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Process evaluation of mild hydrothermal carbonization to convert wet biomass residue streams into intermediate bioenergy carriers

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ABSTRACT

Hydrothermal carbonization (HTC) is a promising process for the upgrading of wet biomass residues. Models of HTC processes, in particular at continuous pilot-scale, are needed to move HTC from lab-scale to industrial scale. This study presents a process model for mild HTC, dewatering and conversion to intermediate energy carriers (bio-pellets and biogas for power and/or heat production) of three wet biomass residue streams: paper sludge, olive pomace and orange peels, based on lab- and pilot-scale experiments. In addition, the process energy efficiency and feedstock utilization of the HTC process is calculated and compared with conventional treatment options for the chosen residues, i.e., direct anaerobic digestion (olive pomace, orange peels) or combustion after conventional dewatering (paper sludge). The process model indicates that the HTC pilot-scale process is much more efficient in terms of feedstock utilization to produce heat and/or power than the reference scenarios. The process energy efficiency of the HTC process (pilot-scale) was calculated to be 26 %, 63 % and 40 % for paper sludge, olive pomace and orange peel feedstocks, respectively. For all feedstocks, both the solid and liquid-generated products are equally important for improving the overall process energy efficiency. This study demonstrates the potential benefits of HTC processes for upgrading wet biomass waste streams based on continuous pilot-scale data.

1. Introduction

The transition from fossil fuels to renewable energy sources is a requirement in the effort to mitigate climate change. Biomass energy will play a role in the energy transition, in particular to balance the energy grid against fluctuations from wind and solar [1]. Among sources of biomass feedstocks, biomass waste and residue streams represent a significant energy potential and are desirable for use because they typically do not compete with food production or land use. In Europe, the availability of biomass waste streams is expected to reach 1.7–5.0 EJ/yr by 2050, including biogas production potential [2]. Wet biomass residues, such as food processing wastes, and biological sludges (including municipal and industrial wastewater sludges), are typically unsuitable and unreliable for direct energy applications because they have relatively low energy density, high water content, high salt content, uneven distribution or seasonality and are susceptible to rapid degradation. As a result, these types of wet residues are usually dewatered, incinerated, landfilled, digested or composted [3], often at high environmental and economic cost and with limited energy recovery where applicable. Thus, upgrading wet biological sludges and food processing sludges would be beneficial to improve their energy density and overall quality for potential use as a fuel source.

One method for upgrading wet biomass wastes to produce energy is via hydrothermal carbonization (HTC). HTC is a thermochemical conversion process that operates at temperatures from 180 °C to 350 °C and is suitable for wet streams without prior dewatering or drying [4]. HTC results in a slurry that can be dewatered to produce a solid hydrochar and a liquid effluent stream. HTC has been applied to treat a wide variety of residue streams, both lignocellulosic (e.g., agricultural residues) and non-lignocellulosic (e.g., wastewater sludge) in nature [5].

Torwash® is a patented process that operates at the "milder" range of HTC, i.e., 150 °C–250 °C. Torwash has been used to treat municipal wastewater sludge and biological sludge from a paper mill [6] at continuous pilot scale, as well as various other wet residue streams. The main purpose of the treatment is improving dewaterability of the residue stream (without chemicals or other dewatering aids) and removing ash and salts from the hydrochar, thus improving its fuel quality. The solids can be further dried and/or densified (pelletized) for use in heat

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generation or combined heat and power (CHP). Whereas some previous studies have not factored in valorisation of the liquid stream from HTC and focus on the solid hydrochar product [7–9], in the mild Torwash process the liquid effluent stream is anaerobically digested to produce biogas. This produced biogas can, in turn, be utilized to alleviate the energy demand of the process. This makes Torwash mild HTC an attractive alternative to traditional hydrothermal conversion processes, especially for wet residue streams.

Development of models to evaluate, optimize and predict performance of HTC processes are needed to scale the technologies from lab-to pilot-to commercial-scale. Challenges in developing models of HTC include the complexity of the reactions and a diverse array of feedstocks and process conditions, resulting in different hydrochar properties [10]. Thus, most research on HTC has been comprised of experimental studies investigating the process conditions, almost exclusively at lab-scale in batch-mode [11] with limited focus on modelling [12]. Modelling research to-date is therefore also predominately based on lab-scale batch process data, for a variety of feedstocks including sludges and food residues such as grape pomace [13], olive pomace [14], avocado stones [15] and sewage sludge [16]. A model of a continuous hydrothermal liquefaction (HTL) process has been published [17], mainly focussing on heat transfer aspects. Extensive modelling work on olive pomace [13] has been published, with focus on the technical design and economics, and in which the authors simplify the biomass to a model component and produce only bio-pellets as product. A recent paper adds the importance of coupling the solids production with anaerobic digestion of the liquid fraction [18]. A techno-economic evaluation of HTC for paunch waste is presented by Ref. [19] focussing on the solid fraction only, and using models designated for coal in the process modelling. The present study specifically aims at presenting a modelling approach aimed at the energy content of both the solid and liquid streams through modelling changes in the composition of the biomass (C,O,H content) and relate this to the heating value and chemical oxygen demand, which are essential elements of the energy efficiency. While all studies make use of experimental data, the present study is novel in that both lab- and pilot-scale data was available and utilized and addresses using the solids product as well as producing biogas from the liquid product. This provides a better understanding of the process as well as of the characteristics of different feedstocks therein.

Models of HTC processes at continuous pilot scale are particularly valuable towards the better understanding, scale-up and commercialization of HTC technology for wet organic wastes. Therefore, the objectives of the current study were to:

i) Develop a process model for the Torwash mild HTC process (including upstream and downstream processes of feed conditioning, dewatering, drying and pelletizing) based on data from lab- and (continuous) pilot scale operation, for various wet biogenic residues: paper sludge, olive pomace and orange peels; and
ii) Assess the benefits and feasibility of the Torwash mild HTC process from a technical perspective (efficiency), compared to reference cases (standard practices including anaerobic digestion and conventional chemical-aided dewatering) for the chosen residue streams.

2. Materials and methods

2.1. Process descriptions and schematics

This study developed process models for the mild HTC of three wet residue feedstocks: paper sludge (PS), olive pomace (OLP) and orange peels (ORP). These feedstocks were chosen for study as they represent wet residue streams in the categories of sludges and agro-food residues. These types of residues are produced in vast quantities and represent a high potential for upgrading to produce energy. Paper sludge represents sludge produced from both the papermaking process (fibre sludge) and from wastewater treatment (biological paper sludge) at a paper mill. Olive pomace represents the residue stream that remains after initial cold pressing of olives to extract olive oil. Orange peels represent the residue stream following orange pressing to remove juice and pulp. Fig. 1 depicts the process schematics for mild HTC of these three residue streams.

For the paper sludge case, as depicted in Fig. 1(a), Torwash mild HTC is used to treat only the biological paper sludge since there is no advantage, with respect to dewatering, of hydrothermally treating the fibre sludge [6]. The HTC process for biological paper sludge involves first using a decanter centrifuge to increase the dry matter (DM) content of the biological sludge from 1% to 4% to approximately 10 %. The pre-conditioned feed then undergoes Torwash mild HTC, producing a wet slurry (mix of solids and liquids) and a gas stream. The gas produced is sent to a hydrogen sulphide (H_2S) adsorber unit, namely an iron sponge, after which the gas is vented. The slurry effluent from the HTC reactor is mechanically dewatered via a filter press. The filtrate, containing some bio-organics, is anaerobically digested to produce biogas. The biogas is subjected to H_2S removal with an iron sponge and subsequently the low H_2S biogas is then used for heat generation in a gas boiler.

The pressed wet solid cakes from the filter press are sent to a drier. Using hot air the moisture content is reduced to 10 wt%. The dried cakes are then sent to a crusher to reduce the particle size. The crushed cakes are sent to a stabilization tank to cool down. The granular feed is then fed to a hammer mill after which the material is pelletized (with conventionally-dewatered fibre sludge) to produce the main bio-pellet product that is exported for dispatchable energy. Included in the analysis is also combustion of the pellets in an industrial boiler for producing low temperature steam. A custom case (CSTM) is possible for the biological paper sludge feedstock which entails excluding the pelletization step and opting for direct combustion of the dried solid cakes using a pre-existing boiler, as indicated in Fig. 1(a).

The process scheme configurations for treatment of olive pomace and orange peels (Fig. 1(b)) are identical to each other and largely similar to that of biological paper sludge treatment. The differences with biological paper sludge treatment are that dilution of the olive and orange feedstocks with water is required and, since there is no clear heat demand on-site, the biogas from anaerobic digestion is utilized for power generation rather than for heat generation.

For comparison with alternative applications of the wet residue streams for energy purposes, a reference scenario (REF) was developed for each of the feedstocks, schematics of which are provided in Fig. 2.

The reference scheme for paper sludge (Fig. 2(a)) is derived from conventional industrial paper sludge treatment. In this scenario, fibre sludge (FS) residue from the paper making process is mixed with biological paper sludge (BS) from the wastewater treatment plant at the mill to form a mixed paper sludge. This is done because the FS acts as a dewatering aid for the BS. This mixed stream, i.e. paper sludge, has a dry matter content of approximately 4 wt% and is dewatered via a gravity table, increasing the dry matter content to 8 wt%. The energy required for this step is 8 kW-hr/tonne$_{ar}$ feed [20]. The dewatering is further aided by the addition of polyelectrolyte (PE) and ferrous sulphate salt. The concentrated sludge is sent to a screw press for further dewatering, which increases the DM to 30 wt%. This step requires an electrical energy input of 10 kW-hr/tonne$_{ar}$ feed [20]. This dewatered stream is sent to the onsite biomass boiler where steam is generated. The remaining effluent from the dewatering is recycled back to the wastewater treatment plant (WWTP).

The reference scenarios for olive pomace and orange peels (Fig. 2(b)) are based on anaerobic digestion of the residue streams. Given the high water content of these streams (>80 % moisture), direct combustion is not an attractive option [21]. Anaerobic digestion of these streams is therefore a feasible option for energy generation. However, it should be noted that other end uses are possible and often practiced. For example, olive pomace still contains residual oil, which is often recovered through

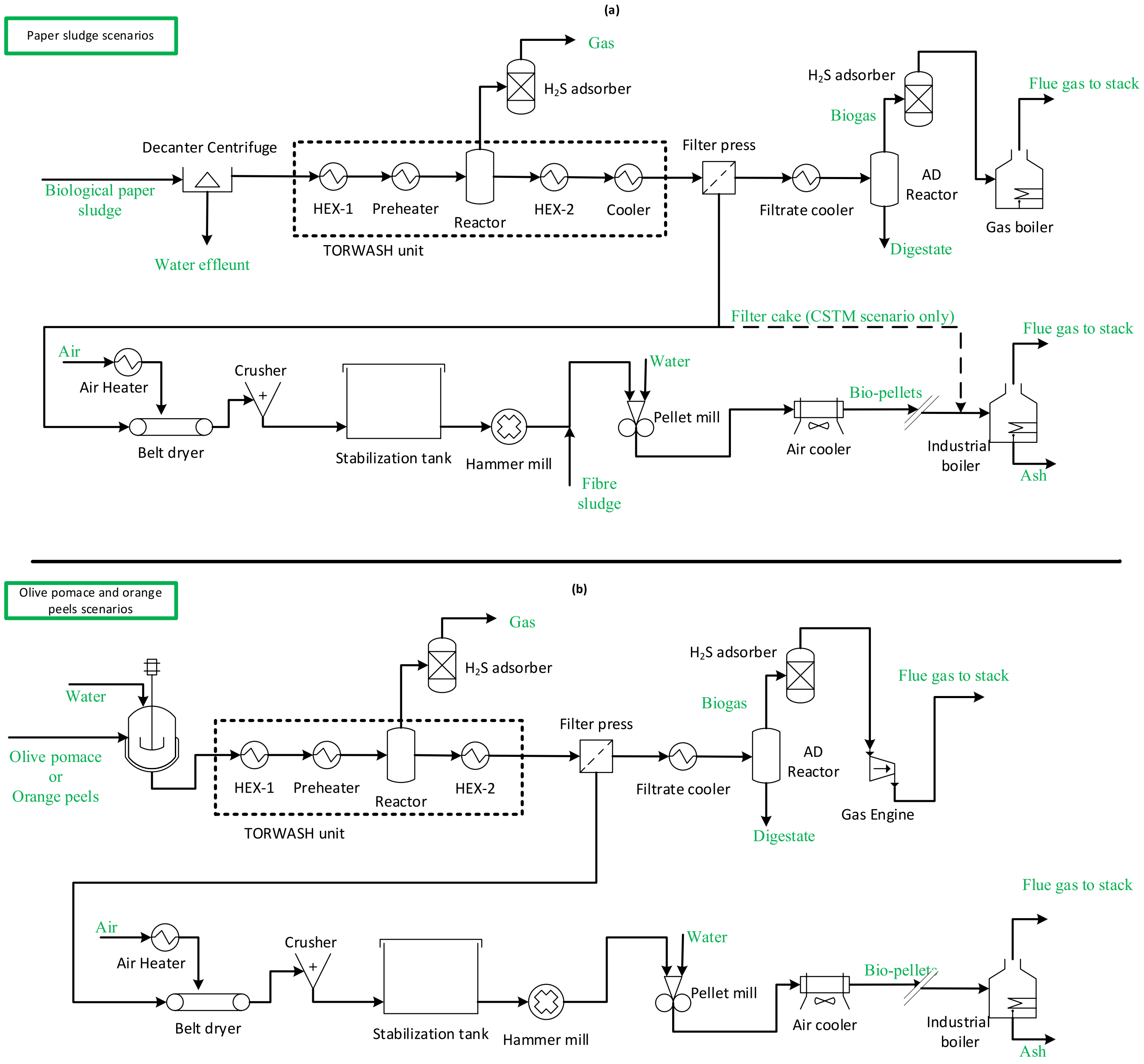

Fig. 1. Process schematics for mild HTC of (a) biological paper sludge and (b) olive pomace and orange peels feedstocks; CSTM = custom scenario, AD = anaerobic digestion, HEX = heat exchanger. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

hexane extraction, refined, and blended to obtain olive pomace oil [22] and orange peels are often composted or used as animal feed [23]. However, for the purposes of this study, anaerobic digestion is chosen as the reference case to compare energy recovery with that from the HTC process. The biogas obtained via anaerobic digestion is cleaned to remove H_2S and used in a gas engine for on-site power generation. The process parameters and biomethane potential were obtained from the literature [24,25].

2.2. Feedstock characteristics and production

A summary of feedstock characteristics, feedstock production rates for a given industrial location and corresponding capacity of the HTC and REF processes are listed in Table 1. For the paper sludge feedstock, the HTC process treats the biological paper sludge without the need for fibre sludge as a dewatering aid. Therefore, the amount of fibre sludge as a waste stream was reduced to a minimum purge volume and the remainder reused in the paper mill for additional Kraftliner production. In the modelling, it is assumed that wet solids are obtained from this fibre sludge purge stream using existing dewatering steps in the REF scenario and then combusted in a boiler for steam generation as indicated in Fig. 1(a).

2.3. Process modelling approach

Processes are evaluated using Aspen Plus V12 software [26] to obtain mass and energy balances. The processes are modelled as steady state where batch and intermittently operated equipment are modelled based on average performance over time. The equipment performance is assumed to not be affected by scale. The thermodynamic method selected is the NRTL-RK method (non-random two-liquid model for the liquid phase with Soave-Redlich-Kwong equation of state for the gas

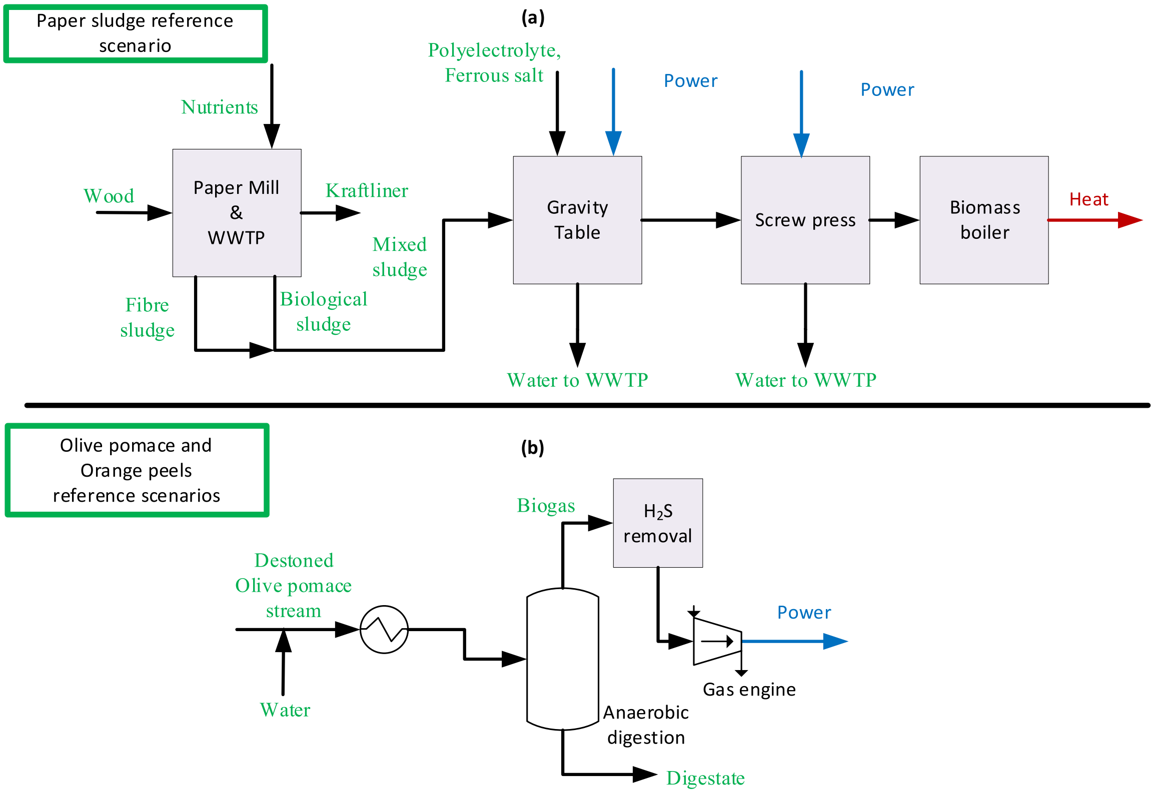

Fig. 2. Process schematics of reference (REF) scenarios for (a) biological paper sludge and (b) olive pomace or orange peels; WWTP = wastewater treatment plant. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Feedstock characteristics and capacity of the representative hydrothermal carbonization (HTC) and reference (REF) processes where plant capacity corresponds to typical industrial production rates of the feedstock streams (at one location); BS = biological paper sludge, FS = fibre sludge, ar = as received, wt = weight.

	Paper Sludge	Olive Pomace	Orange Peels
Feedstock starting points			
Feedstock Production/Plant Capacity (wet basis) ($tonne_{ar}/yr$)	Reference case: 64285 (BS) + 204545 (FS) HTC case: 64285 (BS) + 60606 (FS)	9600	2300
Dry matter content (wt%)	3.5(BS), 1.65(FS)	19.63	20.0
Plant operating time (hours/year)	8600	960	3200
Temperature, as produced (°C)	25	15	15
Feedstock molecular formula			
Bio-organics	$CH_{1.67}O_{0.54}$	$CH_{1.6}O_{0.41}$	$CH_{1.8}O_{0.85}$
Feed composition (wt%, dry weight basis)			
Bio-organics	77.1	95.1	96.5
Nitrogen	7.1	2.1	0.9
Sulphur	1.3	0.2	0.1
Phosphorous	1.4	0.2	0.1
Potassium	0.4	1.4	0.8
Calcium	1.6	0.2	0.7
Rest ash	11.1	0.9	0.9

phase). Non-condensable gases are modelled with Henry's law.

The process model uses two sub-streams for fluid and solids, respectively, where the dry matter (all components except water) is distributed between either of the two phases. The components are represented by bio-organics, nitrogen, and different ash components. Representative chemical components are selected for the nitrogen and ash. A predefined component 'BIOMASS', from the National Renewable Energy Laboratory (NREL)'s ASPEN Plus database [27] is used to model the bio-organics and inputted as a single pseudocomponent, $C_xH_yO_z$, where x, y and z are determined based on elemental analysis of the residue streams (Table 1). For FS, the molecular formula used is $CH_{1.41}O_{0.39}$ based on literature values [28]. The feedstock streams are assumed to be received at ambient pressure.

Of special interest and included in the modelling are nutrient and inorganics flows in the HTC process, which is relevant for the operation of the wastewater treatment plant in the paper mill, anaerobic digestion and environmental assessments. For this purpose, elements N, P, K, S, Ca and "rest inorganics" are included in the model as NH_4NO_3, P_2O_5, KCl, S, $CaCO_3$ and NaCl, respectively.

In the Torwash mild HTC reactor, the feedstock reacts in the presence of water, under high pressures and temperature (Table 2), to give an upgraded feedstock with gas evolution, mainly CO_2 with some H_2S. The optimum operating temperature was determined through lab experiments [6]. To represent this transformation in the modelling, a global reaction (Eq. (1)) is used:

$$\boldsymbol{a}.Bioorganics + \boldsymbol{b}.S \rightarrow \boldsymbol{c}.Upgraded\ Bioorganics + \boldsymbol{d}.CO_2 + \boldsymbol{e}.H_2O + \boldsymbol{f}.H_2S \qquad \text{Eq. 1}$$

The upgraded bio-organics are also modelled as $C_xH_yO_z$, similar to the feed bio-organics, using different values for x, y and z based on actual measurements from lab-experiments (Table 2). A list of elements and their corresponding values used for calculations are given in the supplementary material. The carbon to sulphur ratio in the gas, i.e., between CO_2 and H_2S, is maintained the same as the C:S ratio in the feed. The stoichiometric factors are obtained from the elemental balance through the Torwash HTC reactor.

The schematic for the Torwash mild HTC process is depicted in Fig. 3.

Table 2
Torwash mild hydrothermal carbonization (HTC) process parameters and retention of select elements in the dewatered solids (relative to the feedstock) for different biomass residue feedstocks.

	Biological paper sludge	Olive pomace	Orange peels
Torwash treatment temperature (°C)	200	195	200
Torwash treatment pressure (bar)	16–19		
Heat of reaction, ΔHr (25 °C, 1 bar) (kJ/mol)	−63.85	3.93	−29.85
Upgraded bio-organics	$CH_{1.5}O_{0.34}$	$CH_{1.48}O_{0.33}$	$CH_{1.23}O_{0.41}$
Element retention in dewatered solids			
N	35 %	91 %	81 %
P	100 %	17 %	5 %
K	23 %	9 %	5 %
Ca	100 %	100 %	85 %
S	59 %	93 %	66 %
Rest ash	100 %	31 %	40 %

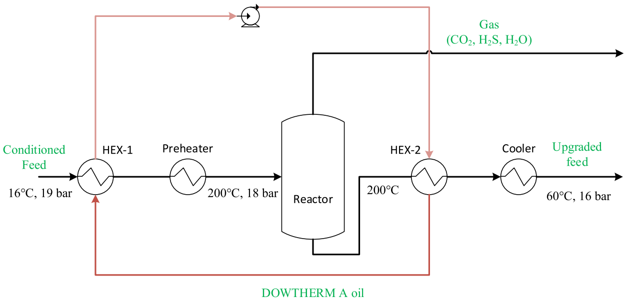

Fig. 3. Torwash mild hydrothermal carbonization (HTC) unit modelling scheme; HEX = heat exchanger.

A regenerative heat integration is carried out between the feed and product stream using a heating oil loop. A preheater further heats the feed to the desired temperature while the cooler cools the reactor outlet to 60 °C. The heat of reaction is taken into account as an additional heat requirement in the cooling oil loop and calculated using the organics and upgraded bio-organics heat of combustion using correlations available in literature [29]. The dissolution of solids for each of the feed components is obtained from elemental analysis of the dewatered cakes after Torwash and used to calculate the retention value (mass of element in pressed cakes/mass of element in Torwash feed) for elements of interest in the solid fraction.

The lab- and pilot-scale experimental methods are described in detail in a related paper [6]. Briefly, at lab-scale, batch experiments are performed in a 20 L autoclave and the treated material is dewatered using a carver die unit. Continuous, pilot-scale experiments are conducted with a Torwash reactor with a maximum capacity of 50 kg/h on-site at a paper mill, an olive mill and an orange processing plant. The treated material is dewatered using a pilot-scale membrane filter press. The proportion of dry matter from the feedstock in the dewatered cakes, i.e. distribution factor = mass of dry matter in pressed cake/mass of dry matter in filter inlet, for lab- and pilot-scale experiments is presented in Table 3. The differences in dry matter content of the solids between lab and pilot-scale tests is attributed to the different dewatering methods and general differences and difficulties in scaling up target process conditions to continuous, pilot-scale.

Pumps, heaters, heat exchangers and phase separators were modelled using standard models. The Torwash reactor was modelled using a combination of a stoichiometric reactor black box separator, and flash vessel for water/vapor separation. The filters, dryers, boiler, and gas engine were modelled using constructs (combinations of standard blocks [30]). The anaerobic digestion was modelled using a stoichiometric reactor producing biomethane according to the Buswell equation [31]. Only part of the biomass is converted into biogas, as characterized by the anaerobic digestibility as listed in Table 4. The process equipment parameters (Table 4) were obtained from experimental data or from the literature.

Table 3
Key data for the dewatering stage of the model for different feedstocks; distribution factor is the mass of dry matter in pressed cake/mass of dry matter in filter inlet; lb = lab, pl = pilot, wt = weight.

	Distribution factor of dry matter (wt/wt)		Pressed cake dry matter (wt%)	
	lb case	*pl* case	*lb* case	*pl* case
Biological paper sludge	0.43	0.43	61	42
Olive pomace	0.71	0.62	68	58
Orange peels	0.84	0.43	59	42

In Table 4, data on filter press power, hammer mill and pellet mill power (only olive pomace and orange peels), anaerobic degradability of filter press effluent, and power requirement for AD system were obtained from experiments. Preconditioning power was obtained from literature and equipment vendor brochures [32–34]. Heat losses to the environment are based on typical numbers assumed for such studies while for TORWASH an assumption was made based on advice given by industry experts. The crusher power was assumed to be 30 % of the hammer mill power based on experts and is line with values from the literature [35]. The hammer and pellet mill power for paper sludge feedstock were obtained from literature [36,37]. The gas engine efficiency was obtained from literature [38]. The remainder are process design choices for the study.

2.4. Key performance indicators

For the technical evaluation of the processes, three key performance indicators (KPIs) are selected: 1) bio-pellet quality, 2) process energy efficiency and 3) relative feedstock utilization.

The bio-pellet quality is assessed by comparing bio-pellet composition as calculated by the process model with the EN*plus* standard for

Table 4
Key process parameters for the models; ar = as received; HTC = hydrothermal carbonization, LHV = lower heating value.

Process Parameter	Paper sludge	Olive pomace	Orange peels
Preconditioning power (kW-hr/$tonne_{ar}$ feed)	5	0.1	0.1
Target dry matter concentration in Torwash mild HTC inlet (wt%)	10	4	4
Heat loss to environment by Torwash unit (% of total heat input)	5		
Filter press power requirement (kW-hr/m^3 feed)	1.3	1.2	1.2
Hot air inlet temperature of dryer (°C)	80		
Temperature difference of air outlet of dryer and dried filter press cake (°C)	20		
Crusher power energy requirement (kW-hr/$tonne_{ar}$ feed)	3.6	2.4	3.8
Hammer mill power requirement (kW-hr/$tonne_{ar}$ feed)	12	8	12.5
Pellet mill power requirement (kW-hr/$tonne_{ar}$ feed)	48.9	32.5	57.5
Anaerobic degradability of filter press effluent (%)	62	57	77
Anaerobic digestion system power requirement (kW-hr/m^3 feed)	0.78	0.16	0.24
Heat losses in steam boiler (% of total energy recovered)	3		
Gas engine efficiency (% of gas LHV)	35		

pellets to be marketed as equivalent to premium wood pellets [39], and against the Illinois No. 6 bituminous composition [40] as a coal replacement.

The process energy efficiency indicates energy retention in the final solid product, i.e., energy content of bio-pellet bio-organics compared to the energy content of the feedstock bio-organics (Equation (2)). The lower heating value (LHV) is calculated based on the elemental bio-organics composition using correlations [40,41] and $\dot{m}$ represents the mass flow rate.

$$Process\ energy\ efficiency = \frac{(\dot{m}.LHV)_{upgraded\ bio-organics\ (db)}}{(\dot{m}.LHV)_{bio-organics\ (db)}} \qquad \text{Eq 2]}$$

Relative feedstock utilization (Equation (3)) is the ratio between the net primary energy that is obtained through the process compared to the energy content of the feedstock. Since this net energy is in the form of both heat and power, an energy conversion is introduced to convert it into primary energy and allow for an equal-basis comparison. It is assumed that the energy conversions LHV efficiencies are the same as those in a typical energy generation system (i.e., 0.88 for heat production, η_h and 0.35 for power generation, η_e). The feedstock utilization indicates how well the feedstock has been used for energy purposes either through pellets, biogas and/or non-pelletized filter press solid cakes (CSTM case for paper sludge) while also taking into account how much the feedstock is upgraded in quality because of the mild HTC process.

$$Feedstock\ utilization = \frac{Net\ energy\ obtained\ from\ process}{Energy\ content\ of\ the\ feedstock} = \frac{\left(\frac{Heat\ Export}{\eta_h} + \frac{Power\ Export}{\eta_e}\right) - \left(\frac{Heat\ Use}{\eta_h} + \frac{Power\ Use}{\eta_e}\right)}{(\dot{m}.LHV)_{bio-organics\ (db)}} \qquad \text{Eq. 3]}$$

3. Results and discussion

3.1. Mass and energy balances

The mass and energy balances for the mild HTC process for each feedstock are calculated based on lab-scale experiments and are presented graphically in Fig. 4. The mass flow rates are listed in terms of total mass flow rate, biogas mass flow rate and dry matter mass flow rate. The lab scale experimental data were translated into the performance of a continuously-operated industrial scale assuming a plug flow in the TORWASH reactor. The energy balance is listed as heat and power duties. The numbers have been rounded to the nearest whole number. The mass and energy balances for pilot plant cases are available in the supplementary material.

The scenarios show significant differences in total mass flow values, with olive pomace having the largest mass flows and orange peels the smallest. Comparing the feedstocks to each other, differences can be seen in the amount of dry matter dissolved during the Torwash mild HTC process (especially significant for orange peels) and in the subsequent conversion of dissolved dry matter into biogas during anaerobic digestion. The mass flow rates in the pelletization and boiler sections are much lower than those in the rest of the process. The results also show that valorisation of biogas is a significant contributor to the energy balance for all feedstocks (and the largest contributor for paper sludge).

An advantage of the modelling approach used in this study is that reaction stoichiometry is available and more detailed information, including specific elemental concentrations (e.g., N, P, K), chemical oxygen demand, and reaction heat can be predicted, which has an influence on the energy balance of the simulations. Other HTC and wet torrefaction simulation studies often use RYield reactors (e.g., Refs. [7,8]) when only yields are known. In the current study, a more fundamental approach is taken by using a reaction with reaction coefficients. Another difference in the current study is that gas generation during the mild HTC process is negligible, whereas typical wet torrefaction processes report approximately 10 % gas generation from the process [42,43]. These higher reported numbers could be due to harsher process conditions and longer residence times of typical HTC, leading to more decomposition of the feedstock and hence more gas formation. The feedstock can play a vital role as well, since not all biomass is the same, with orange peels resulting in more gas formation per kg of dry matter fed into the anaerobic digestion (AD) system than paper sludge and olive pomace.

3.2. Pellet quality

Specific quality parameters (Table 5) are used to assess if the bio-pellets produced from paper sludge, olive pomace and orange peels meet market standards. These quality parameters are taken from the most commonly used bio-pellet standard in the European Union, the EN*plus* B standard [39] for woody pellets from a raw material of virgin or chemically untreated wood. The bio-pellet nitrogen and sulphur content are largely in agreement between the experimental and modelling values for the orange peel and olive pomace feedstocks. Slight variations between experimental and model results can be explained by typical restrictions faced during lab experiments, e.g., feedstock and instrument variations. The differences between lab and model pellet parameters are most significant for the biological paper sludge feedstock, in particular due to an inflated pellet ash and nitrogen content calculated in the model. The N content in the simulations is higher likely as a consequence of the modelling approach which is integrated with experimental results. The ash content is higher because in the mass balance 100 % ash retention in the solids was reported in the lab experiments based on elemental analysis. In reality, it is probable that some ash components end up in the liquid phase during dewatering. It should also be noted that even for experimental data which has lower ash and N values, these parameters still do not meet the target quality standards (Table 5).

In comparison with the global bio-pellet standards, mild HTC of the target feedstocks is not able to produce pellets that meet the En*plus* B standards for wood pellets, mainly due to higher nitrogen and sulphur content in the bio-pellets and higher ash content in the case of paper sludge. The lower pellet quality can be traced back to the lower quality of the waste feedstocks, with high ash content with varied elemental composition, leading to poorer quality bio-pellets than those made from virgin wood feedstock. However, bio-pellets from all three feedstocks meet the EN*plus*B standards for lower heating value (LHV), demonstrating the effectiveness of upgrading the wet sludges with mild HTC. The pellets produced from orange peels and olive pomace have a lower ash and sulphur content when compared to coal (Table 5), and could be suited for applications that do not require premium quality pellets, such as in the power station sector [44]. The bio-pellets created from HTC of biological paper sludge are inferior to all other pellets used in the comparison and should therefore be used on-site, as suggested in the custom modelling case, although previous research has also shown that these solids could also be suitable for partial replacement of coal in blast furnace injection processes [45].

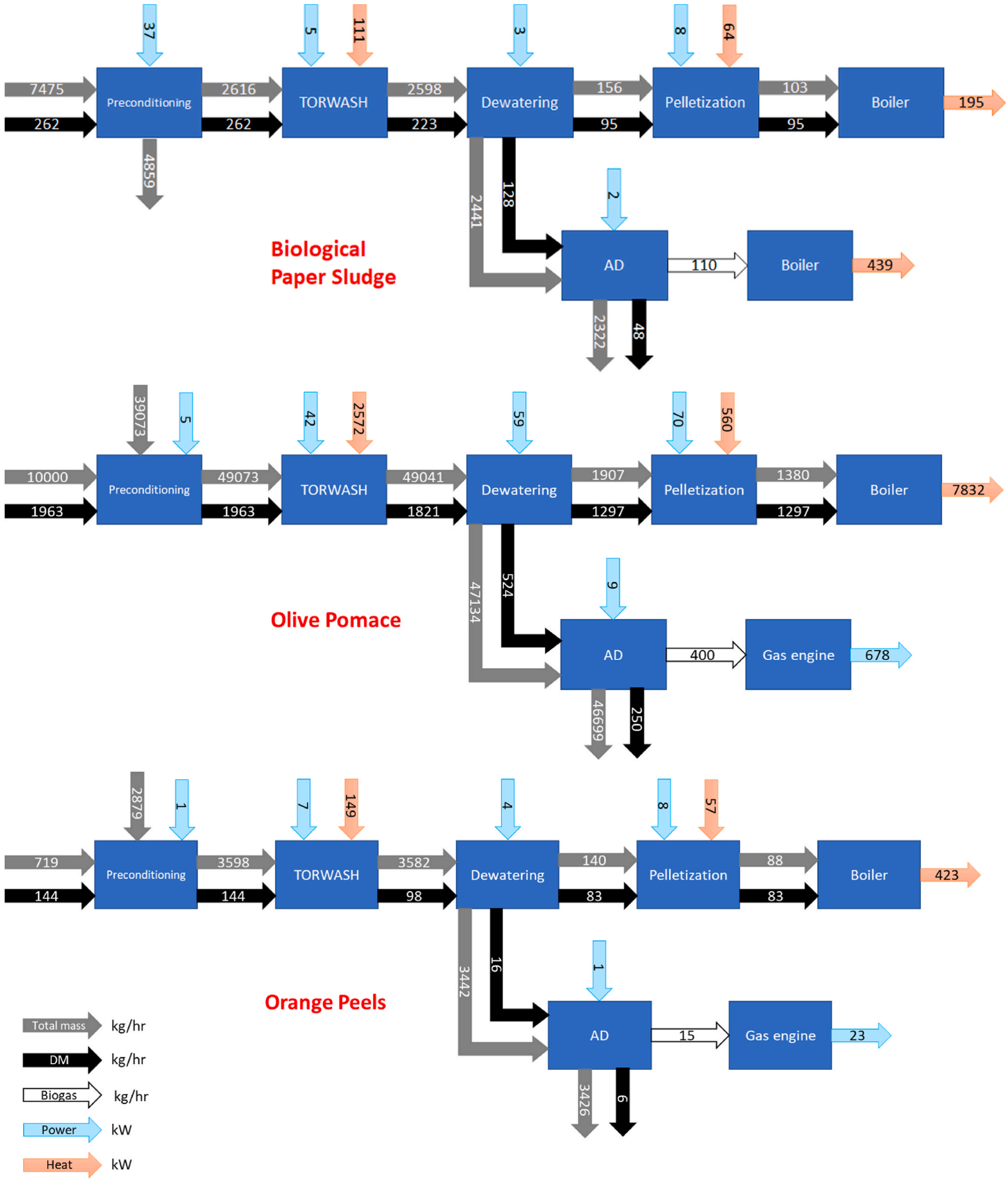

Fig. 4. Mass and energy balances for the hydrothermal carbonization (HTC) process for the three feedstocks, lab-scale (lb) cases; AD = anaerobic digestion, DM = dry matter.

Table 5
Pellet quality for different feedstocks from modelling and experimental data for lab-scale cases, and comparison to ENplus B [39] and coal Illinois No. 6 bituminous [40] standards; ar = as received, db = dry basis. Feedstocks are paper sludge (PS), olive pomace (OLP) and orange peels (ORP).

Parameter			Model			Experimental		
	EN*plus* B	Coal	PS	OLP	ORP	PS	OLP	ORP
Moisture [wt %]	≤10	2	7	6	6	4.9	<10	<10
N (db) [wt%]	≤1	1.3	6.8	2.9	1.6	4.2	2.2	1.6
S (db) [wt%]	≤0.05	2.9	2.1	0.2	0.1	1.3	0.16	0.09
Ash (db) [wt %]	≤2	13.2	41	1.1	2.3	30	0.9	2
LHV (db) [MJ/kg]	–	25.2	18.2	26.3	22.2	18.5	25.8	22.1
LHV (ar) [MJ/kg]	≥16.5	25.2	16.8	24.6	20.7	17.5	23.3	19.9

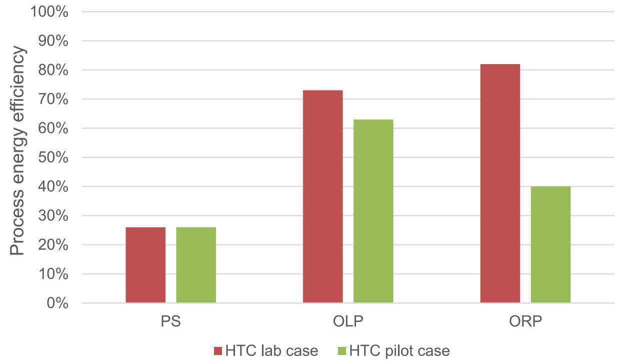

Fig. 5. Comparison between HTC cases for process energy efficiency (representing energy retained in solid pellets).

3.3. Process energy efficiency

The process energy efficiency accounts for the bio-organics captured in the pellets and the higher heat content (i.e., LHV on a dry basis) of the upgraded bio-organics relative to that of the feedstock. Fig. 5 depicts the mild HTC process energy efficiency for the lab and pilot cases of all three feedstocks. Process energy efficiency for the lab-scale experiments for olive pomace (73 %) and orange peels (82 %) are slightly higher than reported efficiency in lab studies with wet feedstocks [46] including grape pomace [7] and fruit peels [47]. Process efficiency for HTC of paper sludge (26 %) is lower than that of studies of municipal wastewater sludges in the literature, which report approximately 40 % process energy efficiency [48,49].

For paper sludge, the lab and pilot cases show the same process energy efficiency, while for olive pomace and orange peels the process energy efficiency at pilot scale is lower than that at lab scale. For olive pomace and orange peels, less dry matter is captured in the solid fraction during pilot-scale dewatering when compared with lab-scale dewatering experiments. This may be due to dewatering methods used, where a smaller volume and higher pressure was applied in lab-scale dewatering when compared to pilot-scale. In addition, operational issues associated with the higher solids content of these feedstocks led to higher HTC residence times at pilot scale than at lab scale, leading to more organic dissolving during HTC. It is hypothesized that this is most significant for orange peels because of their higher content of soluble components such as sugars. In general, further optimization of the process for different feedstocks at pilot scale is required in future studies to address the trade-offs between process severity (temperature, residence time), dewaterability and solids yield.

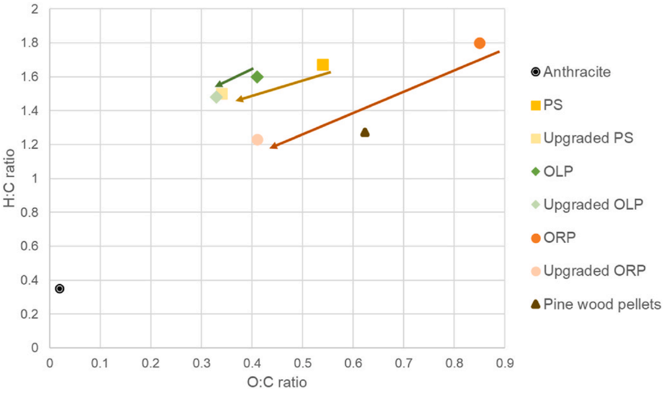

Fig. 6. Van Krevelen diagram for untreated feedstocks and upgraded feedstocks after mild HTC treatment compared to Anthracite and pine wood pellets [51]. Feedstocks are paper sludge (PS), olive pomace (OLP) and orange peels (ORP). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.4. Feedstock utilization

The degree to which each feedstock is upgraded during the mild HTC process is depicted in a van Krevelen diagram (Fig. 6), a type of graph that was initially used to represent various types of coals and their chemical constitution [50]. The diagram presents the ratios of H:C and O:C in the untreated feedstocks and the feedstocks after mild HTC, with anthracite coal and pine wood pellets depicted as references. Following mild HTC, the feedstocks are upgraded and move closer to the elemental ratios of anthracite coal, i.e. a premium solid fuel. Chemically this is seen through a reduction in mainly the O:C content which improves the overall heating values of the solid. Lower H:C and O:C ratios are desirable in solid fuels because this corresponds to less smoke and water vapor and increased energy obtained during combustion [7].

Fig. 7 compares feedstock utilization between the reference and mild HTC scenarios. For all feedstocks, the HTC process results in a higher feedstock utilization than the corresponding reference scenario. In addition, feedstock utilization is lower for the pilot experiments than the lab experiments for all feedstocks. This is attributed to the higher heat input required for drying during the pilot-scale process, where dewatering isn't as efficient as dewatering at lab-scale, resulting in wetter solid cakes.

For paper sludge feedstock, the HTC cases (lab, pilot and custom case) improve feedstock utilization by up to three times when compared to the reference scenario. This is attributed to the HTC process

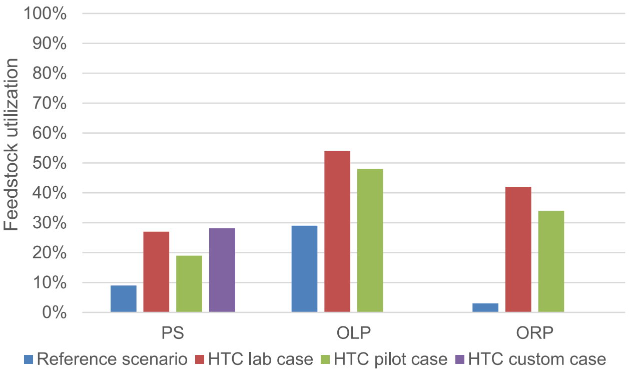

Fig. 7. Feedstock utilization comparison between reference and hydrothermal carbonization (HTC) scenarios for different feedstocks: paper sludge (PS), olive pomace (OLP) and orange peels (ORP). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

improving the dewaterability of the sludge relative to the reference scenario and the use of both the dewatered solids for combustion and the liquid for biogas generation. In addition, in the HTC process heat demand is limited through efficient heat integration in the Torwash reactor. In the custom case for paper sludge, where the solids drying and pelletization are omitted and the dewatered solids are combusted on-site, the feedstock utilization is highest, and improved by almost 50 % relative to the pilot case where drying and pelletization are included. This makes the custom case an attractive option for paper mills and highlights the advantages of tailoring an overall HTC process to be suitable to real-world conditions and industry requirements.

For olive pomace, the HTC process results in a feedstock utilization 1.7 times better than the reference scenario. In the reference scenario biogas is generated from the olive pomace but a large amount of dry matter remains in the digestate. This is reflected in a higher concentration of chemical oxygen demand (COD) in the digestate in the reference scenario when compared to the HTC cases. This impact will be more pronounced in a future economic or in the forthcoming life cycle study [52].

The orange peel feedstock showed the greatest improvement in feedstock utilization with mild HTC, up to 14 times the utilization when compared to the reference scenario. In the reference scenario, the anaerobic digestion has a high heat demand and an overall low biogas generation, which could be attributed to the presence of terpenes such as D-limonene, which are known to be toxic to microorganisms [25]. The heating value of the orange peels is also greatly improved during HTC, as evidenced by the decreasing O:C ratios (Fig. 6).

Remarkably, the amount of dry matter captured in the Torwash HTC and dewatering steps has only a limited effect in on the feedstock utilization. In the HTC process, less dry matter is captured in the solids during dewatering in the pilot case when compared to the lab case, but this impact is not reflected in the biomass utilization to the same extent. This is because the dry matter not captured in the solid cakes is utilized for biogas generation in anaerobic digestion. Other recent studies have also explored the valorisation of the liquid fraction after HTC, showing its relevance for a high process energy efficiency [48,49,53,54]. This makes biogas generation an important step for this process scheme and a relevant consideration for scaling-up of HTC processes.

4. Conclusions

Mild hydrothermal carbonization (HTC) using the Torwash mild HTC process shows promising technical prospects for treating industrial and agricultural wet residue streams, including paper sludge, olive pomace and orange peels. Mild HTC improved the dewaterability of the wet residue streams, resulting in a solid and liquid fraction. The solid fraction can be converted to bio-pellets to generate dispatchable heat and the liquid stream can be anaerobically digested to produce biogas for heat or power generation. Process modelling indicates that both the solid and liquid-generated products are equally important for improving the overall process energy efficiency. Process efficiency is also improved by effective heat integration in the Torwash reactor.

Bio-pellets produced from all three feedstocks do not meet all of the market quality standards for premium wood pellets but do meet the standards for LHV, indicative of successful upgrading of energy density. Bio-pellets from olive pomace and orange peels are suitable candidates to be marketed as a coal replacement based on quality characteristics and market standards. For the pilot process, energy efficiency is highest for olive pomace (63 %), followed by orange peels (40 %) and paper sludge (26 %). Overall feedstock utilization is improved with HTC relative to the reference process for a given feedstock.

Additional benefits were also identified for the treatment of paper sludge, namely treating only biological sludge via HTC and omitting drying and pelletization of the dewatered solids for direct combustion in an on-site boiler. Both of these custom process adjustments could lead to significant cost improvements.

This paper demonstrates the potential of mild HTC for upgrading wet biomass residues, including sludges and agricultural residues, based on both lab- and pilot-scale data and modelling. Further research includes evaluating the process kinetics to better understand differences between lab and pilot-scale performance. In addition, the feasibility of the process must be assessed with respect to environmental and economic factors.

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CRediT authorship contribution statement

Sayujya Shah: Formal analysis, Investigation, Methodology, Visualization, Writing – original draft. **Jan Wilco Dijkstra:** Conceptualization, Investigation, Methodology, Supervision, Validation, Writing – review & editing. **Heather E. Wray:** Conceptualization, Funding acquisition, Resources, Writing – review & editing.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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