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## Fast pyrolysis of torrefied sewage sludge in a fluidized bed reactor



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#### HIGHLIGHTS

- Pyrolysis of dry and torrefied sewage sludge in a fluidized bed reactor was studied.
- The influence of torrefaction conditions on the pyrolysis products was determined.
- The cumulative product yields are not affected by torrefaction.
- Torrefaction step does not improve the liquid homogeneity.
- Torrefaction step does not improve the energy recovery of the organic phases.

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#### ABSTRACT

The pyrolysis of dry and of torrefied sewage sludge in a lab-scale fluidized bed reactor has been studied in order to determine whether torrefaction pre-treatment could enhance the properties of the liquid product obtained after pyrolysis. The aim of this work is to evaluate the influence of the torrefaction temperature (220–320 °C) and average solid residence time (3.6–10.2 min) on the product distribution and the properties of the pyrolysis products. Pyrolysis was conducted at 530 °C with an average solid residence time of 5.7 min and a nitrogen volumetric flow per reactor area of 0.074 m³ (STP) m $^{-2}$  s $^{-1}$  (measured at 0 °C and  $1.01\cdot10^5$  Pa). The experimental results show that torrefaction pre-treatment affects the pyrolysis liquid product, although it does not improve the homogeneity of the liquid. Specifically, it reduces the yields of water and the liquid aqueous phase obtained in the pyrolysis step, especially after torrefaction under the most severe conditions, but it does not have a great effect on the properties of the liquid organic phases obtained. The cumulative yields of gas and organic compounds from the two-step process are not different from the yields obtained from one-step pyrolysis.

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#### 1. Introduction

As the reserves of fossil fuels are running out and energy consumption is increasing, biomass and wastes are seen as potential sources of energy because they can be converted into fuels and chemicals by means of thermochemical and biochemical processes [1–3], being transformed these raw materials into renewable sources of C and H. Fast pyrolysis of biomass for liquid production is among the most promising of these technologies due to the high yield of bio-oil [4]. Sewage sludge is one of the materials that can be used as feedstock [5]. Pyrolysis is a management technology for sewage sludge that, in addition to reducing its volume, could contribute to obtaining renewable energy and valuable chemicals [5–7].

Sewage sludge pyrolysis liquid is a dark-brown complex mixture of organic compounds and water generated from depolymerization and dehydration reactions [8]. This liquid is not homogeneous and it separates into two or three phases [9-12]. Bio-oil from sewage sludge is rich in water (20-70 mass fraction %), resulting from the moisture in the initial material and from dehydration reactions during pyrolysis [5,13,14]. This high water content lowers the higher heating value and flame temperature of the liquid and causes phase separation, but benefits the viscosity and therefore fluidity [5,8,15]. The oxygen content is higher than in the case of crude oil and its distribution in the different compounds depends on the pyrolysis process conditions and on the raw material. This high oxygen content lowers the energy density of the bio-oil and makes it immiscible with hydrocarbon fuels [8]. Besides, the high oxygen content gives bio-oil a high reactivity during storage, which can cause increased viscosity and phase separation [16].

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In order to use bio-oil in turbines or engines as an alternative fuel, it needs to be upgraded due to the drawbacks mentioned above. There are various techniques to improve the fuel properties of bio-oil, either during its production using methods such as in situ catalytic cracking of pyrolysis vapours [17-23], or after it is obtained, by means of hydrodeoxygenation (HDO) [24,25] or decarboxylation (DCO) [26]. Integrated catalytic pyrolysis - in a combined pyrolysis-catalysis reaction system - is less flexible in terms of operational conditions since it operates at a single operation temperature and the catalyst has to be robust enough to resist both the temperature and the environment [27]. Besides, some authors have observed that, when using zeolites, the yield of gaseous products and of water increased and therefore the yield of solid product - char - and of organic compounds decreased [28,29]. The improvement of pyrolysis oil by means of its cracking over a catalyst such as zeolite is now under study. There remain some issues to be solved, such as the decrease in the liquid yield. the excessive deoxygenation that could increase the yield of aromatic compounds, and the catalyst deactivation [26]. The disadvantages of HDO are the high hydrogen consumption (reaching values over 700 m<sup>3</sup> (STP) per ton of bio oil [30,31]) and the low yield of pyrolysis liquid of high quality (at the best, 0.77 dm<sup>3</sup> dm<sup>-3</sup> oil fed [30]). According to Kersten et al. [26], this process would only make sense as a final step in the deoxygenation process. With DCO, deoxygenation by CO2 removal can be achieved with the appropriate catalyst, but the removal of oxygen is insufficient [26]. Upgrading by means of emulsification with diesel and biodiesel [32,33] and esterification [34-36] have also been tested with satisfactory results.

The improvement of the liquid can also be achieved by the pretreatment of the biomass used as feedstock by means of torrefaction [37–39]. Torrefaction (also called roasting or mild pyrolysis [40]) is a thermal treatment which takes place at temperatures between 200 °C and 300 °C under atmospheric pressure conditions and inert atmosphere [40,41]. Torrefaction reduces the oxygen content of lignocellulosic biomass due to the release of water, carbon dioxide, carbon monoxide and lightweight organic compounds [42]. A solid product with lower moisture content and higher energy density is obtained. In the existing literature there are some studies about the effect of torrefaction on the subsequent thermochemical processing of biomass whether through combustion [41,43–45] or gasification [46–49], but also about torrefaction as pre-treatment for pyrolysis [37,50-56]. In these studies, torrefaction and pyrolysis are normally carried out using fluidized bed reactors or auger reactors. Depending on the torrefaction conditions, liquid from pyrolysis of torrefied biomass has a lower water content and oxygen to carbon ratio [37,55] and higher stability [55,56] and energy content [51,55], but at the expense of a reduction in the liquid yield [37,50,51,53,56]. In view of these results, torrefaction pre-treatment could improve the properties of sewage sludge pyrolysis liquid as a fuel. To the best of our knowledge, torrefaction of sewage sludge as pre-treatment for the improvement of pyrolysis liquid properties has not yet been investigated in detail. Ábrego et al. [57] included torrefaction as one of the steps in a three-stage process aimed at valorizing sewage sludge, but they did not analyze the influence of torrefaction on pyrolysis liquid properties

This study describes the effect of sewage sludge torrefaction pre-treatment in a fluidized bed reactor (with temperatures between 220 °C and 320 °C and solid residence times between 3.6 min and 10.2 min) on the distribution and the properties of the products obtained after pyrolysis of the torrefied solid. This pyrolysis was also carried out in a fluidized bed reactor. Dry sewage sludge pyrolysis experiments were also carried out for comparison. The goal of the study is to identify whether sewage sludge torrefaction pre-treatment can enhance the fuel properties of pyrolysis liquid.

#### 2. Materials and methods

#### 2.1. Materials

The dry sewage sludge (SS) was provided by a Spanish wastewater treatment plant located in Madrid. The sewage sludge is anaerobically digested and thermally dried in the treatment plant, which generates over 33 kt per year of dry SS. The properties of the dry SS used are listed in Table 1. The bulk density ( $\rho$ ) was obtained by a non-standardized method. A known volume of dry SS was weighed and the density was calculated. The energy density ( $\rho_{\rm energ}$ ) results from the multiplication of the density by the higher heating value (HHV). The ash mineral composition of the dry SS, analyzed by Inductively Coupled Plasma-Atomic Emission

**Table 1**Properties of dry and torrefied sewage sludge.

	Analytical standard	Units	Dry SS	220/ 10.2	320/ 10.2	220/ 3.6	320/ 3.6	270/ 6.1	270/ 6.1	270/ 6.1	270/ 6.1	220/ 6.1	320/ 6.1	270/ 10.2	270/ 3.6
Carbon	a	% <sup>b</sup>	29.50	30.75	28.86	29.62	29.94	30.07	28.13	29.32	29.11	29.29	30.28	30.36	29.73
Hydrogen <sup>c</sup>	a	% <sup>b</sup>	4.67	4.96	3.27	4.90	4.50	4.68	4.50	4.61	4.65	4.71	3.41	4.57	3.95
Nitrogen	a	% <sup>b</sup>	5.27	5.15	4.71	4.95	5.02	5.18	4.75	4.99	4.83	5.01	5.21	5.14	5.08
Sulfur	a	% <sup>b</sup>	1.31	1.11	0.61	1.07	1.12	1.01	1.17	1.08	0.94	1.13	0.63	1.03	1.03
Oxygen	d	% <sup>b</sup>	20.20	16.90	7.83	18.26	15.24	16.30	19.16	18.35	17.57	18.97	8.44	15.82	18.43
O/C	e		0.37	0.33	0.12	0.31	0.32	0.36	0.40	0.36	0.38	0.38	0.16	0.32	0.35
H/C	e		1.59	1.76	1.19	1.67	1.66	1.71	1.68	1.65	1.76	1.70	1.29	1.66	1.36
Dry matter	ISO-589-1981	% <sup>b</sup>	93.52	96.2	96.5	93.3	97.2	96.8	95.3	95.1	96.9	95.4	98.8	97.0	95.1
Ash		% <sup>b</sup>	39.04 <sup>f</sup>	41.1 <sup>g</sup>	54.7 <sup>g</sup>	41.2 <sup>g</sup>	44.2 <sup>g</sup>	42.8 <sup>g</sup>	42.3 <sup>g</sup>	41.7 <sup>g</sup>	42.9 <sup>g</sup>	$40.9^{g}$	53.0 <sup>g</sup>	43.1 <sup>g</sup>	41.8 <sup>g</sup>
HHV <sup>h</sup>	ASTM D-3286-96	${ m MJ~kg^{-1}}$	12.79	12.90	12.05	13.02	13.62	13.39	13.27	13.23	13.02	13.08	13.25	13.49	13.04
ho		${ m kg}~{ m dm}^{-3}$	0.862	0.872	0.802	0.880	0.848	0.880	0.878	0.873	0.882	0.862	0.792	0.865	0.873
$ ho_{ener}$		${ m MJ~dm^{-3}}$	11.0	11.3	9.7	11.5	11.5	11.8	11.6	11.5	11.5	11.3	10.6	11.7	11.4

<sup>&</sup>lt;sup>a</sup> Ultimate analysis was performed using Carlo Erba 1108.

<sup>&</sup>lt;sup>b</sup> Mass fraction %.

<sup>&</sup>lt;sup>c</sup> The % of hydrogen includes the hydrogen from moisture.

d Oxygen (%) = 100 - carbon (%) - hydrogen (%) - nitrogen (%) - sulfur (%) - ash (%).

<sup>&</sup>lt;sup>e</sup> Molar basis; without moisture O and H.

<sup>&</sup>lt;sup>f</sup> The % of ash in the dry SS was determined according to ISO-1171-1976.

 $<sup>^{</sup>g}$  The % of ash in the TSS was determined taking into account the yield to TSS and the ash content of the raw SS: ash<sub>TSS</sub> (%) =  $100 \cdot \frac{\text{ash}_{SS}}{\eta_{TSS}}$  (%)

<sup>&</sup>lt;sup>h</sup> HHV was determined using IKA C 2000 Basic Calorimeter.

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