ratio of the process under these conditions while examining the impact on the hydrochar quality. Results show that the chemical properties of the produced hydrochar and its heating value were of moderate quality (21.1–24.4 MJ/kgdb), showing similar characteristics like torrefied products. HTC of a 25 % TS-load during 2 h at 180 °C and maximum pressure of 18.3 bar resulted in a char chemical output energy that is twice as high as the electrical energy consumed in the process. If considering the theoretical methane potential of the process water, the energy ratio could be increased to 2.6; while reactor insulation could further enhance this ratio to 3. This article reveals the merits of mild HTC and provides relevant knowledge for attaining an optimized, energy efficient HTC system.

Keywords: Organic waste, energy efficiency, energy ratio, HTC, hydrochar, thermochemical conversion

1. Introduction

Solid waste management (SWM), which aims at protecting human health, preventing environmental degradation and recovering valuable resources, is an essential utility service and a key global challenge of the 21st century, particularly in urban areas [1-2]. Provision of equitable and reliable waste management services remains especially difficult in low- and middle-income countries, a fact that is reflected by low collection rates and widespread evidence of inadequate disposal methods [3-5]. Given the high organic waste fraction in these countries, often constituting more than 50% of the total waste generated [4,6], dysfunctional SWM systems exert adverse impacts on human health, local and global environment, and social and economic development [7-10]. This situation has triggered different research on finding appropriate treatment and valorization technologies for biowaste in cities of low- and middle-income countries [e.g., 11-14]. The underlying idea is to process organic waste such as food scraps and peeling residues from household/canteen kitchens and vegetable/fruit markets into a hygienic product with economic value and market demand [15]. Realizing such environmental and socio-economic benefits through treatment and recycling could then stimulate waste collection, and contribute to the paradigm shift from ‘solid waste management’ to ‘resource management in a circular economy’ [2].

Although hydrothermal carbonization (HTC) has been known for over a century ([16] Bergius, 1913), interest in using it as a method to transform biowaste into a stable, sterile and valuable product has noticeably increased only in the last decade [17-19]. During the thermochemical HTC process the biomass is submerged in water in a pressure vessel and typically heated to 180-250°C for several hours [20]. Gas generation during the reaction increases the pressure to 20-35 bar while the water remains in a liquid state. The exothermal process lowers the oxygen and hydrogen content of the feed (described by the molecular O/C and H/C ratio) mainly through dehydration and decarboxylation [21]. A major advantage of the HTC technology is its high feedstock flexibility in form, composition and moisture content, thus not requiring an energy-intense drying step before processing [21,25]. HTC results primarily in a solid phase enriched in carbon, named hydrochar or char, a liquid phase with dissolved organic compounds (process water) and a small quantity of a gas phase mainly composed of carbon dioxide and trace amounts of other gases [22]. The resulting hydrochar can be dried and used for a range of different applications, such as fuel, functionalized carbonaceous material (activated carbon) or as soil amendment to increase soil fertility while providing a long-term carbon sink [24]. As hydrochar shows enhanced dewaterability properties (low hydrophilicity), high bulk density, and higher heating value similar to coal (20-30 MJ/kg), the focus of the presented study is on the use of hydrochar as solid fuel substituting wood-based charcoal [19, 20, 23].

Prior to the study presented here, batch experiments with canteen foodwaste were conducted to assess the operational process parameters in an experimental HTC reactor [26]. The experiments with varying loading rates (Total Solids (TS) of 2.5 - 15.2%) over a reaction time of 10 hours (minimum 4 hours above 180°C in the reactor) revealed satisfactory char characteristics, with average C-content 66.8% of dry weight and average higher heating value (HHV) of 29.1 MJ/kg_{db} compared to 19.3 MJ/kg_{db} of the initial feedstock. However, comparison of the electrical energy consumed for reactor heating (11.4 kWh) with the energy content of the resulting hydrochar disclosed unfavorable energy ratios below 1. Only the experiment with highest loading rate (15.2% TS) resulted in a char energy content that exceeded the amount of energy consumed during the reaction. Additional experiments with a constant loading rate of 4.9% TS revealed that the char characteristics remain satisfactory (HHV 29.3 MJ/kg_{db}) even in experiments where the high heating temperature (HHT) was reduced to 170°C [26]. Overall, the preliminary study showed that loading rate and temperature are the parameters that mainly influence the energy ratio of the HTC process. Thus these two parameters were varied in the present study.

In general, the HTC process is mainly governed by temperature, biomass-to-water ratio, residence time, pressure, feedstock composition and pH [20]. These parameters directly or indirectly also influence the energy efficiency of the process. The biomass-to-water ratio, generally expressed as loading rate (% TS), has an impact on the reaction and the energetic aspects of the process. Lower loading rates for instance imply more water, which leads to increased energy demand for heating due to the high specific heat capacity of water, and results in a lower total energy output per batch. Residence time influences energy consumption but also affects the carbon content in the char and thus its heating value. The peak temperature during the process, called high heating temperature (HHT), indicates the maximum reached process temperature, which not only affects the electricity consumption required for heating, but has also a major impact on product yield and characteristics. Higher temperatures lead to higher reaction rates and decisively influence the amount of biomass compounds that can be hydrolyzed. Substantial hydrolysis starts at a temperature of 180°C [21]. This is represented by the semi-empirical equation of the reaction severity f ($f = 50 \times t^{0.2} \times e^{-\frac{3500}{T}}$), a formula defined to model the influence of HHT (as T [K]) and residence time (t [s]) on the products, whereby t stands for the duration above 180°C [21, 27]. The higher the reaction severity, the higher the carbon content and heating value of the hydrochar produced. However, there is concern about temperature and residence time being exchangeable parameters and that the achievable coal is defined primarily by the reaction temperature [21]. In addition, some reports also mention carbonization taking place below 180°C [26,28], which could have a positive impact on the energy ratio of the process.

The objective of the study presented here was to improve the energy-related aspects of biowaste carbonization in an experimental HTC reactor. The operational parameters such as loading rate (TS), high heating temperature (HHT) and residence time were varied and their influence on char product quality and energy ratio and energy efficiency examined. The energy aspects were further assessed by including the theoretical energy (methane) potential of the process water and by integrating the calculated energy saving effect of reactor insulation using simplified heat-loss experiments.

The focus of this article lies on the energy aspects, solely considering the energetic efficiency of the HTC process and not of the complete biowaste-to-energy conversion chain. Other aspects that critically influence the applicability of HTC technology as biowaste valorization method in low- and middle-income countries, such as environmental, financial and safety issues are not discussed in this article.

2. Materials and methods

2.1 HTC reactor

The experimental HTC reactor with a capacity of 21.8 L (17 L liquid, 4.8 L gas volume) and weight of 67.7 kg is made of stainless steel and operated in a batch-feeding mode without stirring device (Figure 1; details in [29]). The reactor is certified for a gauge pressure limit of 30 bar and a maximum temperature of 300°C (European Pressure Equipment Directives 97/23/EC). An electric heating mantle (max. 2.5 kW), controlled by a temperature regulator (HT42-30P, Hilesheim GmbH), provided the heat supply. An energy meter (model VSM-120) recorded the energy consumption every 3s. External reactor temperatures were measured at four locations by K-type thermocouples to assess the temperature distribution around the reactor and accurately plan the insulation measures. The lid contains two sensors for measuring and recording of inner temperature (W120.3, Roth&Co AG) and inner pressure (Leo Record Keller AG). These sensors register values every 20 s and transfer the data via USB cable to the computer. Also located on the lid are an overpressure valve (SV510, Spyrax Sarco AG) and a gas drain valve (AV243, Spyrax Sarco AG) to release residual pressure at the end of the reaction. The reactor lid's screws were tightened with a torque wrench at 84 Nm. A graphite-sealing ring between flange and lid ensures hermetic closure of the reactor.

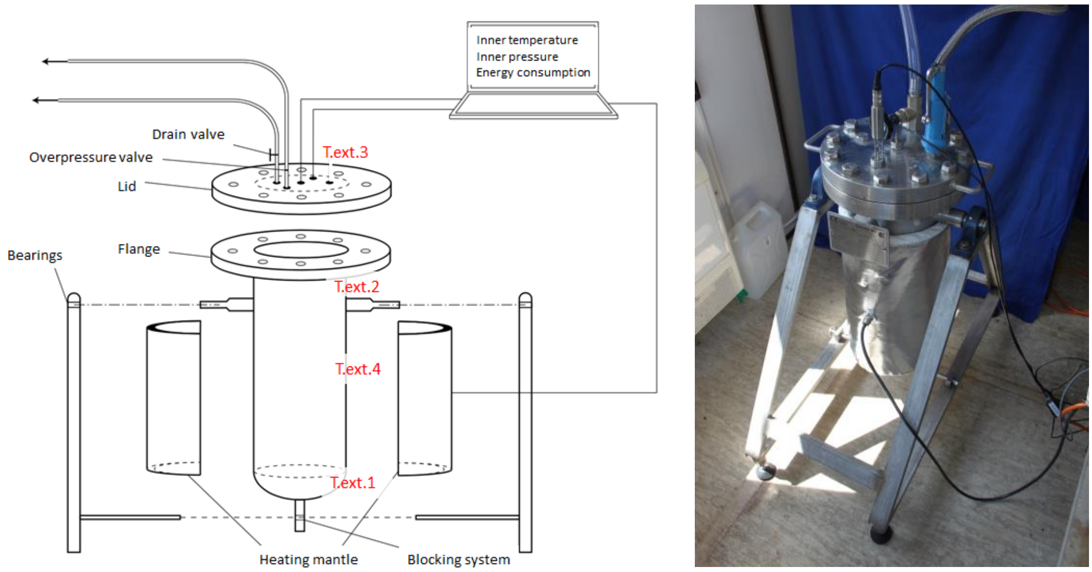

Fig. 1: Schematic and photo of the HTC reactor used [29] with inner temperature and pressure measurements, external thermocouples, and drain and overpressure valve placed on the lid

The feeding procedure was conducted according to the instructions described in [26]. During the reaction time, inner temperature and pressure were continuously monitored. The temperature was reduced when the inner pressure approached the reactor pressure maximum of 30 bar (gauge). After a completed reaction, the reactor was left to cool down for roughly 12 hours. Solid and liquid end products were separated by manual filtration and samples of both phases taken for triplicate analyses. If applicable, based on visual classification, the hydrochar was sorted into three categories of different carbonization degrees before being analyzed.

2.2 Feedstock

A model feedstock was designed with the goal to simulate common kitchen or market biowaste in low- and middle-income countries . Such wastes typically contain starchy, fibrous and cellulose-rich constituents, such as potato, banana,, cabbage, maize and rice. Thus, the following ingredients were selected: potato peeling, banana peeling, potato, cabbage, full maize pellets and rice, and mixed in a ratio of 1:2:4:6:8:8 (on wet basis). This resulted in a mixed feedstock with a TS content of 55%. Water was added to this concentrated feedstock to obtain the desirable loading rate (% TS) for each batch. Details of the biowaste feedstock components used in all experiments are presented in Tab 1.

Table 1 Characteristics of the feedstock ingredients

				Ultimate analysis				*Proximate analysis*		
Ingredients	**Fraction (% ww)**	**TS (%)**	**HHV (MJ/kg)**	**C (%)**	**H (%)**	**N (%)**	**O (%)**	**Ash (%)**	**VM (%)**	**FC (%)**
Potato peeling	3.4	14.8	17.1	40.6	6.6	2.0	53.7	6.5	78.0	15.5
Banana peelings	6.9	10.8	17.7	42.8	5.9	1.3	49.6	11.2	69.5	19.3
Potato	13.8	16.4	17.2	41.5	6.7	1.5	59.5	3.8	80.3	15.9
Cabbage	20.7	9.7	18.0	42.4	6.3	1.9	53.4	6.9	73.2	19.9
Rice	27.6	89.1	17.5	43.0	7.1	1.2	62.2	0.5	89.5	10.0
Maize pellets	27.6	90.7	18.0	42.8	5.9	1.3	49.6	3.5	80.3	16.2
Mixed feedstock	**100.0**	**55.0**	**17.8**	**44.6**	**6.9**	**1.3**	**56.3**	**2.0**	**83.0**	**15.0**

All ingredients were weighted, cut and homogenized with a kitchen blender. To avoid biological degradation, the feedstock was prepared on the day of the carbonization experiment.

2.3 Experiments and analyses

Based on the results of [26] and on literature stating that a total solid (TS) content of at least 15-20% is required to operate a hydrothermal processes in an economic manner [30], the batch experiments were conducted with a TS of 20% and 25%. Maximum temperatures of 160-190°C were targeted with different residence times (2-10 h) above 160°C (Table 2). The reactor was filled with a volume of 17L per batch, resulting in a chemical energy input of 60 MJ for TS 20% and 76 MJ for TS 25%.

Table 2 Overview of experiments with operational parameters

Exp. #	**Loading rate** (% TS)	**High Heating Temperature** (°C)	**Time above 160°C** (h)	**Max. pressure (gauge)** (bar)	**Name used in figures of this article** (Remarks)
1	20	160	10	16.8	TS20_HHT 160 (>160°C = 10h) 16.8 bar
2	20	190	8	28.9	TS20_HHT 190 (>160°C = 8h) 28.9 bar
3	25	160	4	28.8	TS25_HHT 160 (>160°C = 4h) 28.8 bar
4	25	170	5	28.1	TS25_HHT 170 (>160°C = 5h) 28.7 bar
	25	170	5	29.1	Duplicated experiment (mean value used)
5	25	170	3	16.2	TS25_HHT 170 (>160°C = 3h) 13.7 bar
	25	170	3	11.1	Duplicated experiment (mean value used)
6	25	180	2	18.2	TS25_HHT 180 (>160°C = 2h) 18.2 bar
7	25	180	4	28.6	TS25_HHT 180 (>160°C = 4h) 28.6 bar

Total solids content of the feedstock and the resulting hydrochar were analysed according to ASTM E1756-08. Proximate analysis was conducted with a muffle furnace and precision scale (volatile matter: DIN 51720; ash content: DIN 51719; fixed carbon: determined by difference), while the composition of C, N, H and O was analysed with the elemental analyser LECO Truspec. Higher heating value was analysed with IKA C1 according to DIN EN 14918. During the reaction the inner temperature (sensor W120.3L), pressure (Leo Record Keller AG), and external reactor temperatures were measured (4 thermocouples K-type) and recorded (PICO TC-08 data logger), as well as the electricity consumption documented (Voltcraft VSM-120). The process water was analyzed in terms of pH and electrical conductivity (Hach-Lange HQ D40). TOC was analyzed by Shimadzu TOC-L 720°C catalytic combustion.

2.4 Energy calculations

Energy flows

Input energy flows into the reactor comprise biowaste chemical energy and electricity consumption. Output energy flows include the chemical energy of hydrochar and the chemical energy of process water. For simplification and better visualization the heat losses are also included in the output flows. Chemical energy flows of biowaste, process water and hydrochar are calculated based on their analyzed specific higher heating value (HHV) and mass/volume. Electricity consumption was measured and recorded every 3 s.

The energy potential of the process water is derived from two calculations. First the methane yield of the measured carbon in the process water is determined, and secondly the energy content of the methane is calculated:

i) *Molar calculation*

The methane yield of the process water is calculated based on the measured TOC content by using the following chemical equation:

$$2C + 2H_2O \rightarrow CO_2 + CH_4 \qquad \Delta H = 103.341\ \mathrm{kJ/mol} \tag{1}$$

which states that when two moles of carbon (= TOC) react with two moles of water, one mole of carbon dioxide and one mole of methane are produced. It is assumed that carbon (TOC) in process water, although being present in different organic molecules, exists at a leveled oxidation degree of 0. It is further assumed that the complete organic content is transformed into biogas, which is mainly comprised of methane (CH_4) and carbon dioxide (CO_2). Considering the atomic weights of carbon (12.0107 g) and of one mole of water (18.0152 g), and the fact that one mole of any gas has a volume of 22.4 L at standard temperature and pressure, equation 2 is derived:

$$24.0214g\ TOC + 36.0304g\ H_2O \rightarrow 22.4L\ CO_2 + 22.4L\ CH_4 \tag{2}$$

Thus 24.0214 g C produce 44.8 L of biogas, which means that 1 g C (= TOC) produces 1.865 L biogas. Based on the rather conservative estimation that biogas is comprised of 50% methane (CH_4), 1 g C (= TOC) produces 0.9325 L of methane (CH_4).

ii) *Energy calculation*

The energy content of methane, which can be extracted from the process water, is calculated taking into consideration the heating value and density of CH_4 at ambient conditions.

$$Energy\,potential_{\,process\,water} = Energy\,content_{\,CH4} = 0.0363\ MJ/L_{CH4} \tag{3}$$

Heat losses include the heat capacity and heat emission of the reactor system. Heat capacities of the reactor shell (0.5 kJ/kg*K), biowaste (1.6 kJ/kg*K), process water (4.25 kJ/kg*K) and hydrochar (1.45 kJ/kg*K;) were calculated based on [31-33]. Heat emission was determined by six simplified heat-loss experiments. For this purpose, the reactor was filled with water and stabilized at different temperature levels (50 to 200°C in steps of 25°C). The electrical power consumption, which corresponds to energy losses (=heat emissions) in a stable system, were measured at these different temperature levels for an insulated and a non-insulated system. The insulation consisted of a top, lid, reactor body and bottom insulation part. It was made of a thin layer of needled glass wool coated with vermiculite (FIRETEX GFF-FV1 1000 x 600 x 6 mm), fixed to temperature-resistant foam by high-temperature silicone (300°C). The heat loss experiments revealed that through reactor insulation in average 40% of the heat emissions can be avoided. The value of 40% power reduction is used in the calculations of ER_3 and η_3.

Energy ratio (ER) and energy efficiency (η)

Two energy parameters that describe the usable energy output with the energy input were examined. As these parameters strongly depend on the system boundaries and the presumed conditions, their formulas are described in detail.

The Energy ratio (ER) is defined as the energy generated divided by the (electrical) energy consumed during the reaction. An ER above 1 implies that the process produces more usable energy than it consumes. This ratio assumes that the chemical energy stored in the biowaste can be neglected, as the feedstock is a waste material and obtained for free. The following three ERs were determined:

$$ER_1 = \frac{Energy_{\,char}}{Electricity\ consumed} = \frac{m_{char,dry} * HHV_{db}}{EL_{consumed}} \tag{4}$$

$$ER_2 = \frac{E_{\,char} + E_{\,process\,water}}{Electricity\ consumed} = \frac{m_{char,dry} * HHV_{db} + TOC_{process\,water} * E\,(CH_4)\,potential_{TOC}}{EL_{consumed}} \tag{5}$$

$$ER_3 = \frac{E_{\,char} + E_{\,process\,water}}{EL_{\,consumed} - E\,reduction_{insulation}} = \frac{m_{char,dry} * HHV_{db} + TOC_{process\,water} * E\,(CH_4)\,pot._{TOC}}{EL_{consumed} - EL\,red._{insulation}} \tag{6}$$

ER_1 solely takes into consideration the chemical energy of the hydrochar. After the HTC process is completed, water containing a significant amount of chemical as well as thermal energy, leaves the reactor. The former one is in the form of dissolved organic compounds, the latter one as heated water. ER_2 also includes the chemical energy (methane potential) of this process water. ER_3 additionally integrates the calculated minimization of heat losses as a result of reactor insulation.

The energy efficiency η on the other hand is defined as the ratio of useful energy output to total energy input, but contrary to ER also includes the chemical energy stored in the feedstock. The η in biomass conversion processes is always below 100% due to energy (heat) losses.

$$\eta_1 = \frac{Energy_{\,char}}{Electricity\ consumed + Chemical\ energy_{Feedstock}} = \frac{m_{char,dry} * HHV_{db}}{EL_{consumed} + E_{feedstock}} \tag{7}$$

$$\eta_2 = \frac{E_{\,char} + E_{\,process\,water}}{EL\ consumed + Chem.\ energy_{Feedstock}} = \frac{m_{char,dry} * HHV_{db} + TOC_{process\,water} * E\,(CH_4)\,potential_{TOC}}{EL_{consumed} + E_{feedstock}} \tag{8}$$

$$\eta_3 = \frac{E_{\,char} + E_{\,process\,water}}{EL_{\,consumed} - E\,reduction_{insulation} + Chem.energy_{Feedstock}} = \frac{m_{char,dry} * HHV_{db} + TOC_{process\,water} * E\,(CH_4)\,pot._{TOC}}{EL_{consumed} - EL\,red._{insulation} + E_{feedstock}} \tag{9}$$

3. Results

3.1 Char yield and quality

The hydrochar produced in all experiments showed heterogeneous degrees of carbonization. This is probably caused by the uneven heat distribution in the unstirred reactor. As an example, Figure 2 presents the different temperatures measured in different locations (see Figure 1) during the experiment 3 where the max. reached pressure was 28.8 bar. The maximum set temperature of the heating mantle (250°C), the max. temperature inside the unstirred reactor (164°C) as well as the external temperatures of the reactor bottom (T.ext.1; max. 114°C), upper reactor wall (T.ext.2; max. 138°C), outside reactor lid (T.ext.3; max. 127°C) and between heating mantle and reactor wall (T.ext.4; max. 195°C) reveal the uneven temperature distribution, which caused the different hydrochar qualities.

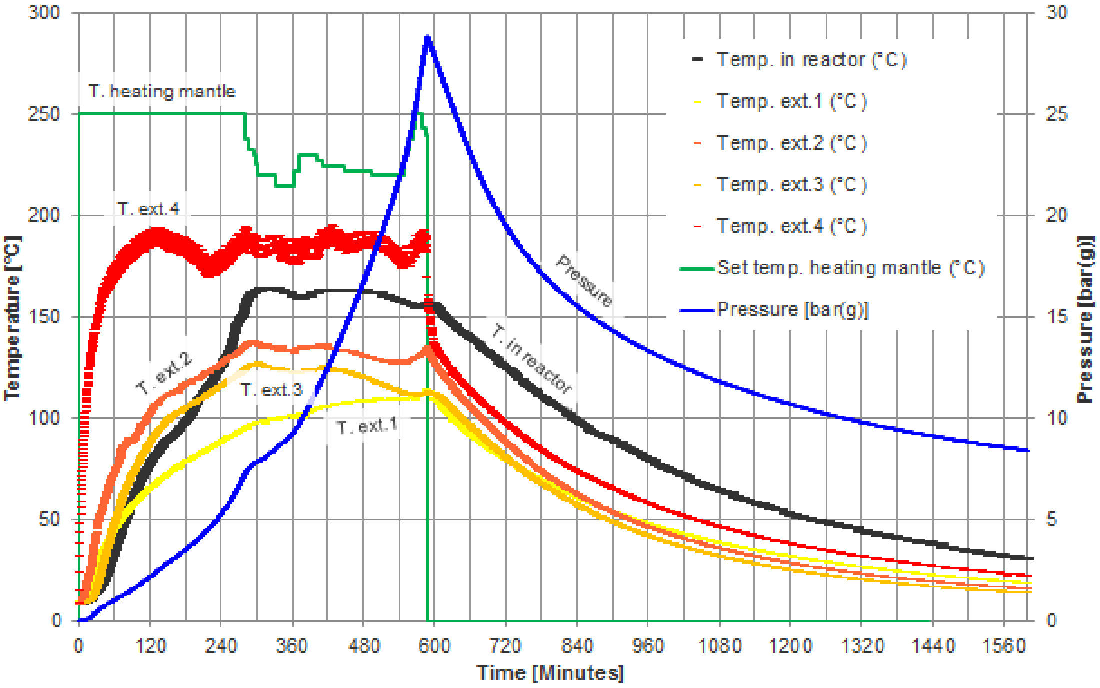

Fig. 2 Temperature and pressure profile of experiment 3 (TS25_HHT 160 (>160°C = 4h), 28.8 bar)

The hydrochar was thus sorted based on visual color classification into three carbonization categories. Char of category 1 (cat.1: carbonized): black, brittle hydrochar, cat. 2 (partially carbonized): dark-brown, viscid hydrochar mainly sticking to the reactor wall, and cat. 3 (hardly carbonized): brown, very sticky material found on the reactor bottom. The analyses of these categories show different carbon contents and higher heating values. Figure 3 presents the average distribution of the carbonization categories based on the total mass of dried hydrochar and their mean higher heating values.

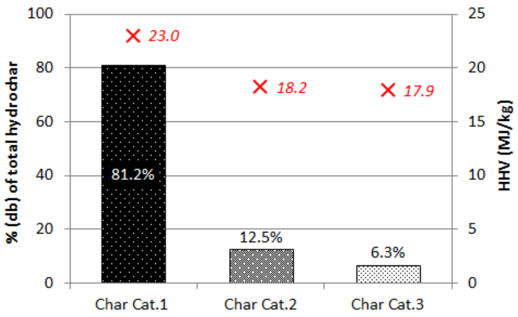

Fig. 3 Distribution and heating value of different char categories (mean values of all experiments)

The hydrochar of category 1 is considered the most interesting solid product due to its highest energy content. The hydrochar yield (char/biowaste*100; [34]) of each experiment is presented in Figure 4. The average char yield of cat. 1 is 47.9% (db).

Figure 5 and 6 present the differences in composition (proximate analysis) and heating value of the hydrochar from the experiments conducted with loading rates of 20% and 25% TS and varying operational parameters such a HHT, period above 160°C and resulting reactor pressure.

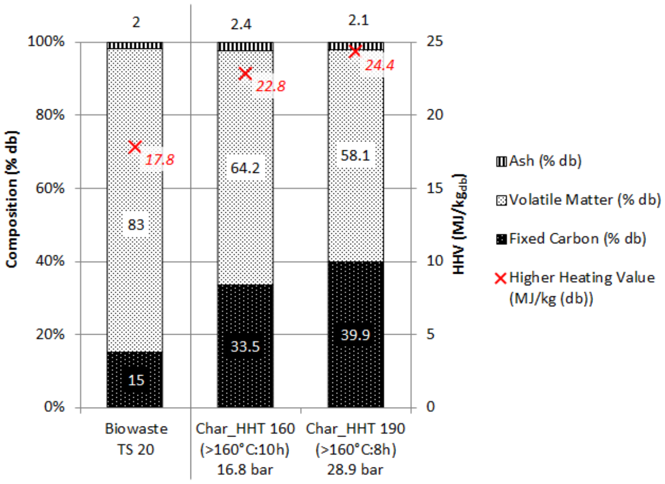

Fig. 5 Biowaste and hydrochar (cat. 1) characteristics of experiments with loading rate TS 20%

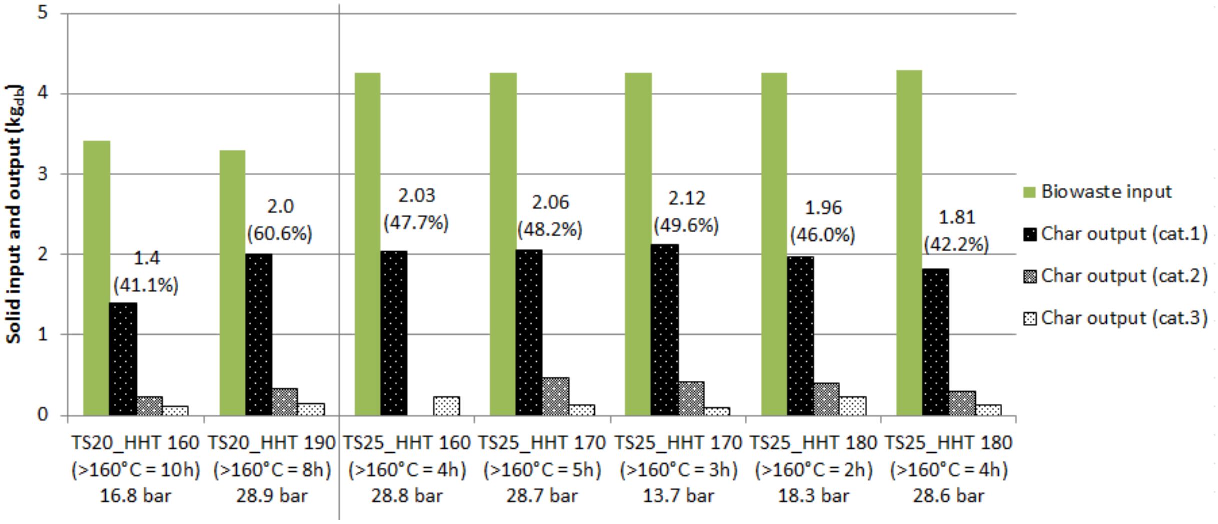

Fig. 4 Hydrochar yields (on dry basis) of all experiments

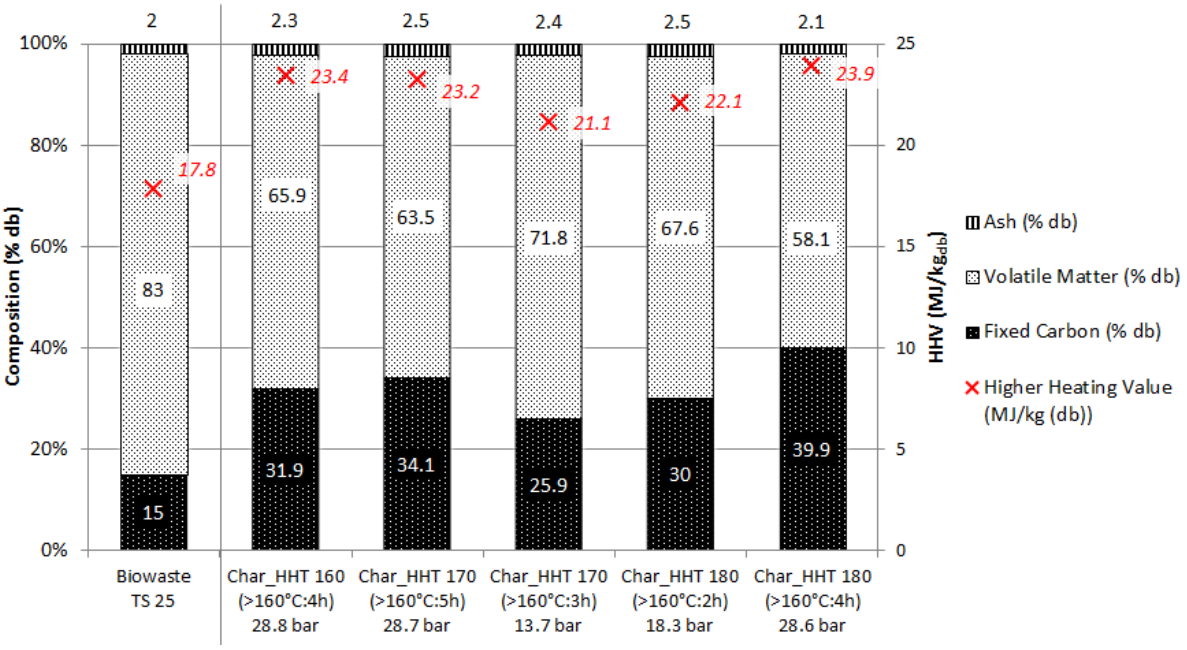

Fig. 6 Biowaste and hydrochar (cat. 1) characteristics of experiments with loading rate TS 25%

3.2 Energy flows and other energy-related aspects

Figure 7 illustrates the distribution of chemical energy measured in hydrochar and in the process water (methane potential based on TOC content). In average 84% of the total chemical energy in the solid and liquid phase is found in the hydrochar, and 16% in the process water.

The electricity consumption among all experiments (Exp.) varied from 26.6 MJ (Exp. 6) to 36.9 MJ (Exp. 2). Exp. 2 produced higher total energy stored in hydrochar (56.9 MJ) compared to 54.8 in Exp. 6. On the other hand, the higher HHT in Exp. 2 resulted in low potential energy found in the process water (4 MJ compared to 13.8 MJ in Exp. 6). Figure 8 presents the energy flow diagrams of these two experiments. The difference between total energy input and total energy output (marked as undef.) is presumably caused by inaccuracies of the electricity meter or in heat loss calculation and due to the lack of chemical energy measurement of the gas phase.

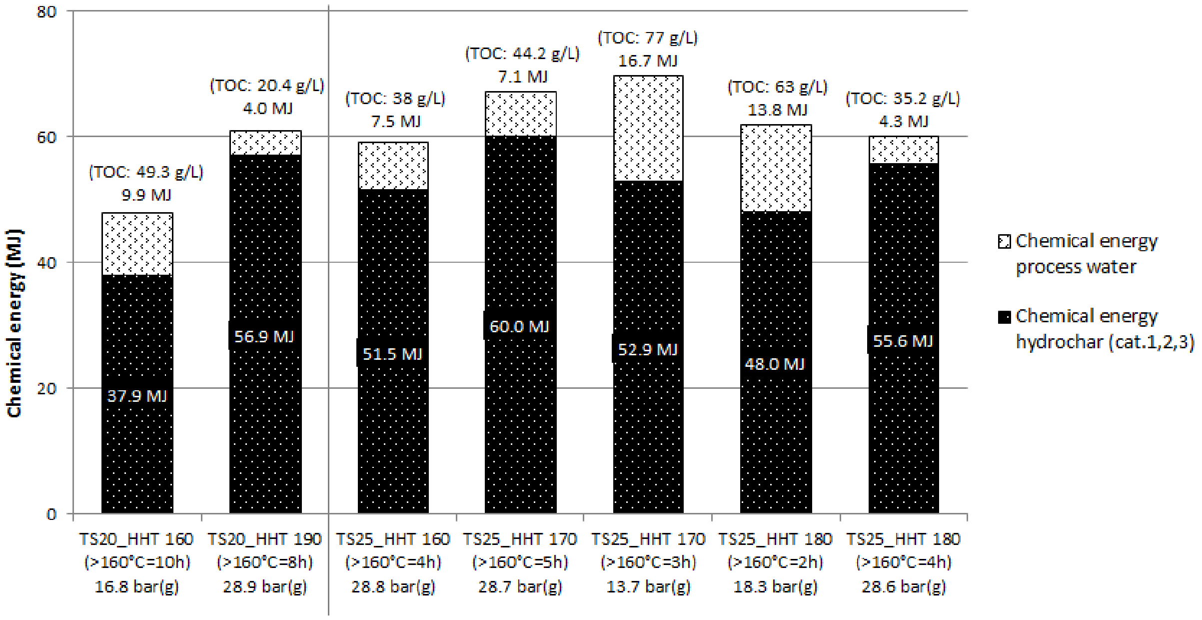

Fig. 7 Distribution of chemical energy in solid and liquid HTC products

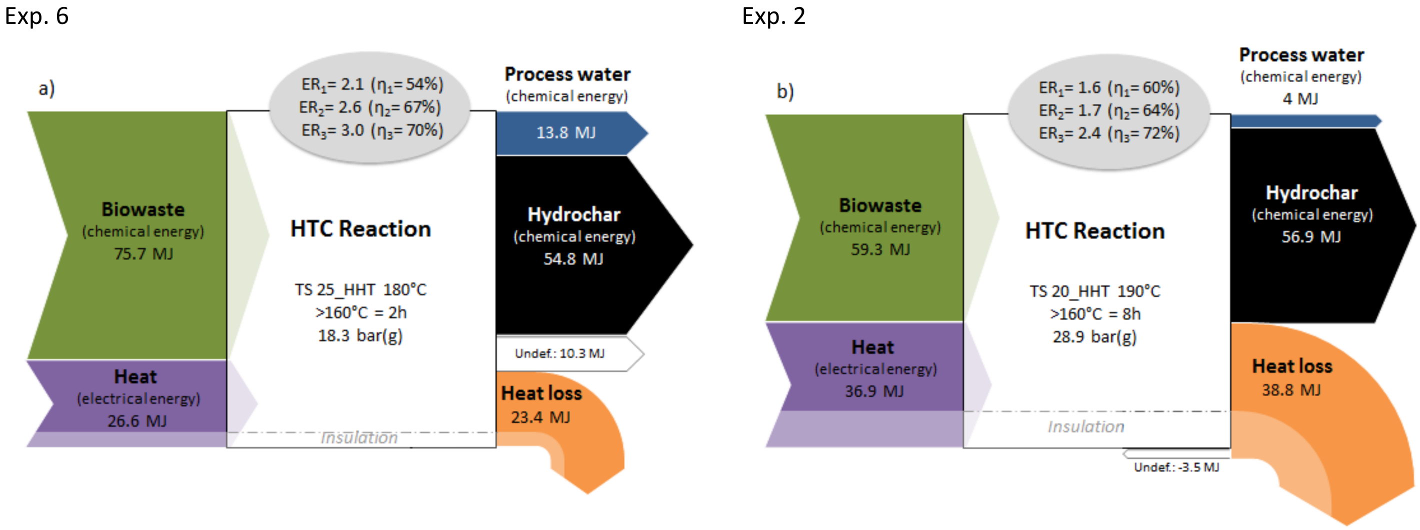

Fig. 8 Energy flow diagram of experiments with highest energy ratios (ERs; exp. 6, left) and highest energy efficiencies (η; exp. 2, right)

The diagram of figure 8a illustrates that the highest energy ratios are obtained at low pressure (18.3 bar; exp. 6) and short process duration. Using the methane potential in the process water results in a substantial increase of the ER_2. Results further show that enhanced reactor insulation leads to a reduced heat input due to reduced heat losses (visualized by the pale coloring and the insulation line), with an improved ER_3 as consequence. Figure 8a shows an ER_3 of 3.0 for the experiments with 25% TS at 180°C and max. 18.3 bar. This means that the theoretically usable energy after carbonization is 3 times higher than the electrical energy used for the HTC process. The experiment which shows the highest energy efficiency η was conducted with 20% TS at 190°C and high pressure of 28.9 bar (exp.2). This energy efficiency can be improved from 60% (η_1) to 64% (η_2) when the energy content of the process water is used and to 72% (η_3) if heat losses are minimized.

As shown in Table 3, all experiments revealed positive energy ratios (ER_1 = 1.04 – 2.06) when considering char of cat. 1, 2 and 3. Highest overall energy ratio was achieved at 180°C at relatively low pressure and short period (Exp. 6). However, the highest energy efficiency can be observed at higher temperature, pressure and time (Exp. 2).

Table 3 Energy ratios and efficiencies of all experiments

Exp. #	Operational parameters	ER_1	ER_2	ER_3	η_1	η_2	η_3
1	TS20_HHT 160 (>160°C = 10h) 16.8 bar	1.04	1.32	1.78	39%	49%	55%
2	TS20_HHT 190 (>160°C = 8h) 28.9 bar	1.55	1.66	2.35	**60%**	**64%**	**72%**
3	TS25_HHT 160 (>160°C = 4h) 28.8 bar	1.61	1.84	2.28	48%	55%	58%
4	TS25_HHT 170 (>160°C = 5h) 28.7 bar	1.78	2.00	2.56	53%	60%	64%
5	TS25_HHT 170 (>160°C = 3h) 13.7 bar	1.59	2.10	2.63	48%	64%	68%
6	TS25_HHT 180 (>160°C = 2h) 18.2 bar	**2.06**	**2.58**	**3.01**	54%	67%	70%
7	TS25_HHT 180 (>160°C = 4h) 28.6 bar	1.47	1.59	2.04	46%	50%	54%

4. Discussion

All HTC experiments conducted with loading rates of 20% and 25% TS under mild operational conditions in terms of temperature (HHT 160-190°C) and time (2-10h above 160°C) exhibited favorable energy ratios and efficiencies while producing hydrochar of moderate quality.

Different hydrochar qualities

The total hydrochar yield on dry weight basis was between 50.4 and 74.8% (average 59.2%), which is in line with results from other studies (50-80% [25]). However, due to uneven heat distribution the hydrochar showed heterogeneous quality (cat. 1, 2, 3). In unstirred reactors, high loading rates lead to feedstock compaction and further enhance uneven carbonization. At higher process temperatures the solid products of the HTC reaction would in general be more homogeneous, dense and uniform [20]. It is recommended to further substantiate the differences in char qualities, which were observed visually and determined by analysis of the heating values, This can be done by providing SEM images or investigating the particle shape, or pore distribution of the different char categories. Only considering the best char quality (cat.1), the hydrochar yield was in the range of 41.1 and 60.6% (average 47.9%). With an average of 61.5% (range: 52.4 - 83%) of the initially present carbon retained in the hydrochar (cat. 1), the carbon recovery [(%C in char*char mass/%C in feedstock*dry feedstock mass)*100], according to [35]) of this study is comparable with batch carbonization experiments found in literature (46-75% [19,36]).

Proximate analysis

The HTC process decreased the volatile matter (VM) content of the initial biowaste feedstock, resulting in an increase of the fixed carbon (FC) weight ratio. VM in hydrochar of all experiments remained above 58%, which is in the range of findings from an HTC study of Asian municipal solid waste (58-71% [37]). Higher VM facilitates ignition of the char, but at the same time char with higher VM is more hygroscopic and burns less cleanly [38]. Taking into consideration that charcoal intended for domestic cooking typically contains 20-30% volatile matter, with a value of 40% being marginally acceptable [39], this produced hydrochar is considered unsuitable for direct combustion in cooking stoves. Thus combustion in industrial burners or incineration plants, equipped with adequate pollution emission control systems need to be considered. With an average of 2.3% the ash content in the produced hydrochar is similar to that of good-quality charcoal (0.5-5% [40]). Limited information is available in literature regarding the fate of metals and nutrients during HTC, which depends on many factors, including reaction time and temperature, process water pH, and feedstock properties [41] (Berge at al., 2015). A better understanding, particularly on the impact of Ni, Na, Ca and Cl on the mechanism of char formation is needed. Addition of ionic salts is known to reduce the HTC reaction pressure, which has an impact on the product quality and the energy ratio of the process. However, in a large-scale process, any cost advantages from reduced pressure may be offset by increased corrosion rates resulting from addition of a chloride salt [42] (Lynam et al, 2012).

Table 4 compares the average results of this study (TS 20/25%, HHT 160-190°C, 2-10h) with HTC experiments using foodwaste (TS 20%) under more severe operational conditions (HHT 250°C, 20h [19]). Substantial differences can be seen in ash and oxygen content. Ash content in the present study is low due to the pure model feedstock with no inorganic contaminants such as salts. Oxygen content is higher presumably due to restricted decarboxylation as a result of the lower carbonization temperature and time. Lower HHT is likely also the reason for the higher char yield, lower energy densification ratio and lower HHV in this study. An explanation for this is that higher temperature and hence pressure leads to higher carbon content of the hydrochar, even though the total solid yield is reduced [20].

Table 4 Comparison of foodwaste HTC in this study with results of [19]

	Literature [19]		*Present study (average)*	
	Input (Foodwaste)	**Output** (Hydrochar)	**Input** (Foodwaste)	**Output** (Hydrochar, cat.1)
VM ($\%_{db}$)	77.6	53.4	83	64.2
FC ($\%_{db}$)	14.8	29.7	15	33.6
Ash ($\%_{db}$)	7.5	11.2	2	2.3
C ($\%_{db}$)	42.5	67.6	44.6	57.2
H ($\%_{db}$)	5.8	5.8	6.9	5.8
N ($\%_{db}$)	3.2	4.6	1.3	1.8
O ($\%_{db}$)	40.8	9.9	56.3	38.2
HHV (MJ/kg_{db})	18.1	29.1	17.8	23.0
Char yield ($\%_{db}$)		43.8		47.9
Energy recovery ($\%_{db}$)[a]		70.3		61.5
Energy densification (ratio)[b]		1.82		1.29
Energy yield ($\%_{db}$)[c]		79.7		62.0

[a] (HHV of char * char mass) / (HHV of feedstock * dry feedstock mass) * 100 [43]
[b] HHV hydrochar / HHV feedstock [20]
[c] Char yield * energy densification ratio [44]

Higher heating value (HHV)
The applied mild HTC process conditions increased the HHV of the biowaste from 17.8 MJ/kg_{db} to hydrochar values (cat. 1) between 21.1 MJ/kg (+19%) and 24.4 MJ/kg (+37%). This is in agreement with literature findings [18]. Thus, the energy content of the hydrochar (cat.1) on dry weight basis with respect to the feedstock is enhanced through HTC by a factor 1.29 (average of all experiments), which corresponds to the range of 1.01 to 1.41 found in literature [36]. An explanation for carbonization even below 180°C is that the feedstock contains hardly any lignin components, which generally requires higher carbonization temperatures than cellulose and hemicellulose [20]. The most influential operating parameter on HHV (R^2=0.79) is pressure, which is a function of the loading rate (TS), process temperature, and process duration. Fig. 9 shows the increase in energy density of char compared to foodwaste feedstock through HTC and the expected significant correlation between the carbon content and the higher heating value of the hydrochar.

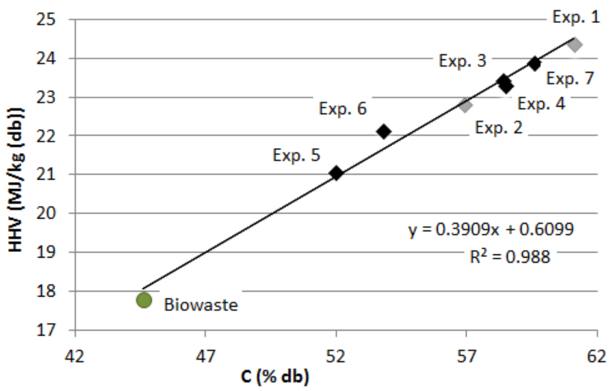

Fig. 9 Correlation carbon content vs. higher heating value of biowaste

As shown in Table 3 the ERs can be further improved by an average of 18% when considering the energy potential of the process water and by an additional 33% through minimizing heat losses of the reactor. Only considering the hydrochar as product under the stated process conditions, the average energy efficiency is 50%, which can be further improved to 58% and 63% when also including the theoretical energy content of the process water and minimizing electricity consumption through insulation.

It has to be noted that the calculated energy outputs, ratios and their improvements are of theoretical nature. In practice, these ratios would be lower due to the following reasons: i) only hydrochar of cat. 1 quality would most likely be used; ii) hydrochar would have reduced heating values as the dried char rapidly absorbs moisture from the humidity of the air, resulting in a moisture content of 5-10% [38]; iii) depending on the hydrochar combustion system, the lower heating value (LHV) rather than the higher heating value (HHV) would have to be applied. This is the case in systems where the heat of vaporization of the water content in the char is not being used; iv) a fraction of the total organic carbon in the process water would not be converted into biogas within an economically feasible retention time due to its recalcitrant nature and would remain in the liquid digestate. Additional enhancement of the energy efficiencies could be achieved by integrating internal heat recovery options (e.g., recirculation or heat-exchange of the hot process water and/or recovering steam from the process in a multiple batch system).
Figure 10 visualizes the coalification of the foodwaste in the Van-Krevelen diagram. Through dehydration and decarboxylation, the H/C and O/C ratio of the feedstock are de-

creased. The length of the vectors between input analysis (green point) and output analysis (grey points) reveal the moderate intensity of the process with the applied operational parameters. Furthermore, the locations of the hydrochars can be compared to typical values depicted as areas of the background coalification diagram [45-46]. The hydrochar composition and calorific quality are still in the range of (roasted) biomass and peat rather than in the expected hatched area marked for coal [25]. This indicates that the resulting hydrochars show similar characteristics to torrefied products (HHV: 20-24 MJ/kg, VM: 55-65%, FC: 28-35%, [47-48]. Overall, the HHVs of the hydrochars produced under diverse operational conditions still signify enhanced product characteristics for use as fuel (reduction of H and O, while increasing the energy density). Experiment 6, which applied short process duration (2h) at 180°C and medium pressure (18.6 bar), showed low electrical energy consumption, resulting in the best energy ratio of all performed experiments.

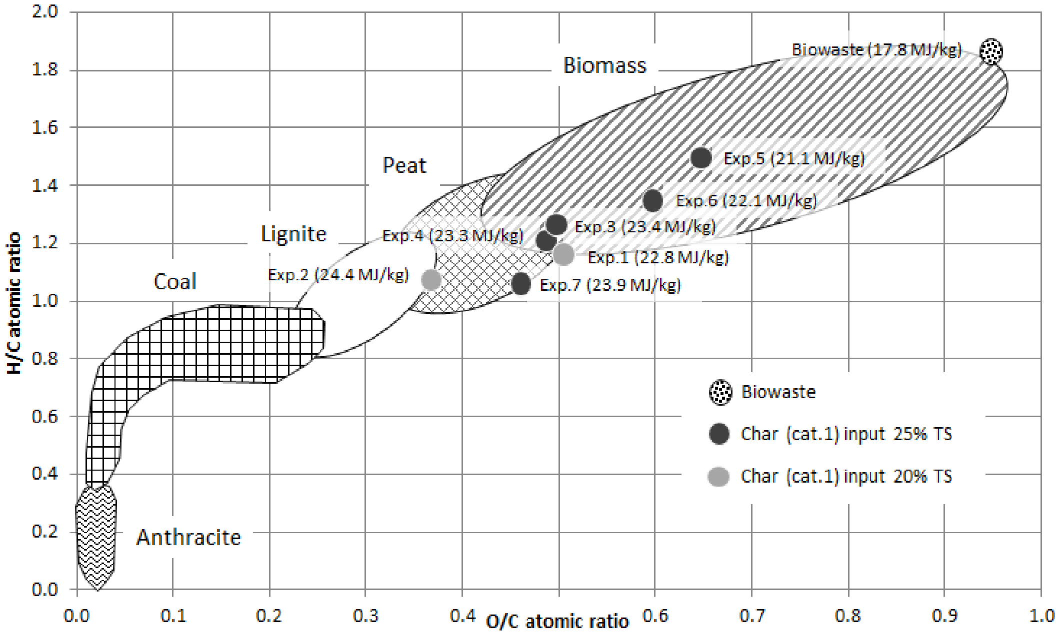

Fig. 10 Van Krevelen Diagram depicting the change in biowaste O/C and O/H ratio after HTC experiments, hatched areas by [44,45]

5. Conclusions

In this study, a model kitchen and market waste feedstock (17.8 MJ/kg_{db}) was carbonized in an experimental HTC reactor with varying loading rates (TS 20% and 25%) under mild operational conditions such as high heating temperature (160-190°C) and process time (2-10 h above 160°C). The aim was to optimize the energy-related aspects of the HTC process while examining the impact on the hydrochar quality. While the chemical properties were similar to torrefied products, the heating value of the produced hydrochar was of moderate quality (21.1-24.4 MJ/kg_{db}). The energy in the char was up to 2 times higher compared to the electrical energy consumed in the process. If the theoretical methane potential of the process water is additionally considered, the energy ratio increases to 2.6. Reactor insulation could enhance this ratio to 3. While high loading rates resulted in an enhanced energy balance of the process, it also led to uneven carbonization in this unstirred system and thus generated different qualities of hydrochar.

The best energy ratio (i.e. not taking into consideration the chemical energy stored in the biowaste) was observed when biowaste of TS 25% was exposed to a short period (2h above 160°C) and peak temperature of 180°C, resulting in rather low maximum pressure of 18.2 bar. This can be explained by the satisfactory energy density (HHV) of the produced char and the low electrical energy consumption resulting from the short heating time. The highest energy efficiency was observed for HTC of TS 20% at 190°C and 28.9 bar, with 8 h above 160°C. This high energy efficiency is due to the lower TS (i.e. lower chemical energy input of the biowaste), and the slightly higher energy output (HHV of the char) resulting from the more severe operating conditions. Summarizing the results of the experiments, favorable energy ratios can be obtained by a high loading rate, short process time and lower pressure. Best energy efficiencies can be obtained by operation at a lower loading rate, long process duration and high pressure. Overall, mild carbonization of canteen waste with acceptable hydrochar combustion properties was also observed at temperature below 180°C.

Although the hydrochar produced under mild process conditions cannot compete with the heating value of coal-like (hydro-)char produced at higher temperature and longer process period found in literature [18,19,23], the resulting char nevertheless shows advantageous properties compared to the raw biowaste: Homogenized, richer in carbon and energy density, with enhanced durability, hygroscopic and grindability characteristics. This char can be used as an alternative to fossil fuels in conventional industrial combustion processes, such as cement burning kilns. To determine the appropriateness of HTC as biowaste-to-energy technology for a specific context, it is essential to conduct an assessment of the complete waste conversion chain including a critical evaluation of the non-technical aspects.

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