

# Effect of Segregation on the Reaction Rate of Sewage Sludge Pyrolysis in a Bubbling Fluidized Bed

A. Soria-Verdugo, A. Morato-Godino, L. M. García-Gutiérrez, N. García-Hernando

**Abstract**—The evolution of the pyrolysis of sewage sludge in a fixed and a fluidized bed was analyzed using a novel measuring technique. This original measuring technique consists of installing the whole reactor over a precision scale, capable of measuring the mass of the complete reactor with enough precision to detect the mass released by the sewage sludge sample during its pyrolysis. The inert conditions required for the pyrolysis process were obtained supplying the bed with a nitrogen flowrate, and the bed temperature was adjusted to either 500 °C or 600 °C using a group of three electric resistors. The sewage sludge sample was supplied through the top of the bed in a batch of 10 g. The measurement of the mass released by the sewage sludge sample was employed to determine the evolution of the reaction rate during the pyrolysis, the total amount of volatile matter released, and the pyrolysis time. The pyrolysis tests of sewage sludge in the fluidized bed were conducted using two different bed materials of the same size but different densities: silica sand and sepiolite particles. The higher density of silica sand particles induces a flotsam behavior for the sewage sludge particles which move close to the bed surface. In contrast, the lower density of sepiolite produces a neutrally-buoyant behavior for the sewage sludge particles, which shows a proper circulation throughout the whole bed in this case. The analysis of the evolution of the pyrolysis process in both fluidized beds show that the pyrolysis is faster when buoyancy effects are negligible, i.e. in the bed conformed by sepiolite particles. Moreover, sepiolite was found to show an absorbent capability for the volatile matter released during the pyrolysis of sewage sludge.

**Keywords**—Bubbling fluidized bed, pyrolysis time, segregation effects, sewage sludge.

## I. INTRODUCTION

AN increase of the primary energy consumption has occurred in the world over the last 50 years, caused by a continuous growth of the world population. The high dependence on fossil fuels, corresponding to more than 80% the primary energy consumed, is responsible for more than 98% the total carbon dioxide (CO<sub>2</sub>) emissions [1]. These CO<sub>2</sub> emissions associated to the conversion of fossil fuels induce serious global warming problems, causing an urgent need to analyze the use of alternative fuels with lower associated pollutant emissions.

Sewage sludge is the residue produced during the treatment of industrial or municipal wastewater. Currently, the main ways of disposing sewage sludge can be divided into three

applications: landfill, agricultural use, and incineration or thermochemical conversion [2]. Nonetheless, the amount of sewage sludge used for landfill is limited by the European regulations. Concerning the agricultural use, sewage sludge is suitable as a fertilizer due to their content on organic matter, nitrogen, and phosphorus. However, the sludge may also concentrate heavy metals and pathogens, which could cause significant environmental problems. In contrast to the drawbacks of the use of sewage sludge for landfilling or agriculture, the thermochemical conversion of sewage sludge [3] counts on several benefits, such as the possibility to recover energy [4], the reduction of the residue volume by 70%, and the thermal destruction of pathogens [5]. 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. Therefore, the thermochemical conversion of sewage sludge with energy recovery might solve the issue of the increase in residues produced due to the population growth, contributing to a reduction of the dependence on fossil fuels.

The physical, chemical, and sanitary properties of sewage sludge might differ significantly depending on the sewage treatment and origin. Nevertheless, sewage sludge is characterized in general 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 are also characteristic of sewage sludge [6], [7].

The technology of Bubbling Fluidized Beds (BFBs) 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 [8]. The performance efficiency and emission level of BFBs are influenced by fuel mixing [9]. Poor fuel mixing may produce a high concentration of volatile matter and char close to the fuel feeding ports [10], resulting in undesired temperature profiles [11] and an increase of pollutant emissions [9], especially when converting high-volatile fuels. The axial mixing of fuel particles in a BFB depends on the differences in density of the fuel particle and the bulk bed. Fuel particles light compared to the bed, usually named flotsam, are typically found close to the bed surface, whereas fuel particles dense in comparison with the bulk bed, known as jetsam, typically dive directly to the bottom of the bed and rest over the bed distributor [12]. Nevertheless, fuel particles with a density close to the bulk

A. Soria-Verdugo is with the University Carlos III, Department of Thermal Engineering and Fluid Mechanics. Avda. de la Universidad 30, 28911, Leganés, Madrid, Spain (phone: +34 91 624 8465; e-mail: asoria@ing.uc3m.es).

A. Morato-Godino, L. M. Garcia-Gutierrez, N. Garcia-Hernando is with the University Carlos III, Department of Thermal Engineering and Fluid Mechanics, Madrid, Spain (e-mail: amorato@ing.uc3m.es, lmgarcia@ing.uc3m.es, ngarcia@ing.uc3m.es).

bed show a proper circulation throughout the whole bed, obtaining a high axial mixing of these fuel particles in the bed [13].

Pyrolysis is the thermal degradation of a solid fuel caused by an increase of temperature in absence of oxygen. The degradation is produced by the dissociation of the chemical bonds of the large molecules that compose the solid fuel due to the thermal energy applied, generating a gaseous mixture, known as pyrolysis vapors, which is composed of lighter molecules. The molecules found in the pyrolysis vapors can be divided into two main categories: i) condensable gases, which can be transform into a liquid fuel when the temperature is reduced, and ii) non-condensable or permanent gases, which is a stable gas fuel. In comparison to the other thermochemical conversion processes, such as combustion or gasification, pyrolysis presents the advantage of producing mainly an easy to store and transport liquid or gaseous product, in particular for those fuels characterized by high volatile matter and low fixed carbon content, such as sewage sludge. Pyrolysis was found to be the optimal thermochemical process for sewage sludge by [14], due to its favorable energy balance, material recovery, and zero-waste conversion.

The products obtained from the pyrolysis of sewage sludge in a fluidized bed reactor are affected by the operating conditions, such as fuel particle diameter, pyrolysis vapors residence time, and reactor temperature. Among the operating conditions, the bed temperature is the most influential parameter; in fact, several authors have focused their research on analyzing its effect on the liquid yield [15]-[17]. The optimal temperature to maximize the liquid yield produced is a moderate temperature around 550 °C, since for lower temperatures the energy supplied to the solid particles of sewage sludge is insufficient to release all the volatile matter content, whereas for higher temperatures secondary cracking reactions of the pyrolysis vapors occur, resulting in an increase of non-condensable gases, reducing the liquid yield generated by condensation. Sun et al. [18] studied the pyrolysis of sewage sludge in a fluidized bed reactor for a wide range of temperatures between 300 °C and 900 °C, concluding that the maximum liquid production from the condensation of the pyrolysis vapors was obtained at moderate temperatures of around 550 °C. In fact, in a following study [19], they focused on the analysis of sewage sludge pyrolysis on a narrow temperature range of 400-600 °C.

In this work, the pyrolysis of sewage sludge was analyzed in a lab-scale reactor. The reactor was operated either as a fixed bed or as a fluidized bed using a lower or a higher nitrogen flowrate, respectively. The sewage sludge sample was supplied through the top of the reactor in a batch of 10 g, once the reactor temperature was stabilized at 500 °C or 600 °C for both the fixed and the fluidized bed. The whole reactor was installed over a high precision scale, so that the mass released by the sewage sludge during the pyrolysis process was measured and used to determine the reaction rate of the sewage sludge, the total mass of volatile matter released, and the pyrolysis time. The bed material employed during the pyrolysis in the fluidized bed was varied, using silica sand and

sepiolite (SG36) particles of the same size but with a different density to analyze the buoyancy effects of the sewage sludge particles on the evolution of the pyrolysis process.

## II. EXPERIMENTAL SETUP

### A. Experimental Facility

The experimental measurements of the pyrolysis of sewage sludge were conducted in a cylindrical lab-scale reactor with and internal diameter,  $d_i$ , of 4.7 cm and a height from the distributor,  $h$ , of 50 cm. The reactor was made of stainless steel and was surrounded by three electric resistors with a power of 500 W each one, one of them located around the plenum chamber and the other two around the bed, above the distributor. The thermal power supplied by each resistor was controlled by a potentiometer. The reactor was supplied with a Nitrogen flowrate measured by a flowmeter PFM750-F01-F from SMC with a measurement range from 1 to 50 l/min. Different Nitrogen flowrates were employed to characterize the differences of the pyrolysis process in a fixed bed and in a BFB. The whole reactor, together with the electric resistors, 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 illustration of the experimental facility is shown in Fig. 1.

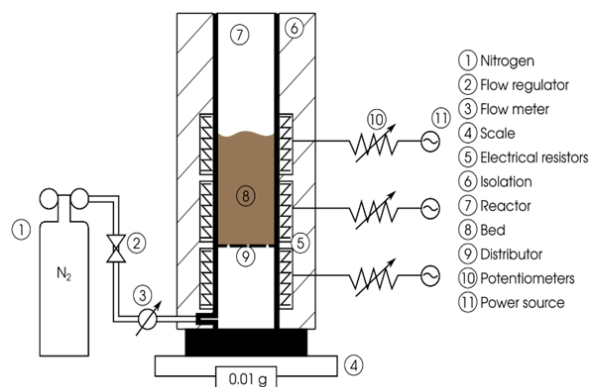


Fig. 1 Schematic illustration of the experimental facility

### B. Bed Material Characterization

Two different bed materials were employed during the pyrolysis tests to analyze the effect of the bed density on the reaction rate of the sewage sludge sample during the pyrolysis process. The particle sizes,  $d_{bm}$ , of both bed materials were identical, in the range of 425 – 600  $\mu\text{m}$ , but their particle densities,  $\rho_{bm}$ , differ, being 2600  $\text{kg}/\text{m}^3$  for the silica sand particles and 1550  $\text{kg}/\text{m}^3$  for the sepiolite (SG36) particles. The fixed bed height,  $h_b$ , was 9.4 cm for both bed materials, 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,  $\epsilon$ , of 0.44, whereas the mass of sepiolite to reach  $h_b$  is 110 g, obtaining a void fraction  $\epsilon$ , of 0.56.

The minimum fluidization velocity,  $U_{mf}$ , was measured for each bed material as a function of the bed temperature,  $T$ . The measurement of  $U_{mf}$  was compared to the estimation of

Carman-Kozeny [20], considering the variation of the gas density,  $\rho_g$ , with temperature as described in Sánchez-Prieto et al. [21]. The density of the gas for a determined bed temperature was calculated considering Nitrogen as an ideal gas:

$$\rho_g = \rho_{g,amb} \frac{T_{amb}}{T}, \quad (1)$$

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<sup>3</sup>.

The variation of the minimum fluidization velocity with the bed temperature can be estimated using the correlation of Carman-Kozeny [20]:

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

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 Nitrogen with the bed temperature,  $T$ , can be determined by the potential law:

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

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,  $U_{mf}$ , measured for both the silica sand, and the sepiolite particles are plotted in Fig. 2, together with the estimation obtained from the Carman-Kozeny correlation (2) for each case, as a function of the bed temperature,  $T$ . An average bed material diameter of  $d_{bm} = 512.5$   $\mu$ m was used for the estimation in both cases, while a sphericity of  $\phi = 0.8$  was employed of the silica sand particles and  $\phi = 0.5$  for the sepiolite particles to estimate the variation of the minimum fluidization velocity,  $U_{mf}$ , with the bed temperature  $T$  using the Carman-Kozeny correlation. Fig. 2 shows a proper agreement between the experimental measurements of the minimum fluidization velocity and the estimation of the Carman-Kozeny correlation for both the silica sand and the sepiolite particles, although the accuracy of the correlation is higher for the silica sand particles than for the sepiolite (SG36) particles.

### C. Sewage Sludge Characterization

The sewage sludge employed in the experimental measurements was obtained from the municipal sewage treatment plant of Loeches (Madrid, Spain) in February 2016. The sludge analyzed was pre-dried in a fluidized bed at 80 °C to a water content under 10% in the sewage treatment plant. The samples of sewage sludge were characterized by a proximate and an ultimate analysis. The moisture, volatile matter, fixed carbon and ash contents of the sewage sludge were determined by a proximate analysis conducted in a thermogravimetric analyzer (TGA) Q500 from TA Instruments. The moisture content was calculated as the mass released by the sample at 105 °C in an inert atmosphere. The ash content was determined as the percentage of mass remaining after heating the sample up to 550 °C using a heating rate of 10 K/min, supplying the furnace with an air flow rate of 60 ml/min, and maintaining a constant temperature of 550 °C until no variation of the mass was detected. 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 °C to 900 °C in an inert atmosphere, obtained introducing a flux of 60 ml/min of nitrogen in the furnace, and carrying out an isothermal process at 900 °C until the mass of the sample is stabilized. Finally, the fixed carbon content was obtained by difference.

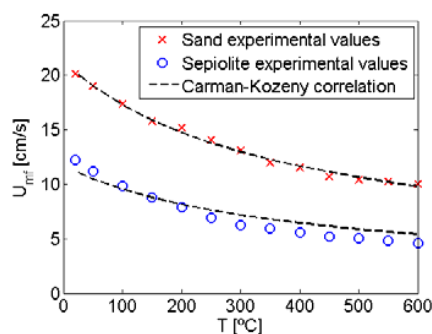


Fig. 2 Variation of the minimum fluidization velocity with the bed temperature

The carbon, hydrogen, and nitrogen contents of the sewage sludge were determined performing an ultimate analysis in a LECO TruSpec CHN analyzer. The carbon and hydrogen contents of the sample were measured using an infrared absorption detector for the exhaust gases obtained from a complete combustion of the sewage sludge carried out in pure oxygen. In contrast, the nitrogen content was determined conducting the exhaust gases through a thermal conductivity cell. The precision in the measurement of the carbon and nitrogen contents is  $\pm 0.5\%$ , whereas the precision of the measurement of the hydrogen content is  $\pm 1\%$ .

Table I shows the results of the proximate and ultimate analyses of the sewage sludge used in the pyrolysis tests. The values obtained for the characterization of the sewage sludge are similar to those obtained for different authors, such as [8],

[22]-[24].

The TGA Q500 from TA Instruments was also employed to study the pyrolysis process of the sewage sludge samples under non-isothermal conditions. A mass of 10 mg of sewage sludge was employed, and a blank experiment was performed for each heating rate to avoid buoyancy effects. Nine constant values of the heating rate (10, 13, 16, 19, 22, 25, 30, 35, 40 K/min) were used to heat the sewage sludge samples from 100 °C to 700 °C, under an inert atmosphere obtained supplying the TGA furnace with a nitrogen flowrate of 60 ml/min. From the TGA curves obtained, the kinetic parameters of the pyrolysis of sewage sludge, i.e. the activation energy and the pre-exponential factor, can be determined. Fig. 3 shows the results for the activation energy and the pre-exponential factor of the sewage sludge as a function reacted fraction,  $V/V^*$ , obtained applying the Distributed Activation Energy Model (DAEM). Further details of the mathematical procedure to obtain the values of the activation energy and the pre-exponential factor from the TGA curves can be found in a previous work [25]-[27].

TABLE I  
RESULTS OBTAINED FROM THE PROXIMATE AND ULTIMATE ANALYSIS OF THE SEWAGE SLUDGE

Proximate Analysis	
Volatile matter [%d]	57.11
Fized carbon* [%d]	8.23
Ash [%d]	34.66
Ultimate Analysis	
C [%daf]	56.46
H [%daf]	7.91
N [%daf]	8.42
O* [%daf]	27.21

d: dry basis, daf: dry-ass-free basis, \* obtained by difference

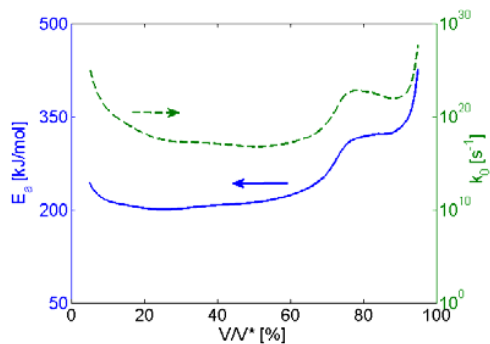


Fig. 3 Activation energy and pre-exponential factor of the pyrolysis reaction of sewage sludge

### III. RESULTS AND DISCUSSION

#### A. Pyrolysis Experimental Measurements

The experiments conducted to characterize the evolution of the pyrolysis process of sewage sludge consisted of recording the mass signal measured by the scale while the pyrolysis reaction of the sewage sludge sample occurred inside the reactor. The high precision in the measurement of the mass permitted the determination of the mass released by the

sample during its pyrolysis. First, the reactor was filled with the bed material particles, either silica sand or sepiolite, that conformed the bed, and the system was heated by the resistors to the desired reactor temperature,  $T$ . Once the reactor temperature of the test was reached, the flowrate of nitrogen, employed as an inert gas, was adjusted to the lower velocity for the fixed bed experiments or to the higher velocity for the fluidized bed tests. When the operating conditions of the bed, i.e. reactor temperature and nitrogen flowrate, were selected, the scale, in which the whole system rested, was tared, and a batch of 10 g of dry sewage sludge particles was introduced through the top of the reactor. When the bed was fluidized, the mass signal measured by the scale during the pyrolysis of the sewage sludge registered the vibration induced by the ascension of bubbles. This vibration of the mass can be filtered using a moving average. Further details of the filtration of the mass signals can be found in a previous work [28].

Different operating conditions were tested, varying the reactor temperature, the fluidizing gas velocity, and the bed material. Two different nitrogen velocities were tested to analyze the differences obtained in the pyrolysis process of sewage sludge occurring in a fixed ( $U = 0.8U_{mf}$ ) or a fluidized bed ( $U = 2.5U_{mf}$ ). For each nitrogen velocity, two different bed temperatures were studied. The temperatures tested, 500 °C and 600 °C, were selected as those for which the liquid yield obtained from the condensation of the pyrolysis vapors is maximized. Finally, in the case of the fluidized bed tests, the effect of the bed density was also analyzed, for each temperature, using two different bed materials: silica sand and sepiolite particles.

#### B. Reaction Rate during the Pyrolysis Process

The mass released by the sewage sludge during the pyrolysis process was measured by the scale and recorded for each operating condition. The mass signal was filtered to remove the vibration induced by the ascension of bubbles when the bed was fluidized, and the filtered signal was divided by the initial mass of sewage sludge supplied to the bed to obtain the percentage of mass remaining inside the reactor,  $X$ . Fig. 4 shows the evolution of the percentage of mass remaining in the reactor as a function of the bed temperature for both the fixed and the fluidized bed using silica sand and sepiolite particles. As can be observed in Fig. 4, the reactor temperature affects the velocity of the pyrolysis process, which occurs faster for higher temperatures. Nonetheless, a stronger effect than that of the bed temperature can be observed when increasing the nitrogen velocity from that of a fixed bed ( $U = 0.8U_{mf}$ ) to that of a fluidized bed ( $U = 2.5U_{mf}$ ). The pyrolysis process of sewage sludge is accelerated in a fluidized bed due to the higher axial dispersion of the particles of sewage sludge, which leads to a higher heating rate of the sludge particles. Concerning the bed material that conformed the bed, a slight increase of the pyrolysis reaction velocity can be observed for the sepiolite particles fluidized bed. This slight increase in the pyrolysis velocity can be attributed to the increase in the heating rate of the sewage sludge particles in

the sepiolite bed, where buoyancy effects are negligible and the sludge particles circulate properly throughout the whole bed. In contrast, the lower density of the sewage sludge particles in comparison to the bulk density of the bed of silica sand particles induces buoyancy effects on the sludge particles, which move in a narrow region close to the bed surface, resulting in a reduction of the heating rate of the fuel particles.

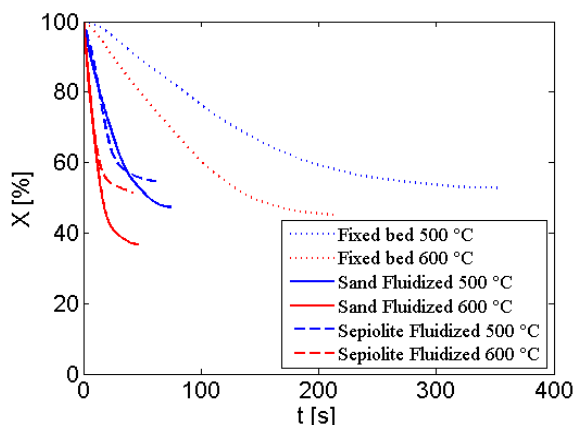


Fig. 4 Evolution of the percentage of mass of the sample remaining in the reactor during the pyrolysis process

Fig. 4 shows also a variation of the mass of the solid residue remaining in the reactor once the pyrolysis process is completed. The percentage of the sample mass remaining after the complete pyrolysis,  $X_{res}$ , was determined for each operating condition and employed to calculate the percentage of volatile matter released during the pyrolysis,  $X_{vol}$ , as  $X_{vol} = 100 - X_{res}$ . The results of  $X_{vol}$  are shown in Fig. 5, along with the percentage of mass of volatile matter released in a pyrolysis test conducted in a TGA. The measurement in the TGA was carried out heating an initial mass of 10 mg of sewage sludge to either 500 °C or 600 °C, supplying the furnace with 60 ml/min of nitrogen to guarantee the inert conditions necessary for the pyrolysis process.

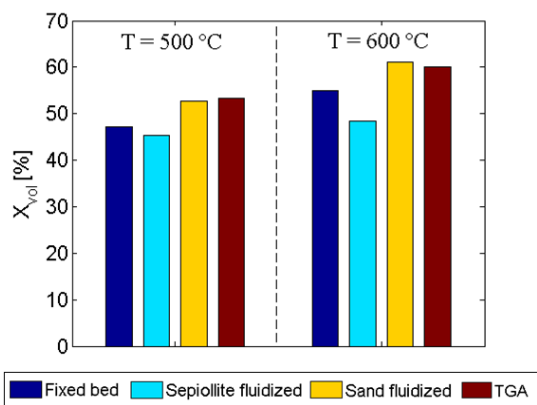


Fig. 5 Percentage of mass of volatile matter released during the pyrolysis of sewage sludge

It can be observed in Fig. 5 that the percentage of volatile matter released by the sewage sludge sample in a fluidized bed conformed by silica sand particles is similar to the total amount of volatile matter released, characterized by the TGA tests, for the two different bed temperatures analyzed. In contrast, when the pyrolysis occurs in a fixed bed, the amount of volatile matter released is lower than in the fluidized bed of sand particles for both temperatures, as a result of the heat transfer effects inside the sewage sludge sample, which is located stagnant over the bed surface in the fixed bed. The amount of volatile matter released by the sewage sludge during the pyrolysis in a fluidized bed of sepiolite particles is significantly lower than that in a fluidized bed of silica sand. Therefore, it can be stated that sepiolite particles absorb part of the volatile matter released by the sewage sludge, contributing to obtain a lower percentage of volatile matter released by the bed.

The reaction rate can be determined as the derivative of the sample mass remaining inside the reactor,  $X$ , shown in Fig. 4. The results of the reaction rate for each operating condition can be observed in Fig. 6. The reaction rate of the pyrolysis of sewage sludge is substantially increased when the reactor is operated as a fluidized bed in comparison to the operation as a fixed bed, as a consequence of the higher heating rate of the sewage sludge particles in a fluidized bed produced by the higher dispersion of the particles induced by the motion of bubbles. Furthermore, a slight increase of the reaction rate can be also observed for a fluidized bed conformed by sepiolite particles compared to a bed conformed by silica sand particles. The reaction rate can be also increased for all the cases tested by increasing the reactor temperature; however, the effect of temperature is lower than that of fluidizing the bed.

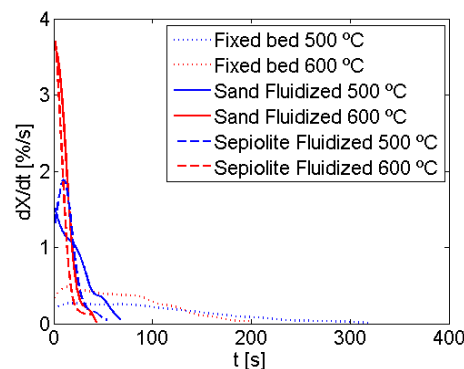


Fig. 6 Evolution of the reaction rate during the pyrolysis of sewage sludge

### C. Pyrolysis Time

The effect of the variable percentage of volatile matter released during the pyrolysis process depending on the operating conditions, shown in Fig. 5, can be removed by calculating the reacted fraction,  $V/V^*$ . The reacted fraction is defined as the percentage of volatile matter released by the sewage sludge sample during the pyrolysis process as a function of time, and can be determined from the percentage of mass remaining inside the reactor,  $X$ , shown in Fig. 4, as:

$$V/V^* = 100 \frac{100 - X}{X_{vol}}, \quad (4)$$

where  $V/V^*$  is the reacted fraction (%),  $X$  is the mass of the sample remaining in the reactor during the pyrolysis process (%), and  $X_{vol}$  is the percentage of the initial mass released as volatile matter during the complete pyrolysis process (%). Therefore,  $V/V^* = 0$  at the beginning of the pyrolysis process and  $V/V^* = 100\%$  when the pyrolysis process is completed for all the cases.

The results of the evolution of the reacted fraction with time for the pyrolysis of sewage sludge under the different experimental conditions tested are shown in Fig. 7. A substantial acceleration of the pyrolysis of sewage sludge can be observed in Fig. 7 when the process is carried out in a fluidized bed reactor in comparison to a fixed bed reactor. Concerning the pyrolysis in a fluidized bed reactor, the process is slightly faster in the fluidized bed conformed by sepiolite particles than in a bed of silica sand particles, i.e. when the fuel particles buoyancy effects are negligible. This fact can be attributed to a higher heating rate of the fuel particles when they are widely dispersed in the axial direction. Furthermore, an increase of the bed temperature also accelerates the pyrolysis process of sewage sludge; however, the effect of the bed temperature is more important when the pyrolysis occurs in a fixed bed reactor.

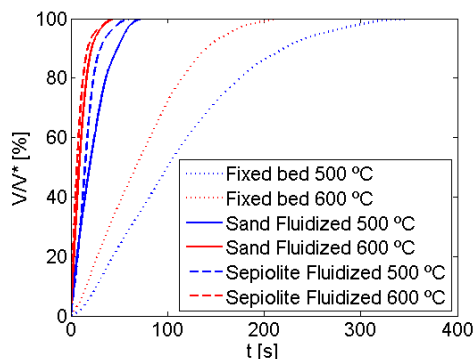


Fig. 7 Evolution of the reacted fraction during the pyrolysis of sewage sludge

The pyrolysis time,  $t_{pyr}$ , can be calculated for each experimental case as the time needed to reach a determined value of the reacted fraction,  $V/V^*$ . In this work, a value of  $V/V^* = 95\%$  was selected to calculate the pyrolysis time. Fig. 8 shows the pyrolysis time for the different pyrolysis tests conducted. Comparing the pyrolysis of sewage sludge in a fixed bed with that in a typical fluidized bed conformed by silica sand particles, the pyrolysis time was reduced by 79% for a bed temperature of 500 °C and by 83% when the bed temperature was increased to 600 °C. Therefore, the fluidization of the bed is proved to contribute to accelerate significantly the pyrolysis process due to the higher heating rates of the fuel particles. This reduction of the pyrolysis time when the pyrolysis is carried out in a fluidized bed reactor

permits an increase of the sewage sludge feeding rate during a continuous operation, which increases the conversion rate of this solid residue into a liquid or a gaseous fuel.

Concerning the use of different bed materials in the fluidized bed reactor during the pyrolysis of sewage sludge, a slight reduction of the pyrolysis time in the bed of sepiolite particles in comparison to a fluidized bed of silica sand particles can be observed. This reduction is a consequence of the higher fuel axial mixing in a bed of lower density, i.e. the bed conformed by sepiolite particles, where the buoyancy effects of fuel particles are negligible and the particles circulate properly throughout the whole bed height. Even though the pyrolysis time can be slightly reduced in a fluidized bed of sepiolite particles, it should also be considered that sepiolite is not an inert particle during the pyrolysis of biomass, like silica sand particles. In fact, sepiolite particles absorb part of the volatile matter released by sewage sludge during its pyrolysis, as can be observed in Fig. 5.

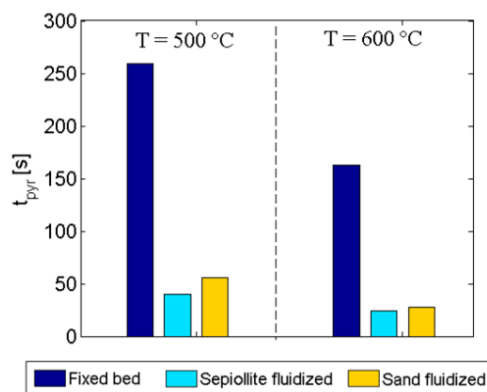


Fig. 8 Pyrolysis time of sewage sludge

#### IV. CONCLUSION

The pyrolysis of sewage sludge in a lab-scale cylindrical reactor was studied using a novel measuring technique consisting in measuring the mass of the whole reactor with enough accuracy to detect the mass released during the pyrolysis. The temperature of the reactor was varied between 500 °C and 600 °C, and two different nitrogen flowrates were analyzed in each case, a low flowrate corresponding to a fixed bed and a higher velocity to properly fluidize the bed. For the fluidized bed pyrolysis tests, two bed materials, differing in particle density, were employed: silica sand, for which the sewage sludge particles show a flotsam behavior, and sepiolite, for which the buoyancy effects of the sludge particles are negligible.

The measurement of the mass of the sewage sludge sample remaining in the reactor was employed to determine the reaction rate, which was found to be higher for the fluidized bed tests, as a result of the higher heating rate of fluidized beds in comparison to fixed beds. The reaction rate was slightly higher when the pyrolysis of sewage sludge occurs in a fluidized bed of sepiolite, where no fuel buoyancy effects

appear, than in a bed of silica sand particles, where the flotsam behavior of the sludge particles force them to move close to the bed surface. However, in contrast to silica sand particles that show an inert behavior during the pyrolysis of sewage sludge, sepiolite particles were found to absorb part of the volatile matter released by the sewage sludge during its pyrolysis in the bed.

The pyrolysis time was also determined for each experimental condition, obtaining a significant reduction when the pyrolysis occurred in a fluidized bed compared to the pyrolysis in a fixed bed. A slight reduction of the pyrolysis time also occurred for a fluidized bed conformed by sepiolite and the temperature of the bed was also found to accelerate the pyrolysis process of sewage sludge in all the cases studied. Therefore, to accelerate the pyrolysis reaction of the sewage sludge sample, a velocity of nitrogen high enough to properly fluidize the bed is required, and increasing the bed temperature would also contribute to a faster pyrolysis process. Concerning the effect of sepiolite during the pyrolysis of sewage sludge, further studies are required to analyze the species of volatile matter absorbed, and the effect of this absorption in the global pyrolysis efficiency.

#### ACKNOWLEDGMENT

The authors gratefully acknowledge the financial support provided by Fundación Iberdrola under the program “Programa de Ayudas a la Investigación en Energía y Medioambiente”.

#### REFERENCES

- [1] A. Demirbas, M. F. Demirbas, *Importance of algae oil as a source of biodiesel*. Energy Conversion and Management, vol. 52, pp. 163–170, 2011.
- [2] I. Fonts, M. Azuara, G. Gea, M. B. J. Murillo, *Study of the pyrolysis liquids obtained from different sewage sludge*. Journal of Analytical and Applied Pyrolysis, vol. 85, pp. 184–91, 2009.
- [3] P. Manara, A. Zabaniotou, *Towards sewage sludge based biofuels via thermochemical conversion – A review*. Renewable and Sustainable Energy Reviews, vol. 16, pp. 2566–2582, 2012.
- [4] W. Rulkens, *Sewage sludge as a biomass resource for the production of energy: overview and assessment of the various options*. Energy & Fuels, vol. 22, pp. 9–15, 2008.
- [5] D. Fytilli, A. Zabaniotou, *Utilization of sewage sludge in EU application of old and new methods – A review*. Renewable and Sustainable Energy Reviews, vol. 12, pp. 116–40, 2008.
- [6] H. Fan, H. Zhou, J. Wang, *Pyrolysis of municipal sewage sludges in a slowly heating and gas sweeping fixed-bed reactor*. Energy Conversion and Management, vol. 88, pp. 1151–1158, 2014.
- [7] G. Liu, H. Song, J. Wua, *Thermogravimetric study and kinetic analysis of dried industrial sludge pyrolysis*. Waste Management, vol. 41, pp. 128–133, 2015.
- [8] B. Leckner, *Developments in fluidized bed conversion of solid fuels*. Thermal Science, vol. 20, pp. S1–S18, 2016.
- [9] B. Leckner, *Fluidized bed combustion: Mixing and pollutant limitation*. Progress in Energy Combustion Science, vol. 24, pp. 31–61, 1998.
- [10] A. Gómez-Barea, B. Leckner, *Modeling of biomass gasification in fluidized beds*. Progress in Energy and Combustion Science, vol. 36, pp. 444–509, 2010.
- [11] I. N. S. Winaya, T. Shimizu, D. Yamada, *A new method to evaluate horizontal solid dispersion in a bubbling fluidized bed*. Powder Technology, vol. 178, pp. 173–178, 2007.
- [12] G. M. Rios, K. Dang Tran, H. Masson, *Free object motion in a gas fluidized bed*. Chemical Engineering Communications, vol. 47, pp. 247–272, 1986.
- [13] A. Soria-Verdugo, L. M. García-Gutiérrez, N. García-Hernando, U. Ruiz-Rivas, *Buoyancy effects on objects moving in a bubbling fluidized bed*. Chemical Engineering Science, vol. 66, pp. 2833–2841, 2011.
- [14] M. C. Samolada, A. A. Zabaniotou, *Comparative assessment of municipal sewage sludge incineration, gasification and pyrolysis for a sustainable sludge-to energy management in Greece*. Waste Management, vol. 34, pp. 411–420, 2014.
- [15] E. S. Park, B. S. Kang, J. S. Kim, *Recovery of oils with high calorific value and low contaminant content by pyrolysis of digested and dried sewage sludge containing polymer flocculants*. Energy & Fuels, vol. 22, pp. 1335–1340, 2008.
- [16] H. J. Park, H. S. Heo, Y. K. Park, J. H. Yim, J. K. Jeon, J. Park, C. Ryu, S. S. Kim, *Clean bio-oil production from fast pyrolysis of sewage sludge: Effects of reaction conditions and metal oxide catalysts*. Bioresource Technology, vol. 101, pp. 83–85, 2010.
- [17] R. O. Arazo, D. A. D. Genuino, M. D. G. de Luna, S. C. Capareda, *Bio-oil production from dry sewage sludge by fast pyrolysis in an electrically-heated fluidized bed reactor*. Sustainable Environmental Research, vol. 27, pp. 7–14, 2017.
- [18] Y. Sun, B. S. Jin, Y. J. Huang, W. Zuo, J. Q. Jia, Y. Y. Wang, *Distribution and characteristics of products from pyrolysis of sewage sludge*. Advanced Materials Research, vol. 726, pp. 2885–2893, 2013.
- [19] Y. Sun, B. Jin, W. Wu, W. Zuo, Y. Zhang, Y. Zhang, Y. Huang, *Effects of temperature and composite alumina on pyrolysis of sewage sludge*. Journal of Environmental Sciences, vol. 30, pp. 1–8, 2015.
- [20] P. C. Carman, *Fluid flow through granular beds*. Transactions of the Institute of Chemical Engineers, vol. 15, pp. 150–166, 1937.
- [21] J. Sánchez-Prieto, A. Soria-Verdugo, J. V. Briongos, D. Santana, *The effect of temperature on the distributor design in bubbling fluidized beds*. Powder Technology, vol. 261, pp. 176–184, 2014.
- [22] S. A. Scott, J. S. Dennis, J. F. Davidson, A. N. Hayhurst, *Thermogravimetric measurements of the kinetics of pyrolysis of dried sewage sludge*. Fuel, vol. 85, pp. 1248–53, 2006.
- [23] A. Soria-Verdugo, N. Garcia-Hernando, L. M. Garcia-Gutierrez, U. Ruiz-Rivas, *Analysis of biomass and sewage sludge devolatilization using the distributed activation energy model*. Energy Conversion and Management, vol. 65, pp. 239–244, 2013.
- [24] K. Jayaraman, I. Gökalp, *Pyrolysis, combustion and gasification characteristics of miscanthus and sewage sludge*. Energy Conversion and Management, vol. 89, pp. 83–91, 2015.
- [25] A. Soria-Verdugo, E. Goos, N. Garcia-Hernando, *Effect of the number of TGA curves employed on the biomass pyrolysis kinetics results obtained using the Distributed Activation Energy Model*. Fuel Processing Technology, vol. 134, pp. 360–371, 2015.
- [26] A. Soria-Verdugo, E. Goos, J. Arrieta-Sanagustin, N. Garcia-Hernando, *Modeling of the pyrolysis of biomass under parabolic and exponential temperature increases using the Distributed Activation Energy Model*. Energy Conversion and Management, vol. 118, pp. 223–230, 2016.
- [27] A. Soria-Verdugo, E. Goos, A. Morato-Godino, N. Garcia-Hernando, U. Riedel, *Pyrolysis of biofuels of the future: Sewage sludge and microalgae – Thermogravimetric analysis and modelling of the pyrolysis under different temperature conditions*. Energy Conversion and Management, vol. 138, pp. 261–272, 2016.
- [28] A. Soria-Verdugo, A. Morato-Godino, L.M. Garcia-Gutierrez, N. Garcia-Hernando, *Pyrolysis of sewage sludge in a bubbling fluidized bed: determination of the reaction rate*. In 12<sup>th</sup> International Conference on Fluidized Bed Technology, Krakow (Poland), May 2017.