

# Waste to Biofuels through Zero-Energy Hydrothermal Solar Plants: Process Design

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Hydrothermal processes enable an effective conversion of waste biomasses into fuels and carbonaceous materials. Covering the heat requirements with concentrated solar energy is a clever strategy to increase the plant efficiency and pursue the principles of circular economy. With the purpose of producing liquid and solid biofuels through zero-energy routes, this work presents two conceptual designs for integrating a concentration solar system (CSS) with a hydrothermal liquefaction (HTL) and a hydrothermal carbonization (HTC) plant. The solar configuration used to cover hydrothermal heat requirements consists of a set of parabolic trough collectors operating with molten salts, which are used as both thermal carrier fluid and thermal energy storage medium. Two different scenarios were modelled to continuously process wood and organic wastes. In the first scenario, the CSS is coupled with a continuous HTL reactor (operating at 400 °C and 300 bar) followed by thermal cracking and hydroprocessing for upgrading bio-crude to a saleable liquid biofuel. The second scenario considers the CSS operating with a continuous HTC reactor (working at 220 °C and 24 bar) for the conversion of organic wastes into a solid fuel (hydrochar). The CSS and both the hydrothermal plants were modelled based on experimental data. Energy consumptions and techno-economic aspects were investigated.

## 1. Introduction

Climate change and resource depletion require the development of new strategies to concretize positive impacting actions. In this context, the valorisation of waste biomass has a key role in a circular economy vision. In particular, hydrothermal processes enable the upgrade of biomass into liquid, solid and gaseous biofuels. Occurring in water, these processes can treat a multitude of non-conventional biomass, as sewage sludge, agricultural residues, and organic wastes. Operating conditions vary according to the desired product. Hydrothermal carbonization (HTC) occurs at mild operating conditions (180-250 °C and 10-40 bar) and lead to a solid phase, known as hydrochar, which can be used as solid fuel (Volpe et al., 2016). Under harsher conditions (300-400°C and 100-300 bar), hydrothermal liquefaction (HTL) occurs, leading to the production of a crude oil. This finds applications in the production of liquid fuels (Castello et al., 2018).

One of the main drawbacks of hydrothermal processes (especially HTL due to the harsher operating conditions) is the large amount of heat required to reach operating conditions (Giaconia et al., 2017). In this regard, the usage of renewable energy to cover energy needs is a possible strategy to accomplish the principles of circular economy and develop a zero-energy plant (Ischia et al., 2020).

This work investigates two possible scenarios in which two hydrothermal plants (a HTC and a HTL) are coupled with a solar concentrator system (CSS). The CSS, equipped with solar collectors and thermal energy storage systems, is designed to cover thermal energy requirements of the hydrothermal plants. An economic evaluation was performed to show the feasibility of these sustainable options.

## 2. Methodology

### 2.1 General layout

Figure 1 shows the general layout of the investigated conceptual plants. The hydrothermal plant schemes are based on previous research articles (Hansen et al., 2019; Lucian and Fiori, 2017), while the configuration of the CSS is based on commercial installations (ESTELA et al., 2016; Kurup and Turchi, 2015). The HTL plant aims to produce a liquid drop-in fuel starting from biomass (500 tons of dry wood per day). The HTC plant treats 200 tons of organic wastes (dry basis) per day, corresponding to about 400 ton of as-received matter per day, (about 3 millions of equivalent inhabitants). The purpose and contextualization of the two systems is different: HTL focuses on the production of a high-quality product, while HTC addresses waste management issues.

Inside the hydrothermal plants, the raw biomass is converted into the desired product. The feedstock of the HTL system is a slurry of wood and water, processed at 400 °C and 300 bar. Regarding HTC, organic wastes are continuously treated at 220 °C and 24 bar (residence time of 3 hours). In both cases the model inputs (e.g. product composition and yields) are based on previous works (Pedersen et al., 2017; Lucian et al., 2018).

The CSS is sized to cover the thermal energy required to perform a continuous thermochemical conversion. To this purpose, a heat fluid carrier (consisting of a mixture of molten salts) is heated up inside the parabolic trough collectors and releases heat to the hydrothermal plant. The solar field is composed of modular solar collectors connected in parallel rows, known as Solar Collector Assemblies (SCA). Each row consists of a 2x100 m collectors characterized by an area of 1113.6 m<sup>2</sup> (Giaconia et al., 2017). To ensure a continuous operation of the plant, thermal energy storage (TES) tanks are used. These have the advantage of mitigating weather fluctuations and extending the heat supply during overnight and cloudy periods. The molten salt mixture adopted is a commercial one (HITEC) and consists of a mixture of NaNO<sub>2</sub>, NaNO<sub>3</sub>, and KNO<sub>3</sub> (Turrini et al., 2018). In particular, under the chosen operating conditions, corrosion effects with stainless steel are almost negligible (Coastal, 2009).

As shown in Figure 1, biomass is initially pre-treated with water to obtain a slurry. Then, the slurry is compressed and heated up to the design conditions by a pump and a counter-current heat exchanger. Here, the mixture of molten salts, previously heated inside the solar collectors, provides the required thermal energy. The outlet temperature of the molten salts stream is maintained constant by varying the flow rate according to the direct solar radiation. A hot TES was positioned downstream the solar field. When the thermal energy from the sun is higher than the process demand, a fraction of heating fluid is stored inside the tank for being used in absence of solar radiation. Then, hot molten salts are conveyed to the heat exchanger, where they heat up the slurry. Afterwards, they are stored in a cold TES and pumped back to the solar field. When the direct radiation is not sufficient to guarantee a proper molten salt exit temperature, the stream is pumped back to the cold tank. TES has a storage capacity of 24 hours. Inside the HTL reactor, wood is converted into bio-crude with a yield of 40 %. This value has been chosen on a conservative basis, since recent studies have demonstrated that higher yields can be reached (Hansen et al., 2019). The gas phase (mainly consisting of CO<sub>2</sub>) represents the 13 %, while solids and the aqueous phase with dissolved organics the remaining 47 %. The solid phase is removed inside a cyclone positioned downstream the HTL reactor. The pressurized hot stream is then cooled down, depressurized and sent to a vessel to separate the three phases (biocrude, water and gas phases), where 90 % of the water phase is recirculated back. The bio-crude is upgraded through thermal cracking (600°C and 4 bar) and hydrotreating (at 360 °C and 77.5 bar). Regarding the HTC reactor, organic waste is converted into the desired solid phase (hydrochar) with a yield of 77.2 %. After the reaction, the slurry is depressurized, cooled down and the liquid phase separated in a decanter. Part of the water is recirculated back.

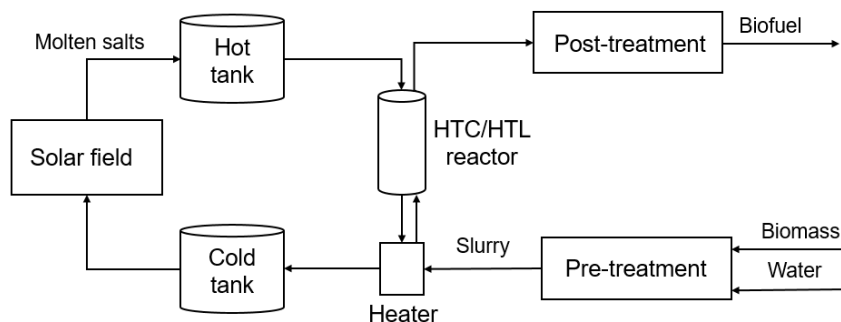


Figure 1: General layout of the CSS-hydrothermal plants.

## 2.2 Modeling of the process

Energy and mass balance computations were performed using Matlab. Thermodynamic properties of water were obtained by using the software Cantera. Energy balances were performed to size the different components. The same procedure was adopted for the HTL and HTC design. The reactor is tubular and in stainless steel AISI 316L. A free volume of 30 % was considered. The reactor volume was chosen to guarantee the nominal volumetric flow rate of the slurry and a residence time of 15 min for HTL and 3 hours for HTC.

The specific heat demand ( $q_{heat}$ ) to rise up the slurry stream from the initial temperature ( $T_0$ , equal to 25 °C) to the set point one ( $T_{set}$ ) was computed according to Eq(1).  $c_{p,bio}$  is the specific heat capacity of dry biomass, assumed constant and equal to 1.2 kJ/(kgK).  $c_{p,w}$  is the specific heat capacity of water, function of temperature and pressure according to available thermodynamic data. B/W is the ratio between dry biomass and water, equal to 0.30 for HTL and 0.25 for HTC.

$$q_{heat} = c_{p,bio} \cdot (T_{set} - T_0) + \frac{W}{B} \cdot \int_{T_0}^{T_{set}} c_{p,w}(T, P) \cdot (T_{set} - T_0) \quad (1)$$

The heat required by the reactor was computed considering both the heat of reaction ( $\Delta H_r$ ) and heat losses ( $q_{loss}$ ). For HTC, the gas was assumed composed entirely of CO<sub>2</sub>, while for HTL the composition proposed by Hansen et al. (2019) was considered. For both HTC and HTL, the liquid phase was assumed composed by water and acetic acid.  $\Delta H_r$  was computed by applying the Hess's law, as shown in Eq(2), where the subscript  $p$  indicates the products. The molar enthalpy of formation ( $\Delta h$ ) of biomass, bio-crude and hydrochar was computed starting from their experimental higher heating values (on a dry ash free basis), on the base of the combustion reactions. This approach has been widely adopted for modeling biomass processes (like torrefaction, gasification and pyrolysis) (Lozano et al., 2019). For conventional components, thermodynamic data were used. Water was assumed non participating to the reaction.  $MY$  and  $MW$  are the mass yield and the mean molecular weight of the i-compound, respectively.

$$\Delta H_r = \sum MY_p \frac{\Delta h_p}{MW_p} - \frac{\Delta h_{bio}}{MW_{bio}} \quad (2)$$

$q_{loss}$  were computed as the sum between conduction, convection and radiation contributions. Reactors are in stainless steel AISI 316L and are thermally isolated by 25 cm of rock wool. Heat transfer relations of a cylinder were applied by using thermal properties of the used materials (Incropera, 2017). The power required by the pump ( $P_{pump}$ ) to increase the pressure of the slurry from  $P_0$  (set equal to 1 bar) to  $P_{set}$  was computed by applying Eq(3), where  $\dot{V}$  is the slurry volumetric flow rate and 0.8 the efficiency of the pump.

$$P_{pump} = \frac{\dot{V}(P_{set} - P_0)}{0.8} \quad (3)$$

The overall thermal requirements of hydrothermal plants ( $P_{tot}$ ) were used to compute the respective molten salt flow rate ( $\dot{m}_{HTF}$ ), assuming a heat exchanger efficiency ( $\eta_{hx}$ ) equal to 90 %. The temperature of the HTL tanks was fixed at 420 and 380 °C, while for HTC at 230 and 210 °C. Then, the CSS was sized. The efficiency of the solar collectors ( $\eta_{solar}$ ) is equal to 72 % and comprises the optical and receiver efficiencies (Giaconia et al., 2017). The projected area of the solar collectors ( $A_{solar}$ ) was computed from Eq(4), where  $I_d$  is the direct solar radiation. This last was assumed equal to 700 W/m<sup>2</sup>, which corresponds to the peak average hourly irradiance in the center of Italy, in summer (ESMAP, 2019).

$$A_{solar} = \frac{P_{tot}}{\eta_{hx} \eta_{solar} I_d} \quad (4)$$

Once the plant component were sized, an economic evaluation was carried out. A plant lifetime of 10 years and a continuous operation (24/24) were assumed. Regarding the HTL plant, its capital and operating costs (CAPEX and OPEX) were estimated on the base of results obtained by Pedersen et al. (2017). For the HTC plant, reference is done to the work of Lucian and Fiori (2017), from which the costs of methane burner were removed. After the actualization, CAPEX and OPEX were scaled by applying Eq(5), where a scaling exponent equal to 0.65 was used (Smith, 2005). The same procedure was applied for the operating costs, making reference to those reported by Lucian and Fiori (2017), reduced by methane costs.

$$Cost_{plant} = Cost_{plant,2019} \left( \frac{Plant\ capacity}{Plant\ capacity,ref} \right)^{0.65} \quad (5)$$

The installation and operating costs of the hydrothermal plants were computed independently of the solar field and TES. In particular, CAPEX of the HTC plant includes the costs for the on-site equipment (like the reactor, pumps, agitator, crusher, filter, dryer, and the pelletizer) as well as depreciation costs (computed using a linear model), land use, utilities, start-up of the plant, and working capital. Meanwhile, OPEX comprises annual costs for electricity, labour, maintenance, and waste-water treatment. For HTL, costs related to the post-treatment phase were also considered. Costs for the thermal cracker and the hydrotreater were included in the total, while the hydrogen purchase was included in the operating costs. CAPEX of the solar plant (solar collectors, molten salts and TES) was computed by averaging and actualizing data reported in previous studies (ESTELA, 2016; Klasing et al., 2018; Giaconia et al., 2017; Kurup and Turchi, 2015). CAPEX of the solar field includes the costs for the solar collectors, molten salts, back up heaters, instrumentations, civil works, installation, balance of plant, and contingency. In particular, CAPEX of 205 €/m<sup>2</sup> and of 28.0 €/kW<sub>th</sub> were assumed for the solar field (comprised of solar collectors and molten salts) and TES, respectively. Operating costs of solar field and TES were assumed equal to 2.2 % of the CAPEX (ESTELA, 2016). These include also the costs for the replacement of molten salts.

Then, the final production cost was computed as a minimum fuel selling price (MFSP). This represents the fuel selling price at which the net present value of the process is equal to zero (Pearce et al., 2016). A cash flow analysis was carried out considering the overall plant life of 10 years and it was assumed an operative plant time of 328 days per year, 5 % of annual interest loan with a payback time of 10 years, and a taxation rate of 37 %. The cost to buy the biomass for the HTL plant was assumed equal to 41.5 USD/ton (wood, comprised of the transportation). Conversely, two scenarios were considered for the MFSP of pelletized hydrochar. The first scenario does not consider any income from the taking in charge of organic wastes (to be conservative), while the second scenario assumes an income of 60 €/ton (common value in the North of Italy).

### 3. Results and discussion

#### 3.1 Thermal energy requirements

Table 1 reports a summary of specific thermal energy requirements. As expected from the harsher working conditions of HTL, the specific heat required to rise the slurry temperature is much higher than that required by HTC. Regarding the heat of reactions, with a positive value of  $\Delta H_r$ , the HTL of wood has an endothermic behaviour. Unlike HTL,  $\Delta H_r$  of HTC is negative, indicating that the reaction is exothermic. This behaviour is well-known in the literature and was already quantified through analytical methods (like differential scanning calorimetry, DSC (Pecchi et al., 2020)) and lab-scale reactors (Merzari et al., 2018).

For the sizing of the solar plant, the heat released during HTC was not considered, in order to adopt a conservative approach. Heat losses are much more important for HTC, representing about 1.5 % of the total thermal energy demand. For HTL, they account for about 0.5 % of the total, meaning the size of the reactors has a higher impact than the operating temperature.

*Table 1: Specific thermal energy requirements (MJ/kg<sub>bio</sub>) of hydrothermal reactors (B/W of 0.30 for HTL and 0.25 for HTC).*

	Heating ( $q_{heat}$ )	Heat of reaction ( $\Delta H_r$ )	Heat loss ( $q_{loss}$ )
HTL	7.09	3.00	0.05
HTC	3.60	-0.32	0.05

#### 3.2 Design parameters and economic analysis

Table 2 summarizes design parameters of the hydrothermal plants and respective solar fields. The difference between the two reactors is evident. Due to the longer residence time required for the reactions, the HTC reactor exhibits a much bigger volume than the HTL one. Regarding the required power, the process operating conditions are decisive, leading to 58.7 and 8.4 MW for the HTL and HTC reactors, respectively. Similarly, with a value of 852.1 kW, the nominal power of the HTL pump is much higher than the HTC one.

Overall, the solar collectors have a huge impact on the overall plant surface. Indeed, to guarantee a continuous operation for the HTL plant, 114 parabolic trough collectors (each of 1136 m<sup>2</sup>) are required. 17 solar collectors are enough for the HTC plant, which requires a lower amount of thermal power. This reflects on the TES volumes, equal to 15.6 and 4.5 m<sup>3</sup>.

Table 2: Design parameters of the hydrothermal plants and respective solar plants.

	HTL	HTC
<b>Hydrothermal plant</b>		
Biomass flow rate (ton/day)	500	200
Total input flow rate (ton/day)	2167	1000
Power (MW)	58.7	8.4
Reactor volume (m <sup>3</sup> )	29	162
Reactor surface area (m <sup>2</sup> )	78	166
Pump power (kW)	852.1	28.9
<b>Solar plant</b>		
Power (MW)	58.7	8.4
Collector area (m <sup>2</sup> )	129456	18620
Number of collectors	114	17
TES volume (m <sup>3</sup> )	15.6	4.5

Table 3 shows the results of the economic analysis. The model predicts a CAPEX of 157.1 and 5.6 M€ for the HTL and HTC apparatus, respectively. The investment cost of the hydrothermal plant is predominant for HTL (69.1 % of the total), while it represents a much lower fraction in the case of HTC (35.7 %). This difference is due to the much higher complexity of the HTL plant, which comprises also the post-treatment stage. TES costs dominate the solar plant investment. In both cases, operating costs involved in the hydrothermal plant represent up to 90 % of the total. The minimum selling prices of bio-oil and hydrochar were computed starting from total costs, considering a life-time period of 10 years (which is thus equal to the payback period). The production accounts for 200 tons of bio-oil and of 120 tons of hydrochar per day. Bio-oil exhibits a MSFP of 1.33 €/LGE (litre of finished fuel, in gasoline equivalent), which is slightly higher than the range 0.56-1.18 €/LGE typical of traditional HTL plants (Castello et al. 2018). The MFSP of pelletized hydrochar equals 163 €/ton, which is a value competitive with wood pellets, whose prices usually range between 150 and 200 €/ton (US EIA, 2019). Interestingly, considering an income of 60 €/ton due for the taking in charge of the as-received organic waste, the hydrochar MFSP reduces to 37.4 €/ton, highlighting the convenience of using the CSS-HTC plant for waste-management purposes. This value is similar to coal prices, currently equal to 20.2 €/ton (lignite) and 59.0 €/ton (bituminous coal) (US EIA, 2019).

Therefore, according to the obtained results, coupling HTL or HTC with a solar concentration apparatus appears as a reasonable strategy towards the development of a zero-energy plant. Solar energy covers the thermal energy process demand, leading to an improved sustainability of traditional systems. Certainly, for a more accurate evaluation, taxation, subsidies and policies should be considered. Finally, it should be noted that all computations were performed in a conservative fashion and with the aim of optimizing the process, certain measures could be adopted. For example, the hot stream exiting from the hydrothermal reactors could be used to pre-heat the slurry in a heat exchange and a part of the off-gases exiting from the HTL reactor could be burnt to cover part of the heat requirement.

Table 3: Economic parameters of the integrated CSS-hydrothermal plants.

	CSS-HTL plant		CSS-HTC plant	
	CAPEX	OPEX	CAPEX	OPEX
MFSP	1.33 €/LGE <sup>c</sup>		163 €/ton <sup>a</sup> 37.4 €/ton <sup>b</sup>	
Hydrothermal plant (M€)	157.1	43.7	5.6	3.0
Solar field (M€)	26.5	0.6	3.8	0.1
TES (M€)	43.8	1.0	6.3	0.1
Total (M€)	227.4	45.3	15.8	3.2

<sup>a</sup> and <sup>b</sup> without and with considering the income derived from the taking in charge of organic wastes, respectively. <sup>c</sup> litre of gasoline equivalent.

#### 4. Conclusions

This work investigates the coupling of a solar concentration plant with a HTL and a HTC plant. Energy requirements necessary to perform liquefaction and carbonization reactions were provided through a stream of molten salts heated inside solar collector. A TES apparatus enables the continuous operation of the plants. Reasonable values of MFSP (bio-oil: 1.33 €/LGE; hydrochar: 37.4 €/ton) show that this solution could constitute an effective strategy to improve the sustainability of the hydrothermal processes. Indeed, thermal

energy requirements are fully covered by solar energy and sub-products derived from the process, like the gas phase rich in H<sub>2</sub> produced during HTL (Hansen et al., 2019), could be used for other purposes (e.g. in the chemical industry).

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