EXPERIMENTAL INVESTIGATION OF OLIVE TREE PRUNING GASIFICATION IN A BENCH-SCALE FLUIDIZED BED

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ABSTRACT: Olive tree pruning was gasified in a laboratory fluidized bed reactor using air as gasifying agent. Different operating temperatures (800, 850 and 900 ºC) and equivalence ratios (ER) (0.12-0.35) were tested and additional tests were performed varying the particle size of the fuel, the biomass feed rate and the oxygen content in the air (13-40%). The composition of the product gas was determined measuring light gas components (CO2, CO, CH4, H2, C2H4, C2H2, C3, N2), tar, inorganic contaminants (NH3, HCN, H2S, HCl), and water. Solid samples collected from the cyclone and the fluidized bed were also examined. It was found that for low temperature (800 ºC) the gasification efficiency was low (0.58) due to the low char conversion. On the contrary at 900 ºC, the gasification efficiency was above 70% even for the highest ER tested (0.35). The quality of the gas was significantly improved by using O2-enriched air up to 40%.
Keywords: Gasification, biomass, Agricultural residues, Fluidized bed

1 INTRODUCTION

Olive tree pruning (OTP) is an important biomass resource in the Mediterranean countries. Despite having a heating value similar to wood, OTP is still mass-burnt in situ in many farms due to the high costs of collecting and transporting it to large-scale utility plants. A great effort has been made to minimize cost of collection/transportation but feasible solutions are still missing.

Autothermal air-blown moving or fluidized bed gasification (FBG) are relatively simple technologies that could produce high electricity efficiency at small to medium scale by burning the gas, previously cleaned of particles, tars and inorganics, in an internal combustion engine.

Downdraft moving bed gasifiers are difficult to control for heterogeneous fuels like OTP, containing particles with different sizes and density (branches and leaves), whereas fluidized bed gasification allows flexible operation for this type of fuel. High gasification temperatures are required to produce a gas with low tar content and to achieve high fuel conversion [1], but the operating temperature in an FBG is limited by the melting of the ash.

Gasification of OTP in air has been studied in fixed bed [2,3]. However, data regarding the gasification of OTP in FB are lacking.

Gasification of different types of biomass and wastes has been studied in fluidized bed (FB). A great deal of experimental data has been obtained using air in lab or pilot gasifiers under allothermal conditions [4-7], i.e. providing heat to the gasifier during the operation, whereas by preheating the air or using a furnace surrounding the gasifier, to maintain the operation temperature in small reactors. Some studies have tested the effect of O2-enrichment in the air [8-10].

The importance of carrying out tests at the pilot scale adiabatically to mimic large-scale operation has been highlighted [4,11]. Therefore, some studies have tried to conduct autothermal gasification with air but in most of cases, the small size of the reactors made it impossible to operate the tests adiabatically.

In this work, the gasification of OTP has been carried out allothermically in a bench-scale FBG varying the temperature, ER, fuel particles size, biomass flowrate and oxygen content in the air. The species measured include light gas components, different tar compounds, inorganic contaminants and water. In addition, solid samples taken from the cyclone and from the fluidized bed were analyzed.

2 MATERIAL AND METHODS

2.1 Experimental setup

The experimental setup is represented in Fig. 1. The reactor is made of stainless steel. It has three sections: a preheating zone, a bottom bed that is 200 mm high and has an inner diameter of 51 mm and a 250 mm high freeboar with an internal diameter of 82 mm. It is surrounded by a 10 kW electric oven, with two independent heating zones, one for the bottom bed and one for the freeboard, and is equipped with 4 thermocouples and two PID controllers, allowing control of temperature in both zones.

The biomass feeder consists of a fuel hopper, a feed screw and a fast rotating screw that introduces the fuel into the lower part of the fluidized bed. The fuel feed rate was adjusted by the rotation speed of the feed screw. The fuel hopper was pressurized to avoid backflow of hot gases from the reactor.

The gas feeding system allows feeding mixtures of air, N2 and O2 with adjustable composition by using three mass flow controllers. At the exit of the reactor there is a cyclone for collecting entrained particles. After the cyclone the gas passes through three scrubbers and a condenser operating at -20 ºC to eliminate tars and other contaminants before the gas analyzer.

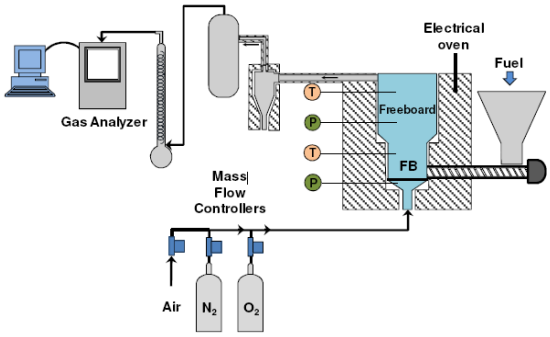

Figure 1: Experimental setup

2.2 Material

The fuel employed was OTP, which is a heterogeneous fuel containing both branches and leale from olive trees. The proximate and elemental composition of the OTP employed is given in Table I and the composition of the main species in the OTP ash is shown in Table 2. Since the fuel contains a large fraction of small branches and leaves, the ash content is higher than values typically found for wood chips or wood pellets that have an ash content of up to 1%.

To enable stable operation of the biomass feeder the OTP was ground to a size below 4 mm. Particles with size below 0.5 mm were eliminated, to avoid excessive entrainment of fines from the bed. Most tests were carried out using particles of 0.5-4 mm, but also some runs were conducted using particles of size 2.5-4 mm.

The bed material employed was bauxite with a particle size between 250 and 500 μm and an experimental minimum fluidizing velocity and particle density of 0.20 m/s and 3200 kg/m3, respectively.

Table I: Proximate and elemental composition of olive tree pruning

		Analysis method
LHV (MJ/kg) (dry basis)	18.37	UNE-164001
Moisture (% as received)	6.72	CEN/TS 14774-1
% weight, dry basis		
Ash	4.54	CEN/TS 14775
Fixed Carbon	15.86	*
Volatiles	79.60	CEN/TS 15148
C	48.84	CEN/TS 11510
H	6.40	CEN/TS 11510
N	1.07	CEN/TS 11510
O	39.15	*

2.3 Operating conditions

The gross of tests were carried out using air as fluidization agent, a biomass flowrate of 1 kg/h and particle size distribution of 0.5-4 mm. The temperature and ER were varied (independently) in the range of 800-900 ºC and 0.17-0.35, respectively, by adjusting the airflow rate and temperature of the test. Then the fluidization velocity (calculated as the fed air flowrate divided by bed section at temperature and pressure of the bed) was varied in the range of 0.56-0.90 m/s. In the following these experiments will be referred to as base case experiments.

Table II: Analysis of OTP ash

Species	wt% in ash
Fe_2O_3	0.90
MnO_2	0.64
MgO	3.16
K_2O	11.54
CaO	38.15
Na_2O	0.14
SO_3	0
P_2O_5	2.65
Al_2O_3	1.79
SiO_2	26.38
TiO_2	0

Some additional tests were conducted varying the following parameters with respect to the base case (keeping the rest of parameters equal): (i) biomass feed rate of 1.5 kg/h; (ii) particle size of 2.5-4 mm; (iii) volume fraction of O2 in the mixture of 13% and 40%. Just one experiment was carried out using an O2 concentration in the fluidizing gas of 13% and this was employed to enable operation with low ER (0.12), but maintaining the gas velocity in the bed similar to the other tests without varying the fuel feedrate.

In Table II, all the tests are listed as well as their operating conditions and most important results.

2.4 Analysis Method

The composition of the exit gas was measured on line with a Siemens analyzer using a non-dispersed infrared method for CO, CO2 and CH4 and thermal conductivity and paramagnetic methods for H2 and O2, respectively. In addition, gas samples were taken with intervals of approximately 5 min, to be analyzed by a micro GC measuring CO, CO2, CH4, H2, N2, O2, C2H4, C2H2, C2H6 and C3. The concentration of C3 is the sum of the concentrations of C3H6 and C3H8.

For analysis of tars, water and inorganic contaminants, gas samples were taken from a port situated just downstream of the cyclone, using a vacuum pump. The sampling port was heated to approximately 315 ºC to avoid condensation of tars and was equipe with a candle filter to eliminate solid particles. For tar and water sampling, the gas passed through a series of impingers filled with isopropanol at -20 ºC. The water content was determined using Karl-Fischer analysis. The gravimetric tar was determined by evaporating the solvent of the samples collected. The individual tar components were measured using gas chromatography and mass spectrometry (GC-MS). A total of 29 aromatic tar species were measured, including compounds with non or low polarity, from light species like toluene or phenol to 5-ring aromatics like perylene.

To study its composition, tar was divided into different classes according to van Paasen and Kiel (2004) [4]. The compounds measured by GC-MS are divided into tar classes 2, 3, 4 and 5 and in addition a GC-MS undetectable tar fraction was estimated by subtracting the yields of aromatic tars measured by GC-MS analysis from the gravimetric tar (toluene and xylene were not considered to be present in the gravimetric tar, as discussed above). The yield of GC-MS undetectable tar is just an approximation, since it is not known exactly to

what extent light aromatics are present in the gravimetric tar sample, or evaporated together with the solvent [4].

For sampling of inorganic contaminants, the gas passed through a series of impingers kept at 0ºC containing different aqueous solutions. H2SO4 solution was employed for capturing NH3, NaOH for HCN and H2S, and distilled water for HCl.

3 RESULTS AND DISCUSSION

3.1 Gas composition

For ER between 0.24 and 0.35 and temperature between 800 and 900 ºC, the measured gas concentrations for H2, CO, CO2, and CH4 were 8.75-14.37%, 11.17-19.42%, 13.41-16.69% and 3.21-4.22%, respectively. As expected, the concentrations of CO, H2 and CH4 decreased with increasing ER (at constant temperature) and increase with increasing temperature, while the tendencies for CO2 and N2 are the opposite.

The concentrations of C2; C2H4, C2H2, C2H6 and C3 hydrocarbons in the gas were also measured. Of these compounds C2H4 is by far the most abundant. The concentrations ranged between 1.4 and 1.8%, 0.05 and 0.11%, 0.04 and 0.11% and 0.02 and 0.24%, for C2H4, C2H2, C2H6 and C3, respectively. The concentrations of C2H6 and C3 decreased with increasing temperature, which is consistent with the conversion of higher hydrocarbons into lighter ones at high temperatures. The variations in the concentrations of C2H4 and C2H2 were smaller, in relative terms. The benzene concentration ranged between 6 and 27 g/Nm3 and as expected it increased with temperature especially between 800 and 850 ºC and at constant temperature it decreases with increasing ER.

The gravimetric tar was high at 800 ºC for low ER (0.24) and decreased rapidly with temperature and ER, leveling off at around 9 g/Nm3.

The contents of inorganic contaminants like NH3, HCN and HCl were also measured, giving maximum concentrations of 0.4%, 0.04% and 0.0004% for NH3, HCN and HCl, respectively. Samples were also taken for measuring H2S, but the concentrations were below the detection limit. The NH3 and HCl concentrations measured here are of the same order of magnitude as results presented by van der Drift et al. [12].

The gas composition (under similar operating conditions) is expected to depend to a large extent on the composition of the biomass, both its contents in cellulose, hemicelluloses and lignin as well as its moisture content and the composition and amount of ash. For example [4] measured different gas compositions for the gasification of beech wood and willow. The gas compositions measured here are quite similar to results obtained previously with wood chips [5,13], while significant differences have been found when comparing to results obtained with wood pellets. The CO, H2 and CH4 concentrations measured here are lower compared to results obtained for wood pellets [6,7]. This discrepancy is probably related to the differences in fuel composition, since the wood pellets had much lower ash content and higher oxygen concentration compared to OTP. In addition, elutriation of char from the bed is expected to occur to a much smaller extent when processing pellets.

The yields of the main light gas components are represented as a function of ER in figs 2-4. Fig 2 shows the yields of CO and CO2, figure 3 shows the H2, CH4 and C2H4 yields and figure 4 gives the yields of C2H2, C2H6 and C3. Figure 2 also shows the total gas yield (N2 and moisture free) in Nm3/kg of dry fuel. ER has the most significant effect on the yield of CO2 (see Fig. 2), which increases due to higher fuel combustion. The yields of CO (see Fig. 2) and H2 (see Fig. 3) decrease slightly with increasing ER, while the CH4 (see Fig. 3) yield remains practically unchanged.

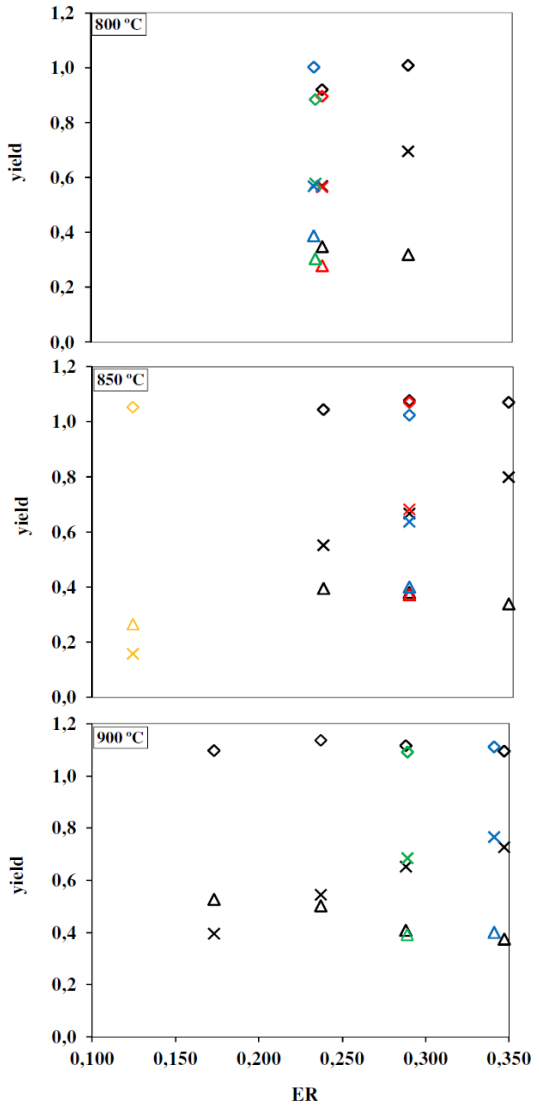

Figure 2: Total gas yield (dry) (Nm/kg of dry fuel) (◊) and yields (g/g of dry fuel) of CO (Δ) and CO2 (×) as a function of ER for different temperatures: 800, 850 and 900 ºC. Black symbols are employed for the base case tests, while colored symbols are employed for tests with fuel particle size of 2.5-4 mm (red symbols), fuel feedrate of 1.5 kg/h (green symbols), with O2-enriched air with 40% O2 (blue symbols) and with N2-diluted air with 13% O2 (orange symbols).

This result is explained by the lower reactivity of CH4 as compared to CO and H2. C2H4 (see Fig. 3) seems to increases with ER, reaching a maximum at ER=0.26 and then slightly decreasing. For a given ER, the yields of CO, H2, CH4 and C2H4 increase with increasing temperature while the CO2 yield decreases. The yields of C2H6 and C3 drop rapidly with

temperature, while the variations of C2H2 are less significant (see Fig. 4). The variations in ER have smaller effect on the yields of C2H6, C2H2 and C3 compared to temperature. The C3 yield seems to increase slightly with increasing ER, while the C2H2 and C2H6 remain practically unchanged. There are no remarkable effects of the fuel particle size or biomass feedrate on the yields of the light gas components, but increasing the particle size seems to lead to a slightly lower methane yield, while the increase in fuel feedrate seems to give somewhat higher C3 yield. The use of O2-enriches air led to a significant increase in the yields of CH4 and C2 and C3 hydrocarbons.

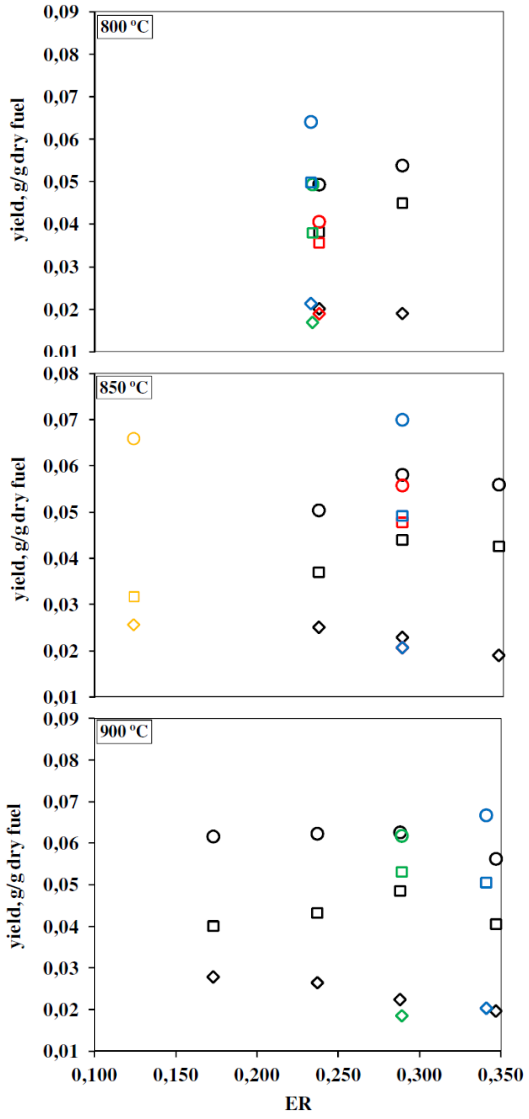

Figure 3: Yields (g/g of dry fuel) of H2 (◊), CH4 (○) and C2H4 (□) of the light gas as a function of ER for different temperatures: 800, 850 and 900 ºC. Black symbols are employed for the base case tests, while colored symbols represent tests with fuel particle size of 2.5-4 mm (red symbols), fuel feedrate of 1.5 kg/h (green symbols), with O2-enriched air with 40% O2 (blue symbols) and with N2diluted air with 13% O2 (orange symbols).

The total gas yield (N2 and moisture free) (see Fig. 2) increased with temperature. At 800 and 850 ºC the gas yield increased when ER is raised from 0.24 to 0.29, but at higher temperature no significant effect of ER on the gas yield was observed. An increase in ER can on one hand favor the conversion of char into gas, but on the other hand it can increase the elutriation of char from the bed due to the increase in gas velocity. The values of total gas yield reported here are similar to results previously obtained with woody biomass [7].

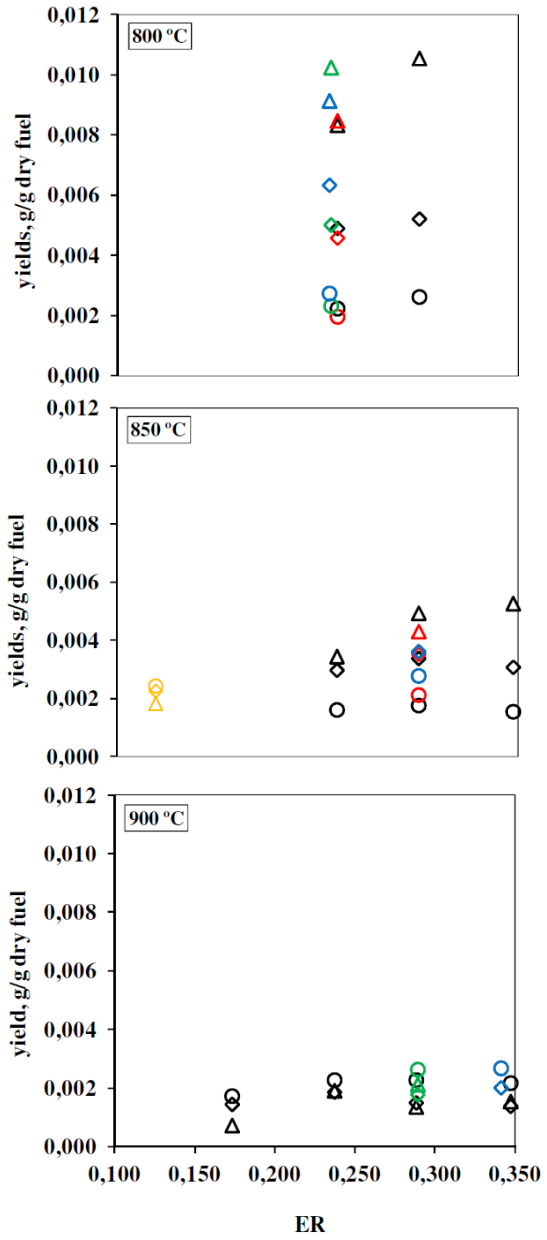

Figure 4: Yields of C2H6 (◊), C2H2 (○) and C3 (Δ) as a function of ER for the three temperatures tested, 800, 850 and 900 °. Black symbols are employed for the base case tests, while colored symbols are employed for tests with fuel particle size of 2.5-4 mm (red symbols), fuel feedrate of 1.5 kg/h (green symbols), with O2-enriched air with 40% O2 (blue symbols) and with N2-diluted air with 13% O2 (orange symbols).

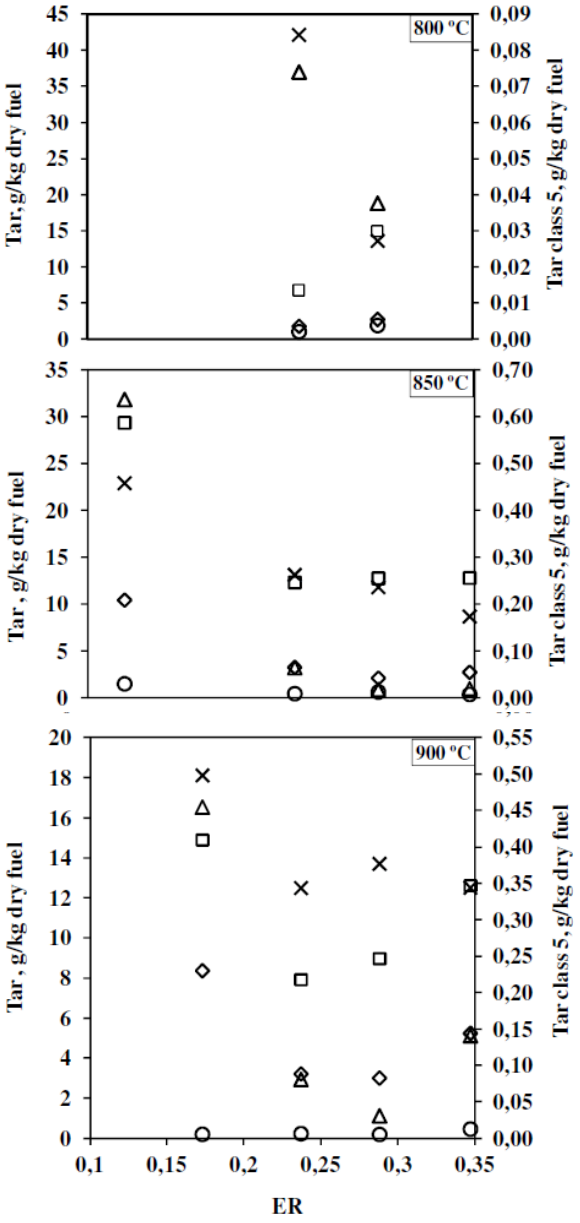

Figure 5: Yields of TC2 (○), TC3 (□), TC4 (◊) and TC5 (Δ) and GC-MS undetectable tar (×), in g/kg of dry fuel, as a function of ER for three temperatures: 800, 850 and 900ºC. Note that the yields of class 5 tar are read on the right side vertical axis and that the scale of the vertical axis are different for the three figures representing different temperatures.

Figure 5 represents the yields of tar classes 2, 3, 4 and 5 as a function of ER for the three temperatures studied. The figure also includes the yields of GC-MS undetectable tar. The most abundant tar class 2 compound at 800 ºC and 850 ºC is phenol but at 900 ºC it is benzonitrile. Toluene is the major tar class 3 compound followed by styrene and Indene. Naphthalene is by far the most abundant tar class 4 compound, while pyrene and chrysene are the major tar class 5 species. At low temperature (800 ºC) the GC-MS undetectable tar is expected to contain mostly primary and secondary tar compounds [14], with high polarity, while at higher temperature it is assumed to contain a larger fraction of more heavy PAH compounds (that are too heavy to be detected in GC-MS analysis) [4].

As expected [15], the yield of TC2 decreases rapidly with increasing temperature, while TC4 and TC5 increase. TC3 decreases as the temperature is raised from 850 to 900 ºC. All the tars tend to decrease with increasing ER, especially the TC5, as can be observed by comparing the results at 850 ºC and ER=0.12 to the rest of the experiments carried out at that temperature. The influence of ER is less clear at high temperature and high ER. An increase in ER increases the concentration of oxygen available, but it also decreases the residence time of the volatiles in the reactor, as the total gas flow increases.

The tar concentrations in the gas measured here differ from those reported in [4], being the contents of TC4 and TC5 significantly lower in the present work. These differences are probably related to the different volatiles residence times in the reactor, since the residence time in the present work was below 0.8 s, while it was between 1.3 and 4 s in [4]. Van Paasen and Kiel (2004) found that rising the gas (volatiles) residence time from 1.3 s to 4 s [4], increased the content of TC4 and TC5 more than 50% and 250%, respectively, while it decreased the TC2 and, to a smaller extent, TC3.

The yield of water for the different tests is given in table 2. The water yield varies between 0.04 and 0.15 g/g of dry fuel. It is difficult to find clear trends of the water content with the temperature and ER. Water takes part in a large number of reactions, including devolatilization, tar reforming, char gasification water gas shift reaction, etc. and these reactions are affected differently by temperature and oxygen concentration in the reactor.

The LHV of the gas is shown in figure 6. Three different values have been calculated: one that includes only the light gas, one that includes both light gas and benzene and one that also includes tar. A heating value of 36 MJ/kg tar was assumed for the non GC/MS tar. When analyzing the results in Fig. 6 it is clear that an important part of the heating value of the gas is present as tars, especially for low ER. This means that gas clearing methods where tars are eliminated rather than converted into light gas lead to a significant decrease in the heating value of the product gas. It can also be seen in Fig. 6 that the LHV of the gas (including only light gas) is about 60% higher when using O2-enriched air. This is of course related to the lower dilution with N2, but also the higher yields of CH4 and C2 and C3 hydrocarbons affect the LHV.

3.2 Carbon conversion and gasification efficiency

The carbon conversion was defined as the carbon measured in the gas, including light gas and tar divided by the carbon introduced with the fuel. The carbon conversion measured at different temperatures and ER is shown in Fig. 7. The carbon conversion is favored by high temperature especially at low ER. At low temperature (800 and 850 ºC) and for ER below 0.26, an increase in ER gives higher carbon conversion, but for higher temperature and ER variations are small. The carbon conversion is closely related to the total gas yield (see Fig. 2) ant it follows the same trends. The carbon conversion achieved in this work is relatively high, since no material is extracted from the bed during the operation. Large scale units on the other hand would need some bed extraction to avoid accumulation of ashes.

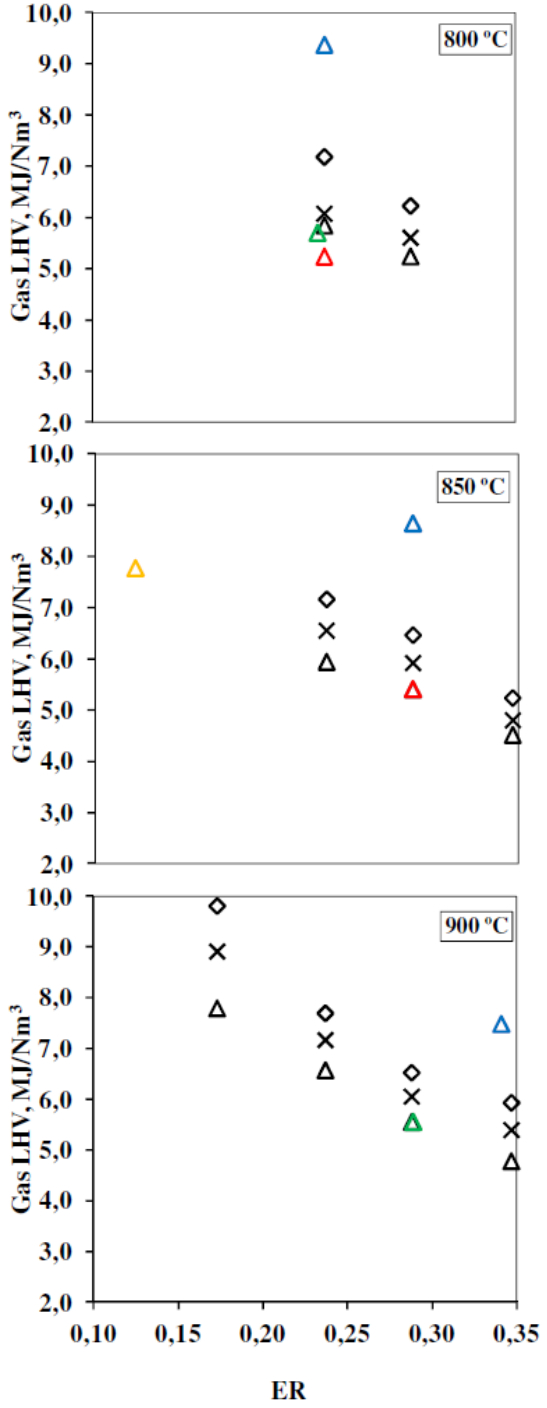

Figure 6: LHV of the gas (MJ/Nm3), including: (Δ) only the light gas; (×) light gas and benzene; (◊) light gas, benzene and tars. Black symbols are employed for the base case tests, while colored symbols are employed for tests with fuel particle size of 2.5-4 mm (red symbols), fuel feedrate of 1.5 kg/h (green symbols), with O2-enriched air with 40% O2 (blue symbols) and with N2-diluted air with 13% O2 (orange symbols).

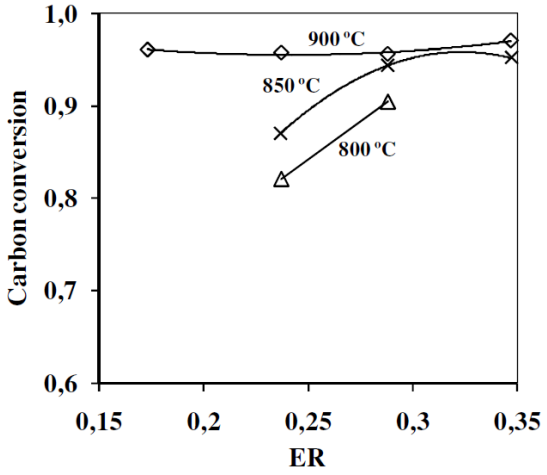

Figure 7: Carbon conversion for the base case tests as a function of ER for three different temperatures: 800 ºC (Δ), 850 ºC (×) and 900 ºC (◊).

The gas concentrations measured during the tests were stable once a start-up period of less than 30 minutes had passed, so it can be assumed that the operation was stable and without accumulation of char in the bed. Only at low temperature (800 ºC) and low ER (0.24) the H2 concentration in the gas was observed to increase slightly during the experiments, so it is suspected that some char accumulation occurred during these tests. In addition the closure of the carbon balance was good except for the tests at 800ºC and ER of 0.24. It was also observed that under the mentioned conditions the mass of carbon in the bed was much higher compared to the other tests. By visual inspection of the material extracted from the bed it has also been observed that the average size of the char particles was significantly higher at low temperature. The rate of particle elutriation is favored by high gas velocity, but it is also affected by the mass of solids in the bed. For example for the experiment carried out at 800 ºC with ER of 0.24, both the rate of particle elutriation and the carbon fraction in the elutriated solids are high, which is logical since for these experiments the mass of carbon in the bed was the highest. It is also noteworthy that increasing the fuel particle size from 0.5-4 mm to 2.5-4 mm (i.e. eliminating the finest particles) lead to significantly lower char elutriation. The increase in fuel feedrate to 1.5 kg/h led to a higher gas velocity in the reactor and to higher rate particle elutriation per g of fuel fed. Also the mass of carbon in the bed was slightly higher during these tests. For the experiments with O2-enriched air, since the gas velocity was lower, so was the rate of particle entrainment.

Two gasification efficiencies were defined, the cold gas efficiency (CGE), to represent the case when the gas is cooled down before its final application, as for example when being burnt in an internal combustion engine, and the hot gas efficiency (HGE) for applications where the gas is not cooled, as when it is burnt directly in a boiler. The CGE and HGE were calculated as:

$$CGE = m_{gas}\, LHV_{gas} / (m_{fuel}\, LHV_{fuel})$$

$$HGE = (m_{gas} LHV_{gas} + H_{gas}) / (m_{fuel}\, LHV_{fuel}),$$

being *mgas* and *mfuel* the mass flow rates of product gas and fuel fed, respectively and *Hgas* the enthalpy of the

gas which was calculated using a reference temperature of 25 ºC. For calculating the CGE the gas was considered to contain benzene but not tars and for calculating the HGE the gas was considered to contain both benzene tars and water. The CGE and HGE for the different temperatures and ER studied are shown in Fig. 8. The CGE is favored by high temperature, all the values obtained for 900 ºC are higher than all the values at lower temperature, even if comparing the CGE at 900ºC and with ER=0.31 to the CGE at 850 ºC and with ER=0.21. At 800 and 850 ºC, the CGE reaches a maximum for ER=0.26, the increase in efficiency between ER of 0.21 and 0.26 is related to the increase in carbon conversion and to the increase of the yields of CH4 and light hydrocarbons and at 800ºC, also the yield of benzene is higher for ER=0.26 On the contrary, at 900ºC the efficiency decreases steadily with ER. For high temperature and low ER, the HGE tends to unity, which indicates that under these conditions the heat supplied by the electrical oven is higher than the heat needed to compensate for the heat losses through the reactor walls, so it would not be possible to achieve these conditions in an autothermal unit.

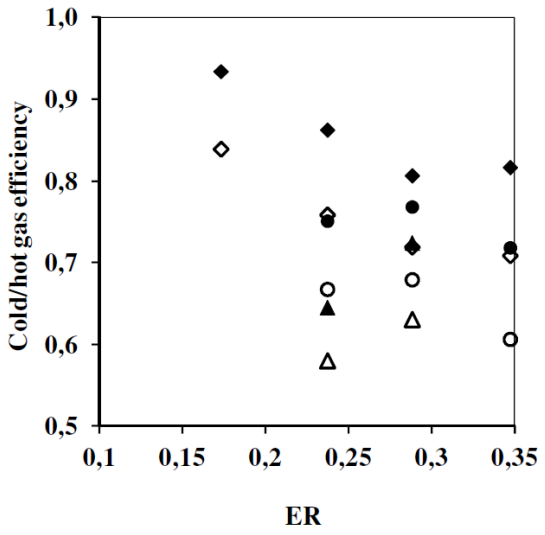

Figure 8: CGE and HGE as a function of ER for the base case experiments at 800 ºC (Δ), 850 ºC (×) and 900 ºC (◊). The CGE is represented by open symbols and the HGE by solid symbols.

4 CONCLUSIONS

The gasification of olive tree pruning, an agricultural residue, has been studied in fluidized bed. The influence of different operating parameters like temperature, ER, fuel particle size, biomass feed rate and O2-enrichment of the air was investigated. Temperatures of 800, 850 and 900 ºC and ER between 0.12 and 0.35 were tested. The composition of the gas at the exit of the gasifier were characterized by measuring the concentrations of light gas components like CO, H2, CO2, CH4, C2, C3, N2; different tar components using GC-MS and gravimetric analysis, as well as water and inorganic contaminants like NH3, HCN and HCl. The results show that at low temperature (800 ºC) and ER (0.24) the char conversion and the gasification efficiency are below 0.54 and 0.78, respectively. As temperature increases both char conversion and cold gas efficiency increase and at 900 ºC the cold gas efficiency was above 0.70 for all ER tested. The experiments were carried out with relatively high gas velocity and therefore the entrainment of particles from the bed was significant. The entrainment of char particles leads to lower char conversion and thus lower gasification efficiency. It was found that rate of particle entrainment could be significantly reduced if the gas velocity was reduced by 20-25% or the fuel particle size was increased from 0.5-4 mm to 2.5-4 mm (i.e. eliminating the finest fraction). It is thus expected that the particle entrainment in real scale gasifier will be less significant than in the laboratory since the particle sizes employed are much larger.

The tar concentration in the gas was found to be too high to burn the gas directly in an internal combustion engine. For the gasification with air, the gravimetric tar content varied between 8 and 30 g/Nm3, but for the majority of the tests it was in the range of 10 g/Nm3. When increasing the temperature and ER, the total tar concentration decreased although the concentrations of heavy tar (tar classes 4 and 5) were favored by high temperatures.

Experiments were carried out using O2-enriched air with an oxygen content of 40% and it was found that in this case a gas with LHV of more than 10 MJ/Nm3 could be achieved.

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6 NOMENCLATURE

6.1 Abbreviations

CGE	Cold gas efficiency
GC-MS	Gas chromatography-mass spectrometry
FB	Fluidized bed
FBG	Fluidized bed gasification or fluidized bed gasifier
OTP	Olive tree pruning