PYROLYSIS OF SEWAGE SLUDGE IN A BUBBLING FLUIDIZED BED: DETERMINATION OF THE REACTION RATE

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Abstract – The pyrolysis of sewage sludge in a lab-scale cylindrical reactor is studied. The reactor was surrounded by three electrical resistors connected to potentiometers that controlled the thermal power supplied by the resistors. The whole reactor and the electrical resistors rested on a high precision scale capable of detecting mass variations of 0.01 g. This work intends to determine the reaction rate, defined as the evolution of the percentage of the fuel volatile matter released as a function of time, during the pyrolysis of sewage sludge, comparing the values obtained in a fixed bed and in a bubbling fluidized bed. Sewage sludge was supplied to the reactor in batch of approximately 10 g by the top of the reactor once the desired bed temperature was reached. The sewage sludge mass evolution with time, measured by the scale, was recorded for each test. Two different bed temperatures of 500 and 600 °C were tested, using for both of them two different Nitrogen flowrates, one of them corresponding to a fixed bed and another one for a fluidized bed. The fluidization of the bed was found to increase the mass of volatiles released and drastically decrease the pyrolysis time of sewage sludge in comparison with the fixed bed tests.

INTRODUCTION

Over the last 50 years, a continuous growth of the world population has occurred, resulting in an increase of the primary energy consumption. Currently, more than 80% of the total primary energy consumption is based on fossil fuels, which are responsible for more than 98% of the CO₂ emissions to the atmosphere, causing the actual global warming problems (Demirbas and Demirbas, 2011). Therefore, there is an urgent need to evaluate the capabilities of alternative fuels which may substitute, or at least reduce, the dependence on fossil fuels, with lower associated pollutant emissions. Furthermore, the population growth in urban areas and the application of the Directive 91/271/EEC lead to the construction of Waste Water Treatment Plants across the European Union, causing also the problem of an increase in the sewage sludge production. According to the European Commission, the annual production of sewage sludge increased from around 5.5 million tonnes of dry matter in 1992 to nearly 9 million tonnes by the end of 2005. The volume of sewage sludge produced annually has continued increasing since 2005 and it is expected to continue rising (Manara and Zabaniotou, 2012).

Sewage sludge is the residue produced during the treatment of industrial or municipal wastewater. The physical, chemical and sanitary properties of sewage sludge might differ significantly depending on the sewage treatment and origin. Nevertheless, sewage sludge is characterized by a large moisture content, which causes the need of a previous drying process to reduce the water content. Regarding the dry sludge, high volatile matter and ash contents may be expected (Fan et al., 2014; Liu et al., 2015). The ways of disposing sewage sludge can be divided in three applications: landfill, agricultural use and incineration or thermochemical conversion (Fonts et al., 2012). Nevertheless, the European regulations try to limit the use of sewage sludge for landfill due to their environmental drawbacks, and the content of heavy metals and pathogens limit their use in agricultural applications. In contrast, the thermochemical conversion of sewage sludge presents several benefits, such as the possibility to recover energy, the reduction of the residue volume by 70% and the thermal destruction of pathogens (Fytili and Zabaniotou, 2008). Therefore, the thermochemical conversion with energy recovery might solve the problem of the increase in the volume of this residue produced due to the population growth, contributing to a reduction of the dependence on fossil fuels.

Several works have been focused on the thermochemical conversion of sewage sludge. Kijo-Kleczkowska et al. (2016) studied the combustion of sewage sludge in comparison with that of coal and biomass. The Lower Heating Value (LHV) of dry sewage sludge is around 20 MJ/kg, similar to that of coal (Jayaraman and Gökalp, 2015), thus a huge amount of energy is contained in the annual production of sewage sludge. Regarding sewage sludge gasification, Choi et al. (2016) analysed the syngas produced in a three-stages gasifier, obtaining a LHV of around 6 MJ/nm³, a similar value to that obtained by Gil-Lalaguna et al. (2014a).
for the gasification of sewage sludge char. Concerning the pyrolysis of sewage sludge, Fan et al. (2016) analysed the composition of the liquid fuel produced. The chemical kinetics of the thermochemical conversion of sewage sludge is typically determined by means of non-isothermal Thermogravimetric Analysis (TGA) (Jayaraman and Gökbalp, 2015).

The technology of Bubbling Fluidized Beds (BFB) is adequate for the conversion of highly volatile fuels such as biomass and organic waste, for which the conversion can occur in the bubbling bed at low temperatures without the need of in-bed heat exchangers (Leckner, 2016). The performance and emission level of BFBs are influenced by fuel mixing (Leckner, 1998). Poor fuel lateral mixing may produce a high concentration of volatile matter and char close to the fuel feeding ports (Gómez-Barea and Leckner, 2010) and, therefore, resulting in undesired temperature profiles (Winaya et al., 2007) and increased pollutant emissions (Leckner, 1998), especially when converting high-volatile fuels.

The thermochemical conversion of sewage sludge in bubbling fluidized beds has been analyzed recently by several authors. As an example of the work performed on the conversion of sewage sludge in BFBs, the incineration of sludge in BFBs was studied by Urciuolo et al. (2012) and Li et al. (2016b), the gasification was analysed by Gil-Lalaguna et al. (2014b) and Nilsson et al. (2013), and the pyrolysis was the focus of the works of Atienza-Martínez et al. (2015) and Jaramillo-Arango et al. (2016). Despite the existence of several works concerning sewage sludge conversion in BFBs, these works are normally specific for a particular thermochemical process and reactor, and there is a lack of general studies focusing on thermochemical conversion of sewage sludge in fluidized bed reactors, capable of measuring the mass released by the sewage sludge when it is immersed in the fluidized bed and moving freely.

In this work, the pyrolysis of sewage sludge in a lab-scale reactor was studied. The reactor was operated with two different Nitrogen flowrates, a low flowrate corresponding to a fixed bed reactor and a higher one corresponding to a fluidized bed reactor. Both for the fixed and the fluidized bed reactor, the sewage sludge was supplied through the top of the reactor in a batch of approximately 10 g, once the reactor temperature was stabilized at 500 °C or 600 °C. The whole reactor rested on a precision scale, so that the mass released by the sewage sludge during the pyrolysis process was measured and used to determine the reaction rate, the total mass of volatile matter released and the pyrolysis time. The fluidization of the reactor was found to decrease substantially the pyrolysis time and slightly increase the total volatile mass released during the pyrolysis.

**EXPERIMENTAL SETUP**

**Experimental Facility**

The experimental measurements were conducted in a cylindrical lab-scale bubbling fluidized bed reactor with an internal diameter, \( d_i \), of 4.7 cm and a total height from the distributor, \( h \), of 50 cm. The reactor, made of stainless steel, was covered by three electric resistors with a power of 500 W each one, one of them located around the plenum chamber and two around the reactor over the distributor. The power supplied to each resistor was controlled by potentiometers. The reactor was supplied with a Nitrogen flowrate determined by a flowmeter PFM750-F01-F from SMC with a measurement range from 1 to 50 l/min. The whole reactor rested on a scale PS 6000 R2 from RADWAG, capable of measuring a total mass of 6 kg with a precision of 0.01 g. A schematic of the experimental facility is shown in Figure 1.

**Bed Material Characterization**

The bed material employed in the tests was silica sand particles with a diameter, \( d_{ba,m} \), in the range 425 – 600 \( \mu \)m and a particle density, \( \rho_{ba,m} \), of 2600 kg/m\(^3\). A fixed bed height, \( h_b \), of 9.4 cm was employed, equivalent to a bed aspect ratio of 2. The mass of silica sand needed to reach this height is 240 g, corresponding to a void fraction, \( \varepsilon \), of 0.44.

The minimum fluidization velocity, \( U_{mf} \), was measured as a function of the bed temperature, \( T \), considering the variation of the gas density, \( \rho_g \), with temperature as described in Sánchez-Prieto et al. (2014). The density of the gas at the bed temperature was calculated considering the Nitrogen as an ideal gas:

\[
\rho_g = \frac{\rho_{g,amb} T_{amb}}{T}
\]

where \( \rho_g \) is the Nitrogen density at temperature \( T \) and \( \rho_{g,amb} \) is the Nitrogen density at the reference temperature \( T_{amb} \). The reference temperature was selected as \( T_{amb} = 300 \) K and the Nitrogen density at this temperature is \( \rho_{g,amb} = 1,14 \) kg/m\(^3\).
Figure 1: Schematic of the experimental facility.

The variation of the minimum fluidization velocity with the bed temperature can be estimated using the correlation of Carman-Kozeny (Carman, 1937):

$$U_{mf} = \frac{(\phi d_{bm})^2 (\rho_{bm} - \rho_g) g}{180 \mu_g} \frac{\varepsilon^3}{1 - \varepsilon}$$  \hspace{1cm} (2)

where $U_{mf}$ is the minimum fluidization velocity, $\phi$ is the sphericity of the dense phase particles, $\varepsilon$ is the void fraction, $g$ is the gravity acceleration, $d_{bm}$ is the diameter of the bed material particles, $\rho_{bm}$ is the density of the bed material particles, $\rho_g$ is the density of the Nitrogen at the bed temperature, and $\mu_g$ is the dynamic viscosity of the Nitrogen at the bed temperature. The variation of the dynamic viscosity of the Nitrogen with the bed temperature, $T$, can be determined by the potential law:

$$\mu_g = \mu_{g,amb} \left( \frac{T}{T_{amb}} \right)^{2/3}$$ \hspace{1cm} (3)

where the dynamic viscosity of Nitrogen at the reference temperature ($T_{amb} = 300$ K) is $\mu_{g,amb} = 1.78 \cdot 10^{-5}$ kg/(m·s).

The experimental results of the minimum fluidization velocity, $U_{mf}$, are plotted in Figure 2, together with the estimation obtained from the Carman-Kozeny correlation (Eq. 2), as a function of the bed temperature, $T$. An average bed material diameter of $d_{bm} = 512.5$ $\mu$m and a sphericity of $\phi = 0.8$ were employed for the estimation of $U_{mf}$ with the Carman-Kozeny correlation. Figure 2 shows a proper agreement between the experimental measurements of the minimum fluidization velocity and the estimation of the Carman-Kozeny correlation.

Figure 2: Variation of the minimum fluidization velocity with the bed temperature.
Sewage Sludge Characterization

The sludge employed in the experimental measurements was obtained from the municipal sewage treatment plant of Loeches (Madrid, Spain) in February 2016. The sludge was taken after the pre-drying process in a fluidized bed in the sewage treatment plant. The samples of sewage sludge were characterized by a proximate and an ultimate analysis. The proximate analysis was conducted in a TGA Q500 from TA Instruments to measure the moisture, ash, volatile matter, and fixed carbon contents of the sample. The moisture content was characterized as the mass released by the sample at 105 ºC. The ash content was determined as the percentage of mass remaining after a heating of the sample up to 550 ºC, supplying the furnace with an air flow rate of 60 ml/min using a heating rate of 10 K/min, and an isothermal process at 550 ºC until the mass of the sample stabilized. The volatile matter content of the samples was measured as the percentage of mass released by the sample during a heating process at a heating rate of 10 K/min from 105 ºC to 900 ºC and an isothermal process at 900 ºC in an inert atmosphere, obtained introducing a flux of 60 ml/min of Nitrogen in the furnace, until the mass of the sample stabilized. Finally, the fixed carbon content was obtained by difference.

The ultimate analysis of the sample was performed in a LECO TruSpec CHN analyzer, where the Carbon and Hydrogen contents of the sample are measured using an infrared absorption detector for the exhaust gases obtained from a complete combustion of the sample carried out in pure Oxygen. The Nitrogen content is determined conducting the exhaust gases through a thermal conductivity cell. The Carbon and Nitrogen contents are measured with a precision of ±0.5%, while the precision of the measurement of the Hydrogen content is ±1%.

The results of the proximate and ultimate analyses of the sewage sludge samples are included in Table 1. The values obtained for the characterization of the sewage sludge are similar to those obtained for different authors, such as Scott et al. (2006), Soria-Verdugo et al. (2013), Jayaraman et al. (2015), and Liu et al. (2015).

Table 1: Results obtained from the proximate and ultimate analysis of the sewage sludge (d: dry basis, daf: dry-ass-free basis, * obtained by difference).

<table>
<thead>
<tr>
<th>Proximate analysis</th>
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<tbody>
<tr>
<td>Volatile matter [% d]</td>
<td>57.11</td>
</tr>
<tr>
<td>Fixed carbon* [% d]</td>
<td>34.66</td>
</tr>
<tr>
<td>Ash [% d]</td>
<td>8.23</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Elemental analysis</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>C [% daf]</td>
<td>56.46</td>
</tr>
<tr>
<td>H [% daf]</td>
<td>7.91</td>
</tr>
<tr>
<td>N [% daf]</td>
<td>8.42</td>
</tr>
<tr>
<td>O* [% daf]</td>
<td>27.21</td>
</tr>
</tbody>
</table>

EXPERIMENTAL MEASUREMENTS

The experimental measurements of the pyrolysis of sewage sludge were conducted in the cylindrical reactor described above, which rested on a high precision scale. The potentiometers controlling the thermal power supplied by each electrical resistor was adjusted to obtained the desired bed temperature $T$ once the Nitrogen velocity of the test, $U$, was selected. Once the desired bed temperature is reached, the scale was tared and the sewage sludge was supplied in a batch of around 10 g by the top of the reactor. The sewage sludge particles were previously sieved to a particle size under 3 mm. Prior to the pyrolysis tests in the reactor, the sewage sludge samples were dried completely at 105 ºC in a Universal Oven UFE 500 from Memmert during 5 hours. The experiment consisted on recording the mass released by the sewage sludge during the pyrolysis process in the reactor. Two different bed temperatures of 500 and 600 ºC were tested, using in both cases two different gas velocities, a low gas velocity corresponding to a fixed bed ($U = 0.85U_{mf}$) and a higher Nitrogen velocity for a fluidized bed ($U = 2U_{mf}$).

Figure 3 shows and example of the mass signal measured by the scale during the pyrolysis of sewage sludge in the reactor at 600 ºC. As shown in Figure 3, the mass signal measured by the high precision scale is quite stable when the bed is operated as a fixed bed. Nevertheless, the vibration induced by the motion of bubbles when the bed is fluidized is detected by the scale, obtaining a variation of the mass measured. The vibration of the mass detected in the raw signal measured by the scale when the bed is fluidized can be smoothed by filtering the signal using a moving average filter, obtaining a clean mass variation with time for the pyrolysis process of sewage sludge in a fluidized bed. In the case of the pyrolysis measurements in a fixed bed, the raw
signal of the mass obtained from the scale was stable, so that the raw and the filtered signal collapsed in a curve.

Figure 3: Raw and filtered signal obtained from the mass variation measurement of the scale during the pyrolysis of sewage sludge at 600 °C, a) Fixed bed (\(U = 0.85U_{mf}\)), b) Fluidized bed (\(U = 2U_{mf}\)).

RESULTS AND DISCUSSION

The filtered signals of the mass variation measured by the scale were employed to determine the evolution with time, \(t\), of the percentage of mass released, \(X\), by the sewage sludge during the pyrolysis process, both in the fixed (\(U = 0.85U_{mf}\)) and in the fluidized bed (\(U = 2U_{mf}\)), for bed temperatures of 500 and 600 °C. The results are shown in Figure 4, where a slight increase of the total volatile matter released during the pyrolysis and a reduction of the pyrolysis time when increasing the reactor temperature can be observed for both the fixed and the fluidized bed tests, as a consequence of the increase of the heat transfer with the temperature of the reactor. Furthermore, operating at the same reactor temperature, the pyrolysis process occurs much faster in a fluidized bed than in a fixed bed reactor, obtaining also a slightly higher total volatile matter released during the pyrolysis of sewage sludge.

Figure 4: Evolution with time of the percentage of mass released during the sewage sludge pyrolysis.

The derivative of the percentage of the mass released, \(dX/dt\), by the sewage sludge during the pyrolysis process is plotted in Figure 5 for the four different tests analyzed. The figure shows a higher derivative of the percentage of mass released for the pyrolysis process in a fluidized bed and for a higher temperature of the reactor. The high mixing rate characteristic of fluidized beds caused a high initial value of the derivative, which decreased from this initial value as the pyrolysis tests progressed. In contrast, the poor mixing typical of fixed beds produced low values of the derivative of the percentage of mass released during the pyrolysis, which slightly increase at the beginning of the test, reaching a maximum value a few seconds after the beginning of the sewage sludge injection.
The reaction rate, \( V/V^* \), defined as the evolution of percentage of the volatile matter released by the sewage sludge as a function of time, can be determined from the percentage of mass released, \( X \), shown in Figure 4 as:

\[
V / V^* = 100 \cdot \frac{100 - X}{X_{vol}}
\] (4)

where \( V/V^* \) is the reaction rate (%), \( X \) is the evolution with time of the mass released during the pyrolysis process (%) and \( X_{vol} \) is the percentage of the initial mass injected released during the whole pyrolysis process (%).

The results of the evolution of the reaction rate with time for the pyrolysis of sewage sludge in fixed and fluidized bed reactors at 500 and 600 ºC can be observed in Figure 6. The pyrolysis time was found to decrease when the reactor temperature increased, for both the fixed and the fluidized bed, because of a higher heat transfer to the sewage sludge. Moreover, a clear reduction of the time needed for the pyrolysis of sewage sludge was found when the reaction was carried out in a fluidized bed, due to the higher heat and mass transfer coefficients of fluidized beds in comparison to those of fixed beds (Kunii and Levenspiel, 1969).
and 600 °C respectively. Therefore, the increase of both the heat and mass transfer coefficients obtained in a fluidized bed was proved with the pyrolysis tests of sewage sludge.

![Image]

Figure 7: a) Total percentage of volatile matter released and b) pyrolysis time during the sewage sludge pyrolysis process.

CONCLUSIONS

The pyrolysis of sewage sludge was studied in a lab-scale fixed and fluidized bed reactor. The mass released by the sewage sludge during the pyrolysis process after an injection of a batch of approximately 10 g in a hot reactor was measured by a high precision scale in which the hole reactor rested. The mass measured by the scale was smoothed using a moving average filter, employing the filtered signal to determine the percentage of mass released by the sewage sludge and the reaction rate during the pyrolysis reaction both in a fixed and in a fluidized bed operated at temperatures of 500 and 600 °C. The pyrolysis reaction in the fluidized bed was found to be much faster than in a fixed bed, obtaining also a greater volatile matter released by the sewage sludge, confirming the higher heat and mass transfer coefficients characteristic of fluidized beds.

NOTATION

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>d_{bm}</td>
<td>diameter of the bed material particles, m</td>
</tr>
<tr>
<td>d_i</td>
<td>inner diameter of the reactor, m</td>
</tr>
<tr>
<td>g</td>
<td>gravity acceleration, m/s²</td>
</tr>
<tr>
<td>h</td>
<td>reactor height, m</td>
</tr>
<tr>
<td>h_b</td>
<td>fixed bed height, m</td>
</tr>
<tr>
<td>m</td>
<td>mass of sewage sludge, kg</td>
</tr>
<tr>
<td>t</td>
<td>time, s</td>
</tr>
<tr>
<td>t_{pyr}</td>
<td>pyrolysis time, s</td>
</tr>
<tr>
<td>T</td>
<td>reactor temperature, °C</td>
</tr>
<tr>
<td>T_{amb}</td>
<td>reference temperature, °C</td>
</tr>
<tr>
<td>U</td>
<td>gas velocity, m/s</td>
</tr>
<tr>
<td>U_{mf}</td>
<td>minimum fluidization velocity, m/s</td>
</tr>
<tr>
<td>V/V*</td>
<td>reaction rate, %</td>
</tr>
<tr>
<td>X</td>
<td>percentage of mass released, %</td>
</tr>
<tr>
<td>X_{vol}</td>
<td>total volatile matter released, %</td>
</tr>
<tr>
<td>ε</td>
<td>void fraction of the bed material, -</td>
</tr>
<tr>
<td>ρ_{bm}</td>
<td>density of the bed material particles, kg/m³</td>
</tr>
<tr>
<td>ρ_g</td>
<td>gas density, kg/m³</td>
</tr>
<tr>
<td>ρ_{g,amb}</td>
<td>gas density at T_{amb}, kg/m³</td>
</tr>
<tr>
<td>μ_g</td>
<td>gas dynamic viscosity, kg/ms</td>
</tr>
<tr>
<td>μ_{g,amb}</td>
<td>gas dynamic viscosity at T_{amb}, kg/ms</td>
</tr>
<tr>
<td>φ</td>
<td>sphericity of the bed material particles, -</td>
</tr>
</tbody>
</table>

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REFERENCES


