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Mass and Energy Integration Study of Hydrothermal Carbonization with Anaerobic Digestion of Sewage Sludge

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Abstract

Anaerobic Digestion is the most common process used for energy generation from sewage sludge but only one half of the sewage sludge is susceptible to biodegradation. Hydrothermal Carbonization is considered an option for harness all the energy embedded in sewage sludge because of its high-value products (Hydrochar and Process waters). The integration AD followed by HTC is a recent approach that is still under development. The challenge is to provide evidence for coupling HTC with the existing infrastructure at wastewater treatment works. In this work, a mass and energy integration study of the potential of coupling HTC with AD for sewage sludge treatment was evaluated. Six proposed process configurations were built using primary sludge, secondary sludge and a mix, in order to evaluate net waste generation, fate of nutrients, net energy production and potential economic benefits. The proposed scenarios showed an overall total solid and COD reduction up to 68 and 66% respectively. The inclusion of hydrochar as a fuel source increased the net energy production 10 times compared when only biogas is considered as an energy source. The potential struvite production ranged from 0.02 to 0.06 kg per tonne of sludge treated. Scenarios with 250°C thermal treatment temperature provided better economic benefits when struvite and hydrochar are considered.

Keywords: *Hydrothermal Carbonization, Anaerobic Digestion, Process waters, Sewage sludge.*

1. Introduction

The use of sewage sludge as a feedstock for renewable energy generation is gaining growing attention. In the UK, the estimated annual production of sewage sludge is 1.4 million tonnes (dry weight) [1] in which the water industry generates approximately 800 GWh/yr of electrical energy [2]. The sewage sludge produced at waste water treatment works (WWTW) has been commonly treated by anaerobic digestion (AD) due its large organic matter content which favours biogas production and brings multiple environmental benefits [3-5]. According to Berglund and Börjesson [5], 40-80% of the energy content of the biogas produced from sewage sludge in a WWTW corresponds to the overall energy input in a large-scale biogas plant. Furthermore the inclusion of AD brings associate benefits as sludge mass reduction, odour removal and pathogen reduction [6]. Nevertheless, one of the main limiting steps of processing sewage sludge via AD is the solubilisation of biodegradable organic compounds through hydrolysis [7, 8]. For that reason, the resulting digested sludge (digestate) from anaerobic digestion still contains large amounts of non-easy biodegradable organic matter that with a proper treatment can be harnessed for additional energy production [3]. Many pre-treatment techniques based on thermal, biological, chemical, mechanical and physical processes have been studied individually and in combination, with the objective to enhance the biodegradability sewage sludge and digestate by breaking down complex organic molecules in order to increase the solubility of organic compounds and produce more energy coming from biogas [6]. However, the main drawback has been the economic constraints for scale-up and commercialisation [6].

Hydrothermal processing is currently being considered as an novel alternative technology to further harness energy from sewage sludge and digestate [9-11] and to reduce the issues related to current disposal of final solid products. Hydrothermal processes (HTPs) involve the treatment of biomass in hot compressed water that can produce either solid hydrochar, a biocrude or a syngas, along with process waters, depending on process temperature and pressure. In addition, HTPs applied to sewage sludge processing not only help to inactivate pathogens and further bacterial activity after disposal, but also produce valuable by-products like hydrochar. In recent years, some researchers have proposed HTC as an alternative to harness better the properties from the sewage sludge and reducing waste generation [3, 9-

18]. HTC main objective is to transform biomass into a carbon-rich product applying heat (180-250°C) and pressure during a certain period of time [9, 18-21]. The main advantage of HTC is that it is carried out in presence of water avoiding the energy-intensive drying step required for thermal processes [3, 9, 21, 22]. Furthermore, the resulting products from the HTC are a solid hydrochar that can be used as a soil amender or fuel source and a process water rich in carbon and organics that can be used for enhanced biogas production [3, 12-15, 20, 23-29].

There are several studies of HTC applied to sewage sludge at lab-scale focused in the process-conditions and their influence on the by-products [3, 12, 18, 23-25, 27, 30-36]. Nevertheless, only few studies have focused in the potential technical performance of industrial-scale of the HTC plants [9, 12, 19, 37-39]. Companies as SunCoal Industries, AVA-CO2 and Ingelia [40-44] have developed commercial applications for HTC, but this technology is still under development. CarboRem and Terranova Energy have developed modular HTC units that makes easier the HTC integration within a waste water system [42, 44]. According to the study carried out by Child [45] and Lucian and Fiori [19], a HTC plant can cost from €1.7 up to €10million depending on treatment capacity (8,000 to 50,000 tonnes of feedstock). Nevertheless, both studies focused on hydrochar production using mostly lignocellulocic biomass as main feedstock.

Other studies have found that the biogas production at AD plants using thermal hydrolysis as pre-treatment is better than using hydrothermal treatments as post-treatment, but the use of hydrochars despite of being considered a low-grade fuel gives an added value boosting up the energy production up to 179% compared with the 43% of the thermal hydrolysis [3]. This makes the integration of hydrothermal treatment as post-treatment a promising option to harness the energy from sewage digestate [3]. Nonetheless, those studies just mention the energetic benefits of integrating hydrothermal processes with AD, but did not consider other implications as energy consumption, potential economic benefits and mass and energy balances [3, 12].

The integration of HTC as a post-step after the AD is a recent approach and some authors suggest that the integration of a hydrothermal treatment step into waste water systems are energy positive [3, 9, 12, 14, 15, 17, 23-25, 27, 46-49]. The use of hydrochars as a source of energy, despite being considered a low-grade fuel, can boost up the energy production up to 179% compared with the 43% of the thermal hydrolysis used as a pre-treatment [3]. This makes the integration of hydrothermal treatment as post-treatment a promising option to harness the energy from sewage digestate. Medina-Martos et al., [9] made a techno-economic and life cycle assessment of an integrated HTC system for sewage sludge. They found that the integration of the HTC system helps to reduce environmental impact compared with the conventional WWT configutarion with AD integrated. The energy efficiency can increase up to 14% but the costs of integrating the HTC+AD system increases up to 42% compared with the conventional WWT with AD.

Considering this novel approach of integrating HTC+AD, the current state of the art and in order to meet sustainable environmental targets in sewage sludge management such as waste minimisation, resource recovery and the overall reduction of treatment costs, it is imperative to provide evidence from integration studies coupling HTC with the existing infrastructure and treatment units at WWTWs. Therefore, the main objective of this study is to assess the integration of HTC with AD through robust mass and energy balances from several proposed process configurations using different sewage sludge streams and supported by results obtained from experimental work. Results were used to evaluate and compare products yields, waste generation, net energy implications and potential economic benefits, in order to integrate HTC as part of a comprehensive sewage sludge management strategy.

2. Material and methods

2.1. Process description

The overall proposed process of hydrothermal carbonization integration to anaerobic digestion of sewage sludge is divided in four main processing areas **(See Figure 1)**. Firstly, the feed undergoes mesophilic anaerobic digestion processing (37 °C) producing digestate and biogas. The next process comprises a thickener which concentrate the digestate to 15% of solids. Next, the thickened digestate is submitted to thermal processing (160 or 250 °C) converting it into process water and hydrochar – i.e., for the energy balance, the thermal recovery efficiency from the heat exchanger is considered in this stage. The treatment temperatures were selected based on previous studies to emulate and compare the conditions of hydrothermal hydrolysis (160 °C) and HTC of sewage sludge in different scenarios [3, 12]. Then, a centrifuge is used to separate the hydrochar (solid fraction) from the process water (liquid fraction). The hydrochar is considered as a potential fuel source based on their higher heating values (HHV), but non-energy recovery process is considered at this step. On the other hand, the process water is anaerobically treated at mesophilic conditions in a second reactor producing biogas. The biogas produced at the first and second AD reactors are mixed combusted in a combine heat power (CHP) unit to produce the energy for the system. The energy produced from the biogas is used to cover the energy requirements of the hydrothermal system and the exceeding energy is used for other equipment.

The aim of the process configuration is to integrate the hydrothermal treatment as a post-treatment to the anaerobic digestion of sewage sludge. The use of different sewage digestate is to compare the energy production between them since the primary sludge seems to have high organic content. As stated by Pérez-Elvira and Fdz-Polanco [50], it is expected that the best option would be to segregate primary and secondary sludge in order to produce more energy in the overall system and that is the overall hypothesis of the work conducted

The assumptions adopted as a basis for the mass and energy balance of the different scenarios built in this study are presented in **Table 1**.

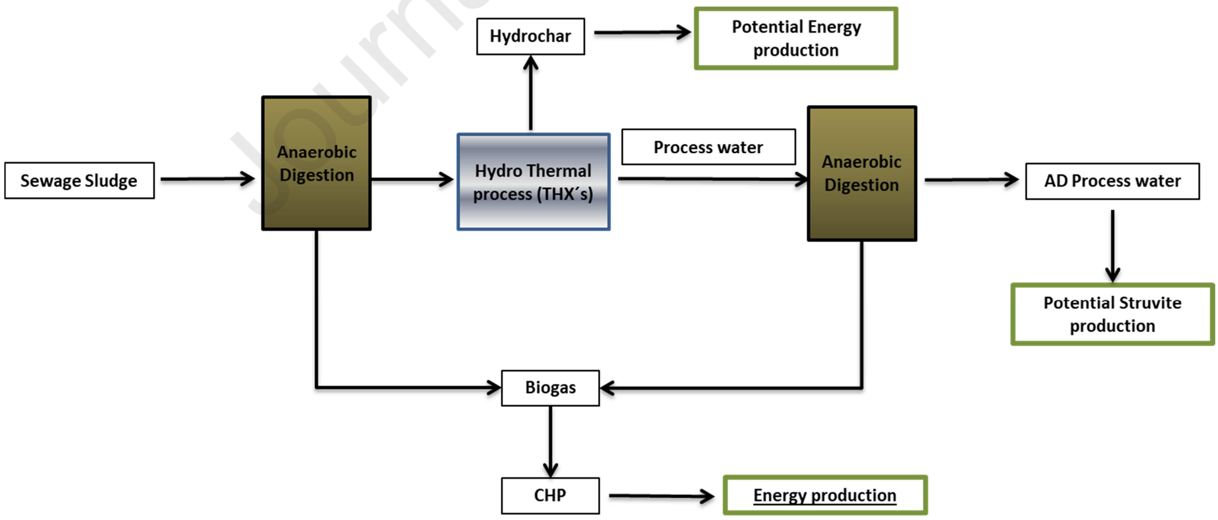

Figure 1.- General process diagram for the proposed scenarios.

2.2. Mass and energy balances

This study is based on the experimental results obtained from laboratory experiments carried out at University of Leeds (UK). Six scenarios were tested using three different sewage sludges (Primary, Secondary and Primary-Secondary Mix) following the proposed process description and undertaking hydrothermal processing at two different temperatures (160 and 250 °C).

2.2.1. Sludge samples

Sludge samples of primary sludge (PS) and secondary sludge (SS) were collected at Yorkshire Water's Esholt WWTW in Bradford, UK. All sludge samples were stored at 4 °C after collection and then used for the hydrothermal treatments prior characterisation. The analytical methods for the characterisation of the raw biomass followed the methodology described in Aragón-Briceño, Grasham, Ross, Dupont and Camargo-Valero [12].

2.2.2. Anaerobic treatments

Primary (PS), secondary (SS) and 1:1 mix of primary-secondary (MIX) sludge were processed by anaerobic treatment for 30 days in the lab, before further hydrothermal processing. Resulting samples were named as follows: digested primary sludge – ADPS; digested secondary sludge – ADSS; and digested mix of PS and SS – ADMIX.

AD tests were carried out following the method described by Aragón-Briceño, Ross and Camargo-Valero [3], [12] using sewage sludge samples and process waters separated from HTC experiments. The inoculum concentration used in each batch for the BMP tests was 1:1 volume inoculum to substrate ratio (≅10 g/L VS of inoculum and ≅ 2 g/L of COD of process waters). The batch experiments were performed in multiple series of 120-mL bottles sealed with a rubber stopper and aluminium cap at 37 °C for 21 days. All the AD tests were conducted in duplicate. Distilled water was used for diluting samples to help them to reach the set COD concentration and volume (60 ml for each reactor). Each bottle was purged with nitrogen to keep anaerobic conditions and avoid the presence of oxygen inside the bottle. Test bottles were kept undisturbed at all time, apart when mixing by hand during biogas production measurements. Methane production was monitored by using a volumetric method with a solution of 1 M NaOH. Volumetric methane production is corrected for temperature and pressure and reported at standard conditions (0 °C and 1 atm). For every measurement, a bottle was taken out and sacrificed for conducting lab analyses. All the analyses were carried out in duplicate.

2.2.3. Hydrothermal experiments

Thermal experiments were conducted in a non-stirred 500 mL stainless steel batch Parr reactor. In each batch experiment 220 mL of sludge sample (2.5% w/w) were loaded in the reactor and sealed. Hydrothermal treatments were performed at 160 °C for 30 min at 5 bar and at 250 °C for 30 min at 40 bar with heating rates of ≅ 5.3 and 4.2 °C/min respectively. After treatment, the reactor was cooled down to 25 °C. The hydrochars and process waters were processed and analysed according to the methodology described by Aragón-Briceño, Ross and Camargo-Valero [3], [12]. Solids samples (Hydrochar and sewage solid fraction) were dried for 7 days at 40 °C in an oven and weighted afterwards.

2.2.4. Solid and liquid samples characterization

The solid and liquid characterization was used to calculate the mass and energy balances of the different scenarios. Raw sludge and different hydrochar were analysed in a CHNS analyser (Elemental Analyser, CE Instruments Flash EA 1112 Series) to perform ultimate analyses of dry hydrochars (See **Table 1**).

The process waters (PW) were processed following standard methods for the characterisation of wastewater samples, for Chemical Oxygen Demand (COD), Total Solids (TS), Total Suspended Solids (TSS), Total Phosphorus, Total Kjeldahl Nitrogen (TKN) and Biomethane Potential (BMP) (See results in **Figures 2 and 3**).

Table 1.- Results from the CHNS analysis of the different sewage sludge and hydrochar in dry basis.

Sample	Ultimate analysis				
	C (%)	H (%)	N (%)	O^a (%)	S (%)
Primary Sludge	40.3	6.6	3.3	23.7	0.4
Secondary Sludge	33.5	5.5	4.1	28.5	0.2
Mix Sludge	35.5	5.8	3.3	28.0	0.1
AD Primary Sludge	31.1	5.1	3.0	24.0	0.5
AD Secondary Sludge	32.2	5.2	3.4	23.5	0.6
AD Mix Sludge	30.9	5.0	3.0	24.1	0.7

Hydrochars from 160 °C - 30 min- 5 Bar					
Primary Sludge	40.3	6.3	2.1	22.5	0.1
Secondary Sludge	33.1	5.3	3.3	21.7	0.7
Mix Sludge	37.5	6.0	2.6	21.6	0.5
AD Primary Sludge	28.9	4.5	2.1	19.8	0.5
AD Secondary Sludge	29.4	4.6	2.4	19.1	0.6
AD Mix Sludge	29.9	4.6	2.5	18.9	0.6
Hydrochars from 250 °C - 30 min- 40 Bar					
Primary Sludge	37.4	5.3	1.0	15.6	0.1
Secondary Sludge	36.1	4.6	1.9	11.6	0.5
Mix Sludge	35.8	5.2	1.1	15.2	0.3
AD Primary Sludge	27.3	3.8	1.1	11.4	0.3
AD Secondary Sludge	27.5	3.8	1.3	11.6	0.4
AD Mix Sludge	26.6	3.7	1.1	12.6	0.3

[a]Calculated as difference between sum of C,H,N,S,ash.

2.2.5. Hydrochar Yield

Hydrochar yield (*Y*) was determined as reported by Aragón-Briceño, Ross and Camargo-Valero [3], [12]:

$$Y\ (\%) = \frac{mass\ of\ dry\ hydrochar}{mass\ of\ dry\ Substrate\ feedstock} * 100 \qquad (1)$$

2.2.6. High Heating Value (HHV)

In order to know the theoretical calorific value of the hydrochar, the Dulong equation reported by Channiwala and Parikh [51] was used.

$$HHV\ (MJxKg^{-1}) = 0.336\ (\ \%Carbon) + 1.433\left(\%Hydrogen - \left(\frac{\%Oxygen}{8}\right)\right) + 0.0942\ (\%\ Sulphur) \qquad (6)$$

2.2.7. Biochemical Methane Potential (BMP)

In order to assess the performance of methane production by gram of chemical oxygen demand (COD) added, the following BMP formula was used:

$$BMP = \frac{V_{CH_4} - V_{CH_4,blank}}{(Mass\ of\ COD\ fed\ in\ biodigester)} \qquad (7)$$

Where:

BMP=Biochemical Methane Potential (ml of CH_4/ g of COD added)

V_{CH4} =Volume of methane produced in bottle (ml)

$V_{CH4, blank}$=Volume of methane produced in the blanks (ml)

Mass of COD=Mass of COD of the substrate (g of COD substrate)

2.2.8. Thermal treatment energy calculations

The energy required for the thermal treatments was based on the energy required calculations to heat water in a closed batch system as stated by Berge, Ro, Mao, Flora, Chappell and Bae [16]. Assuming that the heater has 100% resistance, there is no heating losses in the tank during the thermal treatment and the volume of the water remain constant, the energy required was determined by the followed equations:

$$M_{water\ total} = \rho_{sat.-liq} * V_{sat.-liq} + \rho_{sat.gas} * V_{sat.-gas} \quad (8)$$

$$V_{reactor} = V_{sat.-liq} + V_{sat.-gas} \quad (9)$$

$$H_T = [\rho_{sat.-liq} * V_{sat.-liq} * H_{sat.-Liq} + \rho_{sat.gas} * V_{sat.-gas} * H_{sat.-gas}] \quad (10)$$

Where:

$M_{water\ total}$ is the mass of the water input into the reactor [g].

$\rho_{(sat.-liq)}$ is the density of the saturated water at the thermal treatment temperature [g/L].

$\rho_{(sat.-gas)}$ is the density of the saturated water vapour at the thermal treatment temperature [g/L].

$V_{sat.-liq}$ is the Volume of the water in liquid fraction [L].

$V_{sat.-gas}$ is the Volume of the water in gas fraction [L].

$V_{reactor}$ is the volume of the reactor [L].

$H_{sat.-liq}$ is the enthalpy of the saturated water at the thermal treatment temperature [J/g].

$H_{sat.-gas}$ is the enthalpy of the saturated water vapour at the thermal treatment temperature [J/g].

H_T is the energy required to heat up the water [J],

However in order to get the results into kWh, the following relation was taken into account: 1 J = 2.777 $*10^{-7}$ kWh.

Table 2.- Process assumptions and calculation basis considered for the mass and energy balances of the different scenarios.

Description	Sc1	Sc2	Sc3	Sc4	Sc5	Sc6	References
Sludge to be treated (Kg)	1000	1000	1000	1000	1000	1000	Assumed
Feeding sludge	PS	SS	Mix	PS	SS	Mix	Considered
Solids concentration (%DS)	15	15	15	15	15	15	Assumed
Anaerobic Digestion temperature (°C)	37	37	37	37	37	37	Considered
Anaerobic Digestion retention time (Day)	21	21	21	21	21	21	Considered
Thermal treatment temperature (°C)	160	160	160	250	250	250	Considered
Thermal treatment retention time (H)	0.5	0.5	0.5	0.5	0.5	0.5	Considered
Recovery of heat energy from thermal treatment (%)	85	85	85	85	85	85	Shemfe, Gu and Ranganathan [53] and Sridhar Pilli, Song Yan, R. D. Tyagi and R. Y. Surampalli [54]
Raw sludge COD removal during AD(%)	38	48	44	38	48	44	Experimental Values
Process Water COD removal during AD(%)	47	42	37	59	59	60	Experimental Values
Methane production of raw sludge (m^3 of methane/Ton of COD)	129	116	226	129	116	226	Experimental Values
Methane production of Process Water (m^3 of methane/Ton of COD)	130	207	204	218	212	232	Experimental Values
Energy required for thermal treatment (MJ*Kg-1 of dry feedstock)	3.3	3.3	3.3	5.9	5.9	5.9	Experimental Values
Hydrochar yield (%)	50	55	56	37	40	40	Experimental Values
Solids separator Energy consumption (kW/Dry tonne) – Centrifuge	108	108	108	108	108	108	Smith and Liu [55]
Energy required for Mixing in the AD (kW/Dry tonne)	7	7	7	7	7	7	Oreggioni, Gowreesunker, Tassou, Bianchi, Reilly, Kirby, Toop and Theodorou [56]

3. Results and discussions

3.1. Mass balance

The experimental data obtained from lab experiments and overall considerations made (Table 2) were used for assessing mass and energy balances of the different thermal treatments integrated to anaerobic digestion and to assess the scenarios shown in **Figure 2** and **Figure 3.** Six scenarios were built, compared and assessed, considering three different sewage sludge streams (PS, SS and Mix) at two different thermal treatment temperatures (160 and 250 °C). In this study 1,000Kg as initial mass of sewage sludge was considered with 15% w/w of initial solid concentration. The initial solids concentration considered for the scenarios was based on the minimum solid concentration that shows positive energy balance.

Table 3 shows COD and Solids reduction efficiencies (%) of the proposed scenarios. The percentage of solids removed comes from the sum of solids volatilized during the thermal treatment and solids converted to biogas during the AD treatment. Results showed that higher thermal treatments trends to reduce more solids (See **Table 3**). During thermal treatment, the solids in sludge samples are hydrolysed leading to the increase on water soluble products due the solubilisation of organic and inorganic compounds into the liquid phase [3, 36, 57-59]. Scenarios with the 160 °C hydrothermal treatment integrated showed a solids removal between 47 and 56% and the scenarios with the 250 °C hydrothermal treatment integrated reported 62 to 68% solid removal. Scenarios built with PS and Mix sludge did not show significant differences in terms of solids reduction at the same HTP temperature. However, scenarios with the secondary sludge, regardless the temperature treatment, presented the lowest percentage of solids removal with 47% and 62% at 160 °C and 250 °C thermal treatment, respectively. This might be due the biomass from the SS comes from a biological treatment (aerobic) that makes the organic compounds less biodegradable for the 1st AD stage and affecting the overall performance of the scenarios.

The overall COD removal reported comes from the sum of COD volatilized during the thermal treatment and COD converted to biogas during the AD treatment. During the thermal treatment, most of the COD volatilized is normally converted into CO_2 [57]. The COD removal results showed a similar trend as the solids removal where higher temperatures presented higher COD removals. Scenarios built with Mix sludge presented the highest overall COD removal for both thermal treatments with 58% at 160°C and 66% at 250°C. This is followed by the scenarios built with PS and SS where the COD removals were 56 and 46% at 160 °C and 61 and 51% at 250 °C, respectively. In addition, if it is considered that after the 2nd AD stage the COD concentration corresponded to 4-8% of the total initial concentration for all the scenarios (See **Figures 2 and 3**), that means that most of the COD has the potential to be degraded through energy generation either from biogas or hydrochar combustion. The remaining liquid waste also can be used for irrigation due its high amount of nutrients (see **Table 4**) or reused into the thermal treatment if more liquid is required for co-processing the sewage sludge with other biomass.

Table 3 also shows methane and hydrochar production of the different scenarios. The methane production did not show a clear correlation with the overall COD and solids removed. Scenarios 1 and 4 that used PS as initial feedstock and thermal treatments temperatures of 160 and 250 °C, showed the highest overall methane production with 57 and 67.5 m^3/tonne of sludge, respectively. This might be due the high organic content in the feedstock that has no receive any previous treatment. On the other hand, the SS sample presented the lowest methane production for both thermal treatments at 160 and 250 °C with 20.3 and 22.7 m^3/tonne of sludge, respectively. This might be due the previous biological treatment received which makes the remaining COD less suitable for methane conversion.

Hydrochars produced during hydrothermal treatments varied in all tested scenarios because they heavily depend on feedstock characteristics and process conditions [3, 18, 36]. Scenarios with 160 °C treatment temperature (1 to 3) presented higher hydrochar production than 250 °C treatment temperature scenarios (4 to 6). Reaction temperature condition played a key role in the total hydrochar production for the scenarios. This is because as the reaction temperature increases, the hydrochar yield hydrochar decreases [3, 12]. Moreover, scenarios that used PS as an initial feedstock (1 and 4), showed the lowest hydrochar production with 75.6 and 54.8 Kg/tonne of sludge. Scenarios built with Mix and SS, presented similar hydrochar production at the same treatment temperature – i.e., scenarios 2 and 3 (HTT 160 °C) showed a hydrochar production of 82.5 and 83.6 Kg/tonne respectively and scenarios 5 and 6 (HTT 250 °C) a production of 59.7 and 60.6 Kg/tonne of sludge respectively.

Table 3.- Results from mass balances in each proposed scenario.

	Total solids reduction, %	Total COD removal, %	Total methane production (m^3/tonne of sludge)	Total Hydrochar Production (Kg/tonne of sludge)
Scenario 1	56%	56%	57.0	75.6
Scenario 2	47%	46%	20.3	82.5
Scenario 3	56%	58%	34.0	83.6
Scenario 4	68%	61%	67.5	54.8
Scenario 5	62%	51%	22.7	59.7
Scenario 6	68%	66%	37.5	60.6

Scenarios built with PS (1 and 4) showed the highest biogas and hydrochar production and can be considered as suitable alternatives for WWTWs without a secondary treatment. On the other hand, scenarios 2 and 5 are not convenient due their low overall biogas and hydrochar production and quality. However, considering the potential costs due the necessary modifications to integrate HTC-AD at a current WWTWs, scenarios that involve MIX sludge (3 and 6) are more suitable compared with the rest of the scenarios. This is due less modifications would be needed to adapt the HTC at the WWTWs, compared with the rest of the scenarios that would need separated treatments for PS and SS.

One of the most promising process for simultaneous nitrogen and phosphorus recovery in the waste water sector is through formation and precipitation of ammonium magnesium phosphate ($NH_4MgPO_4{\bullet}6(H_2O)$), also known as struvite [60]. Some authors have demonstrated that during hydrothermal treatment, there nitrogen and phosphorus solubilisation from sewage digestate and most of the nitrogen extracted is present within the liquid fraction [3, 36, 61]. **Figure 2 and 3** presents nitrogen and phosphorus mass balances and **table 4** the nitrogen and phosphorus available for the potential struvite production per ton of sludge in each tested scenario. Scenarios 2 and 5 presented the highest nitrogen concentration extracted; this is due to the fact that SS samples mainly contain biomass from the aerobic treatment which take up nitrogen and phosphorus from waste water. Most of the nitrogen solubilized during thermal treatment was in ammonium form (NH_4^+), which has the potential to be used for struvite precipitation. Previous studies have proven that during thermal treatment the proteins present in sewage sludge hydrolyse forming ammonium which is released into the process waters [3].

On the other hand, most of the phosphorus remained within the solid fraction. The phosphorus solubilisation is carried on during the thermal process due the organic phosphorus compounds (complex phospholipids, DNA and phosphates monoesters) break down into phosphate [36, 61]. Although, the fate of P is highly feedstock dependent during hydrothermal treatment and is linked to the levels of metals presented in the feedstock [3, 36]. The phosphorus extraction ranged from 0.03 to 0.07 Kg per tonne of sludge in those scenarios (1 to 3) with 160 °C treatment and 0.8 Kg per tonne of sludge in those scenarios (4 to 6) with 250 °C treatment. This suggest that 250°C treatment favoured the phosphorus fixation within the hydrochar unlike the 160°C that favoured the phosphorus solubilisation. The majority of phosphorus extracted was inorganic phosphorus (PO_4^{-3}) ranging from 50 to 75%. However, the amount of soluble phosphorus determined was very low which might be due to the low initial P content in sludge samples as biological P removal is not part of the processes at Esholt's WWTW, where sludge samples were collected. Despite of that, **Table 4** shows the potential struvite production in each scenario. Scenarios 4 and 6 showed the highest potential struvite production with 0.06 and 0.05 kg per tonne of sludge treated respectively.

Table 4.- Nitrogen and Phosphorus available for potential struvite production.

	*$Nitrogen_{Sol}$ (Kg)	*$Nitrogen_{Liq}$ (Kg)	*Ammonia (Kg)	*Total $Phosphorus_{Sol}$ (Kg)	*Total $Phosphorus_{Liq}$ (Kg)	*PO_4^{-3} in the liquid (Kg)	*[1]Mg addition (Kg)	*Struvite Production (Kg)
Scenario 1	1.8	3.8	3.1	1.0	0.03	0.02	0.00[2]	0.02
Scenario 2	2.6	5.8	4.6	0.8	0.07	0.04	0.01	0.04
Scenario 3	2.3	4.8	3.8	0.8	0.04	0.02	0.00[2]	0.02
Scenario 4	1.3	4.6	3.7	0.7	0.08	0.06	0.01	0.06
Scenario 5	1.9	6.8	4.7	0.6	0.08	0.04	0.01	0.04
Scenario 6	1.6	5.7	4.7	0.6	0.08	0.05	0.01	0.05

*per tonne of sludge
[1]Mg needed to be added
[2]Value below the decimal position.

3.2. Energy balance

The summary of the energy balance from the different scenarios is presented in **table 5**. The energy production from methane favoured those scenarios with hydrothermal treatments at the highest reaction temperature. The PS sludge had the highest net energy production in all the scenarios followed by the Mix Sludge and the SS sludge with the lowest. Scenarios 1 and 4 (PS) samples had a potential energy production from methane of 221.1 and 261.6 kW per tonne of sludge respectively. This was followed by scenarios 3 and 6 (MIX) and scenarios 1 and 5 (SS) with a potential energy production from methane of 131.7, 145.5, 78.8 and 88 kW per tonne of sludge respectively.

On the other hand, the net energy balance favoured the scenarios with HTC processes at the lowest reaction temperature, when energy was produced only from biogas. All the scenarios presented a positive energy balance; scenarios 1 and 4 showed the highest net energy balance, producing extra 163.3 and 187.8 kW per tonne of sludge treated, followed by scenarios 3, 6, 2 and 5 with 73.9, 71.8, 21 and 14.2 kW extra tonne of sludge respectively. This potential extra energy produced can be used either for the total WWTW energy needs or for electricity exportation.

When only biogas production is considered as the only energy source in the proposed systems, the net energy production reduces significantly in comparison when the hydrochar is included as an energetic. For that reason it is important the hydrochar inclusion as a fuel source within the system in order to make a more self-sustainable. Despite of the quality of hydrochar samples, the net energy production in each scenario is enhanced with the inclusion of the energy that comes from hydrochars if used as a low-grade fuel. According to Aragón-Briceño, Ross and Camargo-Valero [3], between 56 to 59% of the energy produced in an AD + HTC process comes from the hydrochar. Scenarios with lower temperature treatment (1, 2 and 3) have more potential for energy production from the hydrochar. The hydrochar yield is higher compared at lower treatment temperatures compared with those scenarios with a higher treatment temperature (4, 5 and 6). The hydrochar fraction represents between 40 to 68% for scenarios with 160°C treatment and between 24 to 51% for the scenarios with 250 °C treatment of the overall potential energy production. Therefore, the inclusion of the hydrochar as an energy source is directly reflected on the net energy balance which would be enhanced between 89 to 642% for scenarios with 160 °C treatment and between 42 to 435% for the scenarios with 250 °C treatment. Other studies have reported that the implementation of the thermal treatment at the end of the process favours the overall energy production up to 179% in comparison with the traditional AD [3]. Wang, Chang and Li [37] reported a positive overall energy recovery from the HTC of sewage sludge coupled with AD (668 MJ per tonne treated), where hydrochar contributed to 59% of the potential energy recovered. Medina-Martos, Istrate, Villamil, Gálvez-Martos, Dufour and Mohedano [9] reported in their study a contribution of 77% (562 MJ) from hydrochar to the total potential energy production of a HTC+AD system.

Table 5.- Results from energy balance per tonne of sludge processed in each scenario

	*Energy consumed (kWh)	*Energy produced from Methane (kWh)	*Net Energy balance (kWh)	*Potential Energy from the hydrochar (kWh)	*Net Energy balance including hydrochar (kWh)
Scenario 1	57.8	221.1	163.3	149.6	312.9
Scenario 2	57.8	78.8	21.0	169.6	190.5
Scenario 3	57.8	131.7	73.9	176.4	250.2
Scenario 4	73.8	261.6	187.8	82.4	270.2
Scenario 5	73.8	88.0	14.2	90.5	104.7
Scenario 6	73.8	145.5	71.8	89.7	161.4

*per tonne of sludge

3.3. Economics

In the United Kingdom the tariff rates for electricity exportation from renewable sources is established by the Office of Gas and Electricity Markets [62]. This is a non-ministerial government department and an independent National Regulatory Authority, recognised by EU Directives and governed by the Gas and Electricity Markets Authority (GEMA). The tariff rates for anaerobic digestion depends on the plant capacity for electricity generation. In this study, the lowest tariff of 5.30 p/kWh (Euro) was considered as the based tariff for the electricity produced by the methane produced [62]. The same tariff was considered for potential electricity production from the hydrochar since there is not a clear information about it.

In **Table 6,** the summary of economic benefits of integrating HTC with AD are shown. The potential profit from methane production ranged from €1.12 to €10.32 per tonne of sewage sludge treated. The economic analysis for the scenarios using PS (1 and 4) had the highest potential economic benefits with €9.02 per tonne of sludge at 160 °C and €10.32 per tonne of sludge at 250 °C showing an increase when as the reaction temperature increases. Scenarios with MIX sludge (3 and 6) presented an economic benefit of €4.28 and €4.17 per tonne of sludge, respectively and SS scenarios (2 and 4) showed the lowest economic benefit with €1.12 and €1.28 per tonne of sludge respectively.

The potential economic benefit from the hydrochar production is slightly higher compared with the biogas production. Some studies have found that the hydrochar production can represents the 55% of the total revenue [9]. Hydrochar yield played a key role on the economic benefit showing higher benefits for those scenarios with 160 °C thermal treatment. For scenarios with 250 °C thermal treatment, the economic benefits had not significative difference regardless the type of sewage used as feedstock. The benefit ranged from €4.36-€4.79 per tonne of sludge treated. The economic benefits from the hydrochar production might be increased if co-processing sewage sludge with other biomass feedstocks is considered in order to increase the quality properties of the hydrochar [63].

For this study, the price for the struvite was considered based on that average price for the struvite in the fertiliser market is €475.5 per tonne [60]. The economic benefit per tonne of sludge ranged from €9.10 to €18.08 for scenarios with 160 °C thermal treatment and between €20.19 to €26.24 for scenarios when the 250 °C thermal treatment was considered. Nonetheless, despite of the cost of phosphorus recovering as struvite can range from €2 up to €8 per kg of P recovered, the struvite production from the process waters still showed to be a good opportunity area for increase the overall profit, especially if feedstocks with high phosphorus content are co-processed with the sewage sludge [60, 64].

In 2010, the average sewage sludge generation per person in the UK was 22.5 kg of dry sewage sludge annually [1, 65]. Considering a large size WWTW could serve a population of about 100,000 p.e. – i.e., an annual production of 15,000 tonnes of sewage sludge (15% of dry solids) – the economic benefits per year depending on the tested scenario would range from €16,832 to €1514,788 from biogas production; €65,429 to €140,102 from hydrochar production; and €136,449 to €393,652 from struvite production (without considering the price of Mg addition and pH regulation process). The scenarios where the 250 °C treatment was applied showed the best potential for increasing profits with €613,869 and €486,528 for scenarios 4 and 6, respectively (See **Table 6**).

Table 6.- Potential economic benefits of integrating HTC with AD.

	Electricity production profit from methane per tonne of sewage sludge (€)	Electricity production profit from Hydrochar per tonne of sewage sludge (€)	[a]Struvite production profit per tonne of sewage sludge (€)	Total profit per tonne sludge (€)	Total profit for WWTW serving a 100,000 p.e. (€)
Scenario 1	9.02	7.92	9.21	26.15	392,306
Scenario 2	1.48	8.98	18.08	28.54	428,094
Scenario 3	4.28	9.34	9.10	22.72	340,798
Scenario 4	10.32	4.36	26.24	40.92	613,869
Scenario 5	1.12	4.79	20.19	26.10	391,518
Scenario 6	4.17	4.75	23.52	32.44	486,528

[e]xchange rate £1=€1.1197 [66].

Therefore, taking into account all the variables involved in this analysis (waste reduction, energy production and economic benefits), scenarios 4 and 6 showed to be the more suitable for the integration of HTC with AD in a WWTW. However, more variables and many other aspects have to be considered for a more complete analysis as the cost of the WWTW´s modifications needed, cost of an struvite production process plant, cost of an energy generator from solid biomass fuel, etc.

4. Conclusions

A complete HTC+AD system for integration in a WWTW was proposed. Six different scenarios were evaluated built with 3 different sewage sludge (PS, SS and MIX) and 2 different thermal treatments (160 and 250 °C). It was found that the integration of HTC-AD, through the proposed scenarios, presented environmental benefits (waste reduction) such as an overall total solid and COD reduction up to 68 and 66% respectively. In addition, the integration of the HTC-AD process showed a positive energy balance in all the proposed scenarios with a maximum net energy production of 312.9 kWh per tonne of treated sludge if the hydrochar is considered as a fuel source.

The economic analysis for the proposed scenarios showed a potential benefit up to €613,869 as long as the struvite production is considered as a part of the whole system. Scenarios that included PS and MIX sludge with 250 °C thermal treatment showed to be the more suitable options for HTC+AD integration if organic matter removal, energy harnessing and economic feasibility are considered as main variables. Nonetheless, still many aspects have to be considered regarding capital costs for retrofitting existing WWTWs and the addition of complementary equipment for the use of hydrochar as an energy source. In addition, HTC co-processing of other feedstocks with AD sludge needs to be considered as a complementary option as it might enhance the quality of AD products (process waters and hydrochar) and therefore the overall economic and environmental benefits.

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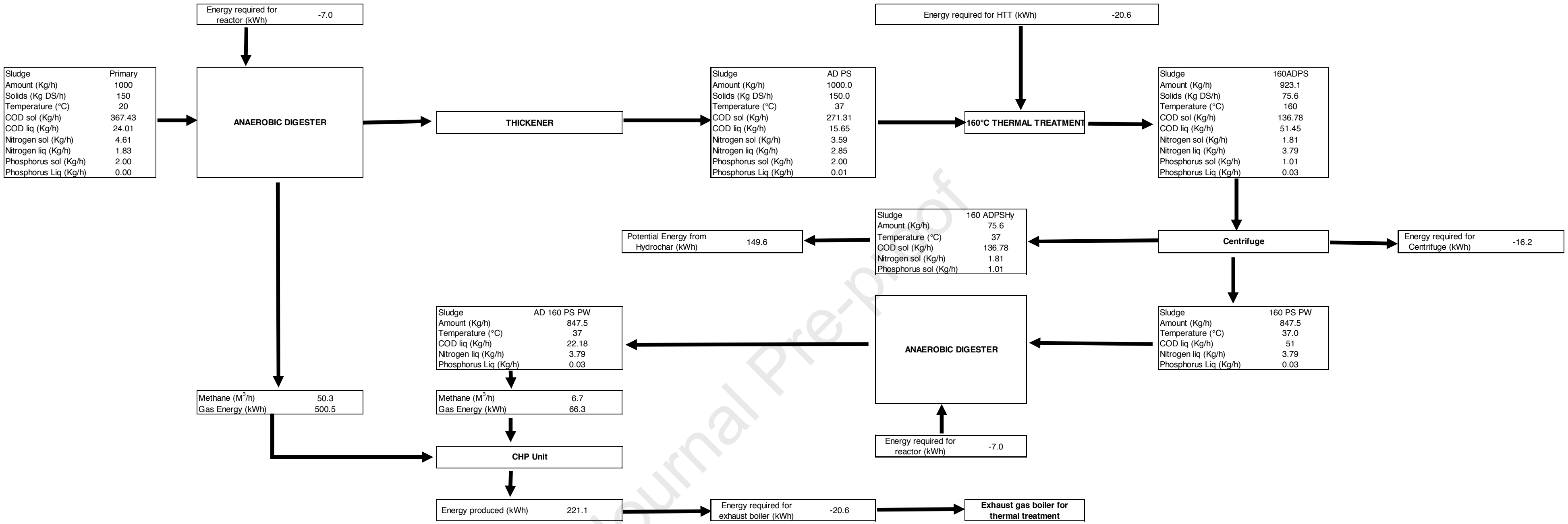
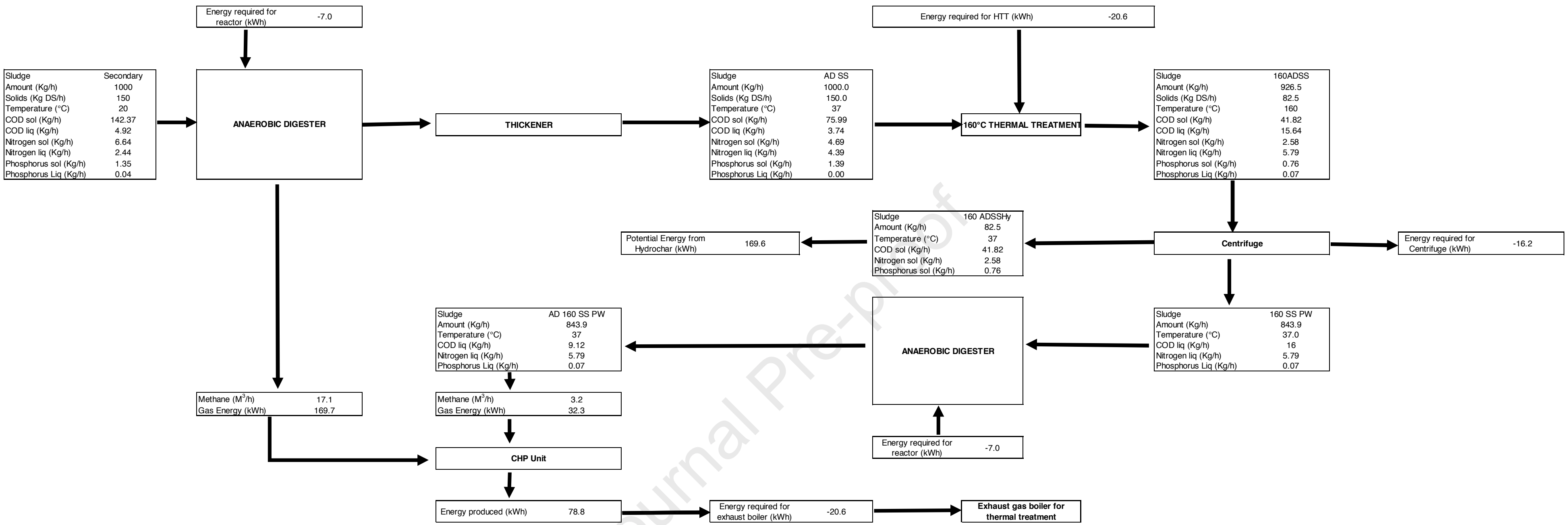
a) Scenario 1


Energy required for reactor (kWh) -7.0
Sludge Primary
Amount (Kg/h) 1000
Solids (Kg DS/h) 150
Temperature (°C) 20
COD sol (Kg/h) 367.43
COD liq (Kg/h) 24.01
Nitrogen sol (Kg/h) 4.61
Nitrogen liq (Kg/h) 1.83
Phosphorus sol (Kg/h) 2.00
Phosphorus Liq (Kg/h) 0.00
ANAEROBIC DIGESTER
THICKENER
Sludge AD PS
Amount (Kg/h) 1000.0
Solids (Kg DS/h) 150.0
Temperature (°C) 37
COD sol (Kg/h) 271.31
COD liq (Kg/h) 15.65
Nitrogen sol (Kg/h) 3.59
Nitrogen liq (Kg/h) 2.85
Phosphorus sol (Kg/h) 2.00
Phosphorus Liq (Kg/h) 0.01
Energy required for HTT (kWh) -20.6
160°C THERMAL TREATMENT
Sludge 160ADPS
Amount (Kg/h) 923.1
Solids (Kg DS/h) 75.6
Temperature (°C) 160
COD sol (Kg/h) 136.78
COD liq (Kg/h) 51.45
Nitrogen sol (Kg/h) 1.81
Nitrogen liq (Kg/h) 3.79
Phosphorus sol (Kg/h) 1.01
Phosphorus Liq (Kg/h) 0.03
Centrifuge
Energy required for Centrifuge (kWh) -16.2
Sludge 160 ADPSHy
Amount (Kg/h) 75.6
Temperature (°C) 37
COD sol (Kg/h) 136.78
Nitrogen sol (Kg/h) 1.81
Phosphorus sol (Kg/h) 1.01
Potential Energy from Hydrochar (kWh) 149.6
Sludge 160 PS PW
Amount (Kg/h) 847.5
Temperature (°C) 37.0
COD liq (Kg/h) 51
Nitrogen liq (Kg/h) 3.79
Phosphorus Liq (Kg/h) 0.03
ANAEROBIC DIGESTER
Energy required for reactor (kWh) -7.0
Sludge AD 160 PS PW
Amount (Kg/h) 847.5
Temperature (°C) 37
COD liq (Kg/h) 22.18
Nitrogen liq (Kg/h) 3.79
Phosphorus Liq (Kg/h) 0.03
Methane (M3/h) 50.3
Gas Energy (kWh) 500.5
Methane (M3/h) 6.7
Gas Energy (kWh) 66.3
CHP Unit
Energy produced (kWh) 221.1
Energy required for exhaust boiler (kWh) -20.6
Exhaust gas boiler for thermal treatment

b) Scenario 2


Energy required for reactor (kWh) -7.0
Sludge Secondary
Amount (Kg/h) 1000
Solids (Kg DS/h) 150
Temperature (°C) 20
COD sol (Kg/h) 142.37
COD liq (Kg/h) 4.92
Nitrogen sol (Kg/h) 6.64
Nitrogen liq (Kg/h) 2.44
Phosphorus sol (Kg/h) 1.35
Phosphorus Liq (Kg/h) 0.04
ANAEROBIC DIGESTER
THICKENER
Sludge AD SS
Amount (Kg/h) 1000.0
Solids (Kg DS/h) 150.0
Temperature (°C) 37
COD sol (Kg/h) 75.99
COD liq (Kg/h) 3.74
Nitrogen sol (Kg/h) 4.69
Nitrogen liq (Kg/h) 4.39
Phosphorus sol (Kg/h) 1.39
Phosphorus Liq (Kg/h) 0.00
Energy required for HTT (kWh) -20.6
160°C THERMAL TREATMENT
Sludge 160ADSS
Amount (Kg/h) 926.5
Solids (Kg DS/h) 82.5
Temperature (°C) 160
COD sol (Kg/h) 41.82
COD liq (Kg/h) 15.64
Nitrogen sol (Kg/h) 2.58
Nitrogen liq (Kg/h) 5.79
Phosphorus sol (Kg/h) 0.76
Phosphorus Liq (Kg/h) 0.07
Centrifuge
Energy required for Centrifuge (kWh) -16.2
Sludge 160 ADSSHy
Amount (Kg/h) 82.5
Temperature (°C) 37
COD sol (Kg/h) 41.82
Nitrogen sol (Kg/h) 2.58
Phosphorus sol (Kg/h) 0.76
Potential Energy from Hydrochar (kWh) 169.6
Sludge 160 SS PW
Amount (Kg/h) 843.9
Temperature (°C) 37.0
COD liq (Kg/h) 16
Nitrogen liq (Kg/h) 5.79
Phosphorus Liq (Kg/h) 0.07
ANAEROBIC DIGESTER
Energy required for reactor (kWh) -7.0
Sludge AD 160 SS PW
Amount (Kg/h) 843.9
Temperature (°C) 37
COD liq (Kg/h) 9.12
Nitrogen liq (Kg/h) 5.79
Phosphorus Liq (Kg/h) 0.07
Methane (M3/h) 17.1
Gas Energy (kWh) 169.7
Methane (M3/h) 3.2
Gas Energy (kWh) 32.3
CHP Unit
Energy produced (kWh) 78.8
Energy required for exhaust boiler (kWh) -20.6
Exhaust gas boiler for thermal treatment

c) Scenario 3

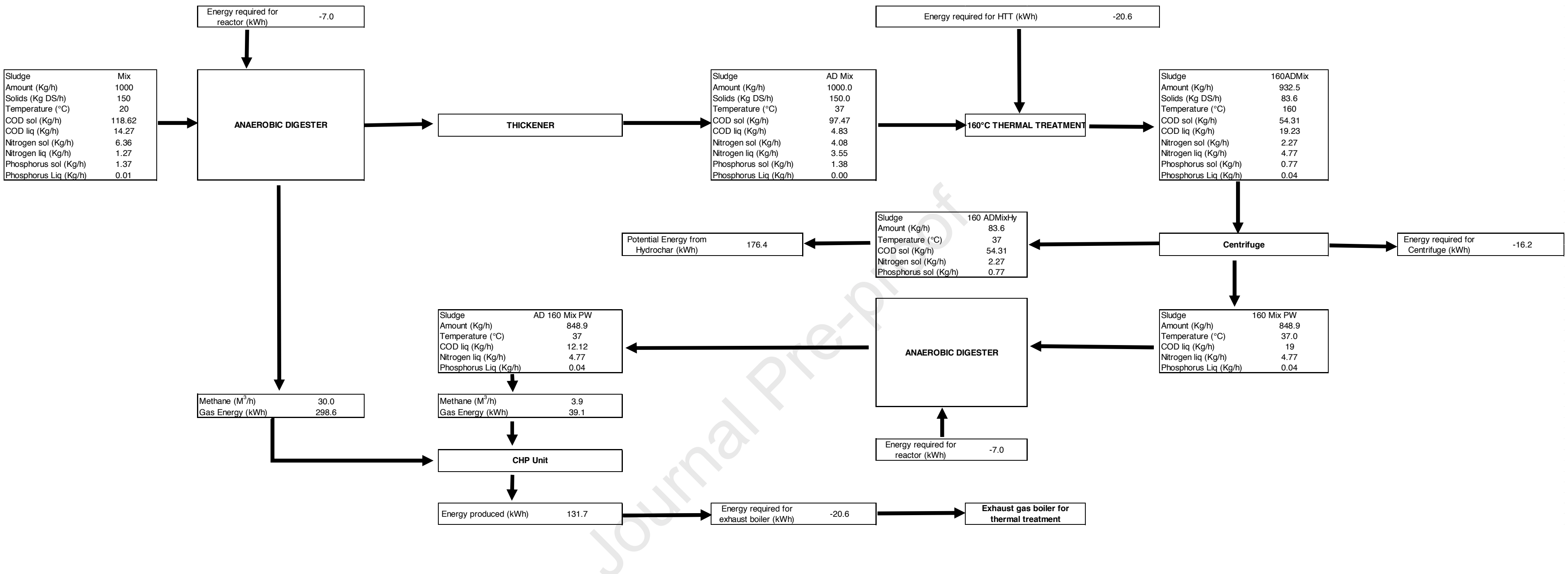
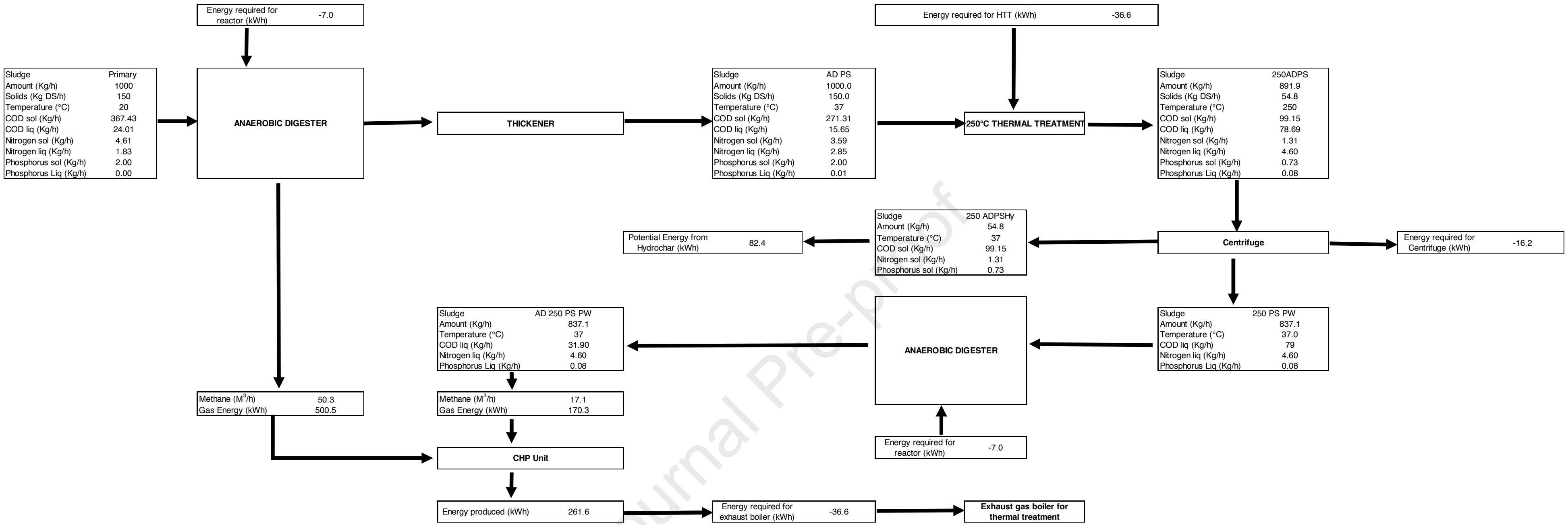
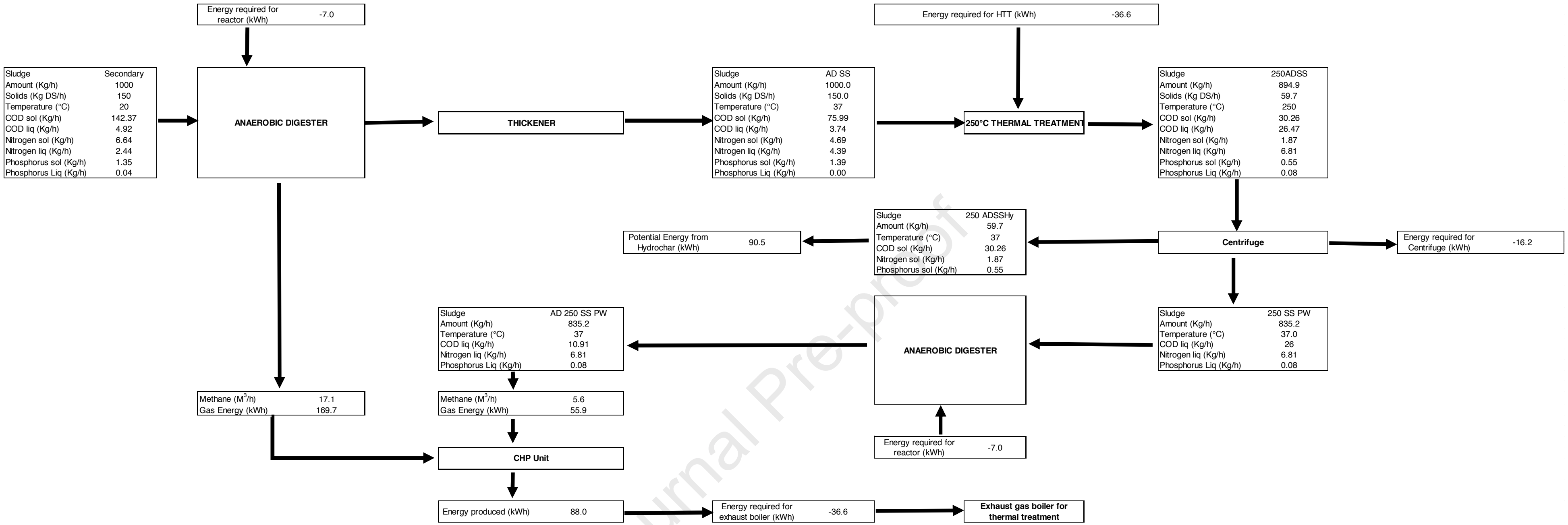


Figure 2.- Mass and energy balance scenarios of the a) Primary Sludge, b) Secondary Sludge and c) Mix Sludge at 160°C thermal treatment.

a) Scenario 4


Energy required for reactor (kWh) -7.0
Energy required for HTT (kWh) -36.6
Sludge Primary
Amount (Kg/h) 1000
Solids (Kg DS/h) 150
Temperature (°C) 20
COD sol (Kg/h) 367.43
COD liq (Kg/h) 24.01
Nitrogen sol (Kg/h) 4.61
Nitrogen liq (Kg/h) 1.83
Phosphorus sol (Kg/h) 2.00
Phosphorus Liq (Kg/h) 0.00
ANAEROBIC DIGESTER
THICKENER
Sludge AD PS
Amount (Kg/h) 1000.0
Solids (Kg DS/h) 150.0
Temperature (°C) 37
COD sol (Kg/h) 271.31
COD liq (Kg/h) 15.65
Nitrogen sol (Kg/h) 3.59
Nitrogen liq (Kg/h) 2.85
Phosphorus sol (Kg/h) 2.00
Phosphorus Liq (Kg/h) 0.01
250°C THERMAL TREATMENT
Sludge 250ADPS
Amount (Kg/h) 891.9
Solids (Kg DS/h) 54.8
Temperature (°C) 250
COD sol (Kg/h) 99.15
COD liq (Kg/h) 78.69
Nitrogen sol (Kg/h) 1.31
Nitrogen liq (Kg/h) 4.60
Phosphorus sol (Kg/h) 0.73
Phosphorus Liq (Kg/h) 0.08
Sludge 250 ADPSHy
Amount (Kg/h) 54.8
Temperature (°C) 37
COD sol (Kg/h) 99.15
Nitrogen sol (Kg/h) 1.31
Phosphorus sol (Kg/h) 0.73
Potential Energy from Hydrochar (kWh) 82.4
Centrifuge
Energy required for Centrifuge (kWh) -16.2
Sludge AD 250 PS PW
Amount (Kg/h) 837.1
Temperature (°C) 37
COD liq (Kg/h) 31.90
Nitrogen liq (Kg/h) 4.60
Phosphorus Liq (Kg/h) 0.08
ANAEROBIC DIGESTER
Sludge 250 PS PW
Amount (Kg/h) 837.1
Temperature (°C) 37.0
COD liq (Kg/h) 79
Nitrogen liq (Kg/h) 4.60
Phosphorus Liq (Kg/h) 0.08
Methane (M3/h) 50.3
Gas Energy (kWh) 500.5
Methane (M3/h) 17.1
Gas Energy (kWh) 170.3
Energy required for reactor (kWh) -7.0
CHP Unit
Energy produced (kWh) 261.6
Energy required for exhaust boiler (kWh) -36.6
Exhaust gas boiler for thermal treatment

b) Scenario 5


Energy required for reactor (kWh) -7.0
Energy required for HTT (kWh) -36.6
Sludge Secondary
Amount (Kg/h) 1000
Solids (Kg DS/h) 150
Temperature (°C) 20
COD sol (Kg/h) 142.37
COD liq (Kg/h) 4.92
Nitrogen sol (Kg/h) 6.64
Nitrogen liq (Kg/h) 2.44
Phosphorus sol (Kg/h) 1.35
Phosphorus Liq (Kg/h) 0.04
ANAEROBIC DIGESTER
THICKENER
Sludge AD SS
Amount (Kg/h) 1000.0
Solids (Kg DS/h) 150.0
Temperature (°C) 37
COD sol (Kg/h) 75.99
COD liq (Kg/h) 3.74
Nitrogen sol (Kg/h) 4.69
Nitrogen liq (Kg/h) 4.39
Phosphorus sol (Kg/h) 1.39
Phosphorus Liq (Kg/h) 0.00
250°C THERMAL TREATMENT
Sludge 250ADSS
Amount (Kg/h) 894.9
Solids (Kg DS/h) 59.7
Temperature (°C) 250
COD sol (Kg/h) 30.26
COD liq (Kg/h) 26.47
Nitrogen sol (Kg/h) 1.87
Nitrogen liq (Kg/h) 6.81
Phosphorus sol (Kg/h) 0.55
Phosphorus Liq (Kg/h) 0.08
Sludge 250 ADSSHy
Amount (Kg/h) 59.7
Temperature (°C) 37
COD sol (Kg/h) 30.26
Nitrogen sol (Kg/h) 1.87
Phosphorus sol (Kg/h) 0.55
Potential Energy from Hydrochar (kWh) 90.5
Centrifuge
Energy required for Centrifuge (kWh) -16.2
Sludge AD 250 SS PW
Amount (Kg/h) 835.2
Temperature (°C) 37
COD liq (Kg/h) 10.91
Nitrogen liq (Kg/h) 6.81
Phosphorus Liq (Kg/h) 0.08
ANAEROBIC DIGESTER
Sludge 250 SS PW
Amount (Kg/h) 835.2
Temperature (°C) 37.0
COD liq (Kg/h) 26
Nitrogen liq (Kg/h) 6.81
Phosphorus Liq (Kg/h) 0.08
Methane (M³/h) 17.1
Gas Energy (kWh) 169.7
Methane (M³/h) 5.6
Gas Energy (kWh) 55.9
CHP Unit
Energy required for reactor (kWh) -7.0
Energy produced (kWh) 88.0
Energy required for exhaust boiler (kWh) -36.6
Exhaust gas boiler for thermal treatment

c) Scenario 6

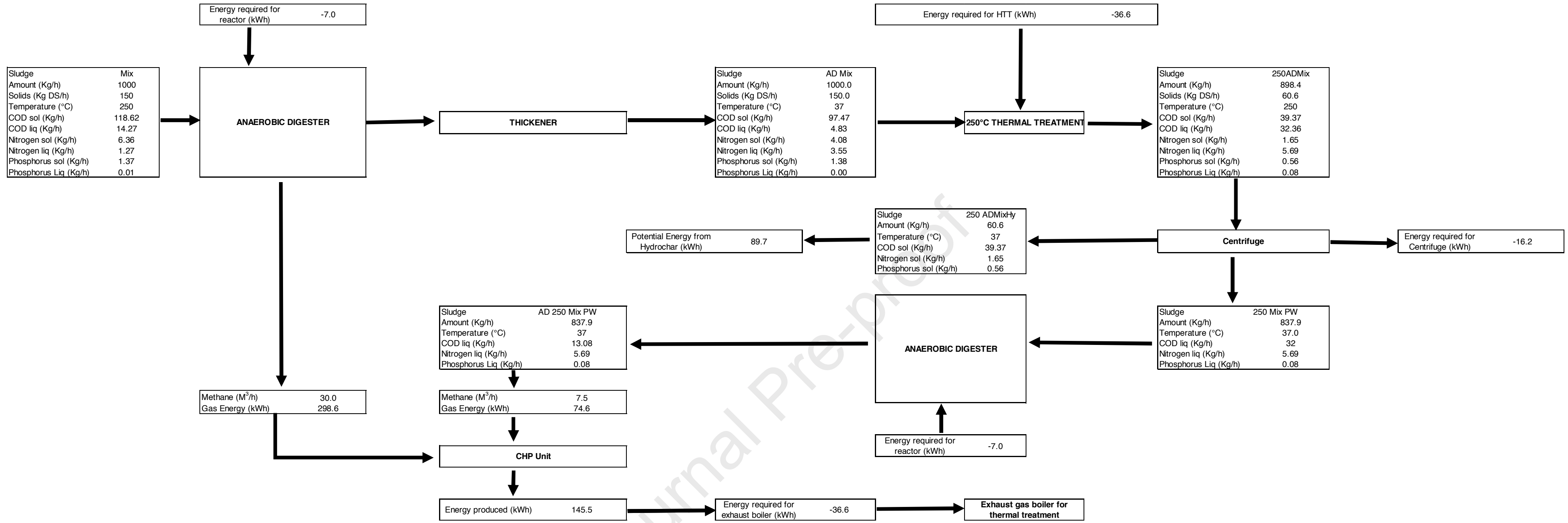


Figure 3.- Mass and energy balance scenarios of the a) Primary Sludge, b) Secondary Sludge and c) Mix Sludge at 250°C thermal treatment

- Mass and Energy balance of the HTC-AD integration are presented.
- Six HTC-AD scenarios with different process temperature and sewage sludge were evaluated.
- Scenarios assessed with 250 °C process temperature showed higher energy and economic benefits.
- Scenarios with MIX sludge stream showed to be the most suitable option for HTC-AD integration.