Hydrothermal Carbonization: a Pilot-scale Reactor Design for Bio-Waste and Sludges Pre-treatment

L. Zaccariello[1], D. Battaglia[1], B. Morrone[2] and M.L. Mastellone[1,3,*]

[1]DiSTABiF, Università della Campania "Luigi Vanvitelli", Caserta, 81100, Italy
[2] Dept. Engineering DI, Università della Campania "Luigi Vanvitelli", Aversa (CE), 81031, Italy
[3]Athena srl, Via Vivaldi 43 – 81100 Caserta, Italy

Keywords: hydrothermal carbonization, hydrochar, digestate, biowaste

Abstract

The purpose of the paper is to illustrate the basis of design of a pilot-scale HTC reactor realized starting from an extensive series of process data obtained by an experimental research. The hydrothermal carbonization of industrial waste has been previously studied in the bench scale reactor with the aim to optimize the process parameters such as the temperature, reaction time, water-to-dry matter ratio and then scaled-up at a scale 30 times larger.
The new pilot-scale reactor has been designed by considering an operating temperature of 300°C, a volume of mixture of 0.1 m^3, an operating pressure of 86 bar. The design has been structured into two steps: a) process design; b) mechanical design. The main result from the process design step has been the evaluation of the amount of heat to be provided by an external source, the method to provide it as well as the related reaction time necessary to reach the given yield. By a mechanical point of view, the reactor geometry has been chosen to minimise the non-homogeneity of temperature along the section supplying the necessary heat from the external boundary to the reactor center, to allow an easy removal of both solids and gas/vapors, have a low footprint and be easily scaled-up.
The paper describes the pilot-scale prototype that has been realised starting from a well-designed experimental campaign at bench-scale level.

Introduction

Bio-based waste represents an important fraction of the waste produced daily by both domestic and industrial activities. The consistent fraction of water contained in such waste like those produced from household' kitchen and restaurants, food and beverage industries, sludge from wastewater plant treatment, etc. does not allow this kind of waste to be treated in conventional incinerators or used for energy recovery. The disposal costs and environmental challenges linked to these processes increasing constantly in the last decades due to the energy costs as well as the restrictions related the emission of bad odours, bioaerosols and leachate. It is a very severe environmental, social and economic problem in both developed and developing Countries, accounting today for a production of over 2 Gton/year and expected to be more than 3.4 Gton/year in 2050 (Kaza et al. 2018). The correct management of this peculiar waste is a critical issue in the waste system: its landfilling increases the global warming potential by producing methane and carbon dioxide and has been prohibited in many developed Countries; the energy recovery requires a preliminary drying step to remove the excess of moisture (usually larger than 70%), resulting in a negative energy balance; biochemical processes do not require any drying but yields of conversion are very limited, with a corresponding low mass reduction. Anaerobic digestion, a biotechnique that involves biowaste fermentation in controlled anaerobic bioreactors, for example, converts around 12-14% of substrates into biogas while the remaining part of the mass needs to be treated with high economic cost to be mineralised (Mastellone 2015).

Emerging trends in this field are gaining increasing researchers' attention. In particular, hydrothermal carbonization (HTC) offers several advantages, such as: minimization of nasty odour dispersion, destruction of pathogens that can cause crop, soil and water contamination, production of a carbonaceous material (hydrochar) that can be used to replace soil amendment and/or fertilizer (Ischia et al. 2021), (Lucian et al. 2020).

Scope of the research and applied methodology

The whole project started from the basic studies related the process of hydrothermal carbonization realized by considering its targeted application. The application was mainly focused on pretreatment prior the final disposal or the energy recovery of biowaste such as kitchen waste, food, sludges, digestate (intermediate of anaerobic digestion plant), etc. In this specific application, the quality of the biowaste (e.g. contaminants) and that of the resulting product (hydrochar) as well as the lack of regulations allowing its commercialization and use, have secondary importance. More specifically, the pre-treatment of biowaste by using HTC process can transform heterogeneous substrates having high moisture content, sometimes larger than 90%, into homogenous product with high carbon content, low nitrogen content, substantially hydrophobic and having a low moisture content (<30%). This transformation implies to apply a combination of thermal treatment in water bulk (HTC) followed by a mechanical treatment (centrifugation) that is more efficient due to the hydrophobic property of the resulting hydrochar.

The research plan has been then developed by considering each step with a precise target: building a prototype to demonstrate the feasibility of the application at industrial scale as a suitable pre-treatment of many types of biowaste. Virtually, all the biowaste with a high water fraction can be stabilized. This process would impede from releasing odors, a long storage with no reactions, high bulk density, easiness of water removal, good calorific value, etc.

The sequence of steps that characterized the research development is sketched in the Figure 1.

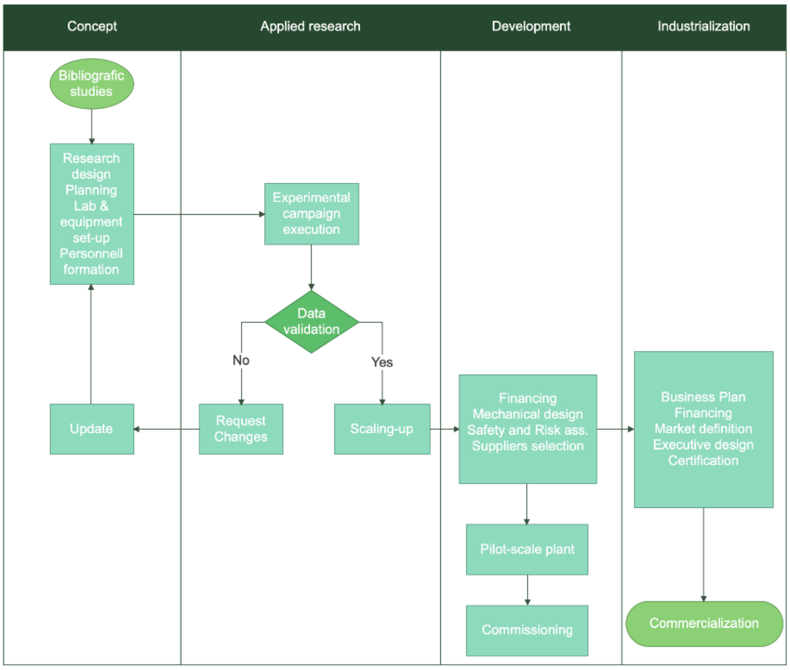

Figure 1 Planning of HTC research from the concept to the prototype installation

Process description

HTC is a thermo-chemical process that uses water under subcritical conditions to convert biomass into a C-rich product known as hydrochar. HTC process takes place at relatively low temperature (generally in the 180 – 250 °C range) under autogenous pressure. Whereas conversion to char via dry pyrolysis is restricted to biomass with dry water content, this process opens up the field of potential feedstock to substrates with a high moisture content such as food waste, biowaste in general, wet agricultural residues and fecal sludge. Hydrochar is an easy to handle product with good dewatering properties and when dried, has a high calorific value. It can be used as a soil conditioner and a carbon neutral combustible, but also for a wide range of other environmental, electrochemical and catalytic applications (Sevilla and Fuertes 2009; Sevilla, Fuertes, and Mokaya 2011; Jain, Balasubramanian, and Srinivasan 2016; Zhang et al. 2019).

The bench-scale apparatus and experimental results

The experimental campaign carried out on the bench scale aimed to investigate the effect of operating parameters on the yield and composition of hydrochar produced by using a biowaste produced by anaerobic digestion of food and kitchen waste. The experimental tests have been conducted utilizing an externally heated stirred batch reactor having a reaction volume of 3.6 litres. The simplified P&I diagram of the reactor and auxiliary equipment is shown in the Figure 2.

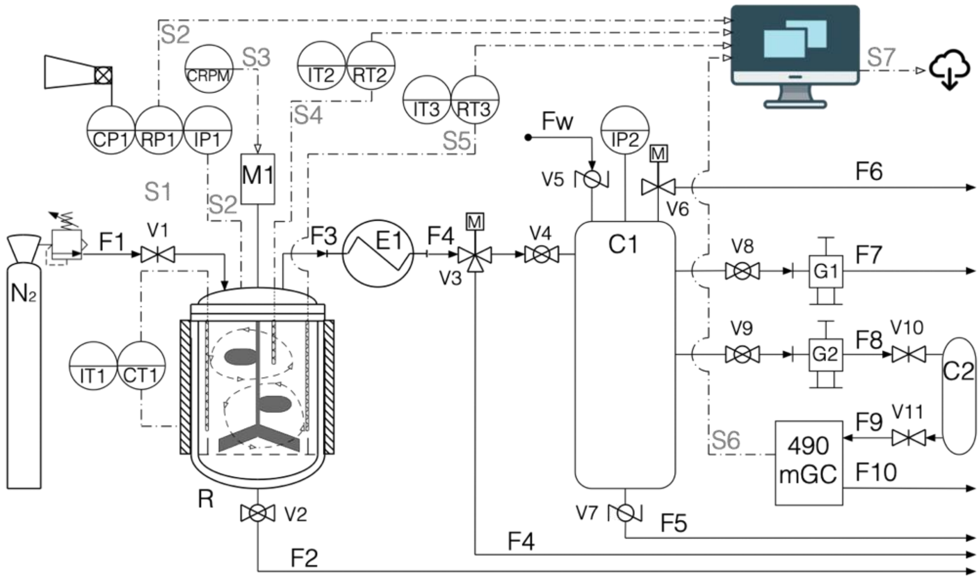

Figure 2: Schematic illustration of the hydrothermal carbonization apparatus. It is composed of the main process units: Reactor (R); Heat exchanger (E1) and a Condenser (C1).

Table 1 reports the main characteristics of the reaction mixture that was composed by biowaste and water at different ratios. The effect of this parameter together with the reaction time and the operating temperature has been studied and published elsewhere (Zaccariello et al. 2020).

C, $\%_{db}$	H, $\%_{db}$	N, $\%_{db}$	O, $\%_{db}$	Ash, $\%_{db}$	Moisture, $\%_{ar}$
35.9	4.5	2.4	20.0	37.2	67.8

*Table 1. Ultimate and proximate analysis of the biowaste utilized for the experimental runs. * db: dry basis; ar: as received*

Bench-scale experimental results

The experimental tests have been realised by modifying the water/(dry matter) ratio, the temperature of the reactor and the reaction time. After each test, samples of solids, liquid and gas have been analysed. Several indicators have been then calculated: the yield of remaining solids, mainly hydrochar, the yield of permanent gases, the carbon partition between the phases. The hydrochar yield decreases with the reaction time while the carbon content increases due to the progressive release of nitrogen, oxygen and hydrogen.

The preliminary experimental tests of hydrothermal carbonization of digestate indicated that for both investigated temperatures, i.e. 180 and 250°C, the hydrochar yield decreases from 0.89g/g to 0.81g/g and from 0.79g/g to 0.74g/g as the reaction time was increased from 2 to 6 h. A similar effect is produced by the increase of reaction temperature that affects the kinetic rate: the yield reduced from 0.89g/g to 0.78g/g, after 2 hours of reaction time, and from 0.81g/g to 0.74g/g, after 6 hours of reaction time.

The results indicated that both reaction temperature and reaction time affect the yield of the obtained hydrochar and their characteristics. This means that a high yield is not necessarily a positive result and that the yield itself cannot be used as a performance parameter. In particular, the yield decreases as temperature and reaction time increase.

Pilot-scale design basis

The pilot-scale reactor has been designed by considering the following input data and restrictions:

- Optimised set of operating conditions as obtained from the experimental campaign on the bench-scale
- No need of mechanical mixing to avoid gas leakage
- External heating with fast and easy control of the internal reactor temperature
- Modularity of the reactors instead of scaling-up the geometry
- Small footprint
- Easy maintenance such as internal cleaning

The reactor has been sized on the basis of the following set of parameters (Table 2) including the waste mixture (biowaste from food collection and wastewater sludge). The other parameters are related to the process management; the most important data are the number of cycles in a day (24h), the duration of the emptying of the reactor, its cleaning and filling as well as the real reaction time. The process conditions have been fixed on the basis of the experimental results in the bench scale: a process temperature of 200°C and a reaction time of 10 h guarantee a good conversion of dry matter into hydrochar and a limited energy cost.

Biowaste (food)	32%	
Waste content (d.b.)	15%	-
Moisture content	98%	-
Carbon fraction (d.b.)	47.0%	-
Sludge	68%	
Waste content (d.b.)	0%	-
Moisture content	97%	-
Carbon fraction (d.b.)	7%	-
Waste removal efficacy during pulping & filtering	90%	
Water/solids ratio (minimum requested)	10.00	-
Daily cycles for each reactor	2.00	1/d
Emptying, cleaning and feeding time	1.00	h
Reaction time	10.00	h
Heating time	1.00	h
Reaction temperature	200.00	°C
Gas yield (gas/mixture)	5%	t/t
Hydrochar yield	50%	$t_{IC\text{-}dry}/t_{SOLIDS\_in}$
Maximum allowed reactor pressure	70.00	bar

Table 2 Input data for the pilot-scale design

The reactor has been designed as a unit (single reacting volume) that will be part of a module composed by six units and a unique heating system. Each unit has the characteristics reported in the Table 3 and is sketched in the Figure 2.

Amount of gas produced	kg/cycle/reactor	5
Equilibrium Liq/Vap pressure (Antoine law)	bar	17.2
Gas product pressure	bar	20.6
Total gas pressure	bar	37.9
Reactor volume	m^3	0.1
Height	m	2.0
Diameter	m	0.3

Table 3 Pilot-scale geometry and design characteristics

The reactor unit is a batch filled by the top and externally heated. The temperature sensor controlling the heat power is located at bottom where a denser phase is present due to sedimentation of substrate and solid product. During the heating time the temperature continuously increases, and the power is set-up at its maximum level. When the operating temperature and pressure are reached the controlling of temperature is realised by means a PID control of heating power. During the process, exothermic reactions occur so that the power can be taken at such level to balance the losses. No cooling system is installed.
At reactor unit top, a head space is present where the gas produced from the reaction and the vapor in equilibrium with the liquid water are accumulated during the process. A safety valve (relief valve) is installed on the top so allowing the release of gaseous phase in case pressure exceeds the threshold limit. Once the process is completed the valve installed on the top opens by releasing gas and steam: the flow is addressed to the condenser where the pressure was 1 bar at beginning of the emptying process. The evaporation process continues until the equilibrium at the new process and temperature is reached. The pressure decreases from 38 bar to about 1 bar (the condenser promotes the fast passage from steam to liquid) so the change of enthalpy at saturation pressure for the water changes from 944 to 126 kJ/kg. The energy balance allows to estimate that almost 35% of liquid water passes in the vapor form to be recondensed. This leads to estimate the hydrochar-water ratio in the final sludge the 45% of the initial one. This sludge is removed by opening the valve located at bottom that is almost wide as much as the section of the reactor and addressing the sludge in the hopper and from this to the centrifuge.

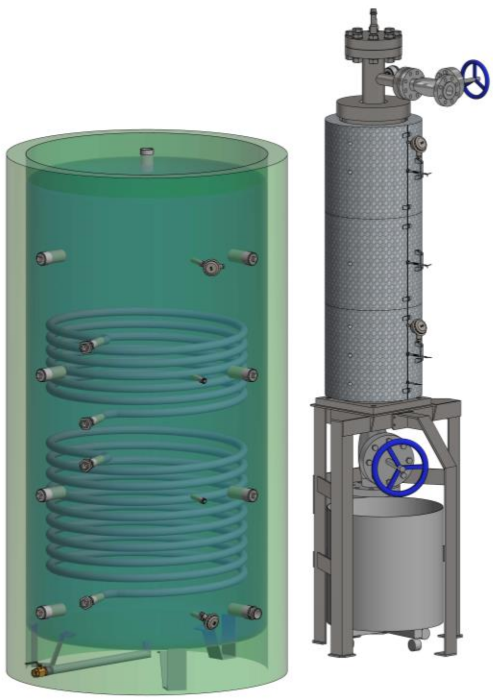

Figure 2 Pilot-scale HTC unit composed by reactor (on the right) and condenser (on the left). (Property of Athena srl)

Ambient temperature	20.0	°C
Reaction temperature	200	°C
Amount per module	90	kg/unit
Heating time	1.00	h
Thermal power (per unit)	19	kW

Table 4 Reactor (single unit) heating

Conclusion

The hydrothermal carbonization has been applied to several biowaste with a high water content to promote the inertisation of the organic carbon obtaining a stable (hydro)-char.
The experiments have been conducted in a bench scale reactor at different operating conditions in order to know the effect of reaction time, temperature and water-to-solid ratio on the yield and mineralization degree of hydrochar.
This work illustrates how the obtained data at lab-scale size have been scaled up at a pilot-scale so allowing the demonstration of the process reliability and scalability.
The pilot-scale unit is a batch-wise reactor having a volume enough small to avoid the mechanical mixing and a tubular geometry allowing an almost uniform temperature profile along the radius of the reactor.
The heating time, reaction time and cleaning/filling time require a cycle of 12hours so allowing two cycles/day for the treatment of 200liters of sludges/biowaste with 90% water content. The hydrochar produced will be about 10kg while the leachate to dispose will be 61liters instead of 200liters. The remaining water will be evaporated at very low rate in such a way to avoid stripping of organic compounds and condensed under form of distillate water.
The economic balance of this system can be then quite interesting depending on the cost for heating and the possible value for the hydrochar; this analysis is strongly depending by the Country's regulations.

Acknowledgments

V: ALERE 2019 grant support from Università degli studi della Campania "L. Vanvitelli" of CHIMERA project is gratefully acknowledged for financial support of the experimental activity.

References

Ischia, G., Fiori, L., Gao, L., and Goldfarb, J.L., 2021. Valorizing Municipal Solid Waste via Integrating Hydrothermal Carbonization and Downstream Extraction for Biofuel Production. Journal of Cleaner Production 289: 125781.
Jain, A., Balasubramanian,R., and Srinivasan, M. P., 2016. Hydrothermal Conversion of Biomass Waste to Activated Carbon with High Porosity: A Review. Chemical Engineering Journal.
Kaza, S., Yao, L C.; Bhada-Tata, P. and Van Woerden, F., 2018. What a Waste 2.0. A Global Snapshot of Solid Waste Management to 2050. Washington: World Bank Publications.
Lucian, M., Volpe M., Merzari F., Wüst D., Kruse, A., Andreottola, G., and Fiori, L., 2020. Hydrothermal Carbonization Coupled with Anaerobic Digestion for the Valorization of the Organic Fraction of Municipal Solid Waste. Bioresource Technology 314.
Mastellone, M L., 2015. Waste Management and Clean Energy Production from Municipal Solid Waste. Waste and Waste Management. New York: Nova Science Publ.
Sevilla, M., Fuertes, A. B., and Mokaya, R., 2011. High Density Hydrogen Storage in Superactivated Carbons from Hydrothermally Carbonized Renewable Organic Materials. Energy and Environmental Science.
Sevilla, M, and Fuertes, A. B., 2009. The Production of Carbon Materials by Hydrothermal Carbonization of Cellulose. Carbon 47 (9): 2281–89.
Zaccariello, L., Mastellone M.L., D'Amelia, L.I., Catauro, M., and Morrone, B., 2020. Assessment of Integration between Lactic Acid, Biogas and Hydrochar Production in OFMSW Plants. Energies - MDPI 13 (24): 1–19.
Zhang, S., Zhu, X., Zhou, S., Shang, H., Luo, J., and Tsang, D.C.W., 2019. Chapter 15 - Hydrothermal Carbonization for Hydrochar Production and Its Application. In Biochar from Biomass and Waste edited by Ok,Y.S., Tsang, D.C.W., Bolan, N. and Novak, J.M., 275–94. Elsevier.