

BIOMASS GASIFICATION: EFFECT OF EQUIVALENCE RATIO ON SYNGAS QUALITY IN THE CASE OF EXTERNALLY HEATED REACTOR

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ABSTRACT: Biomass gasification is a thermal process that aiming at obtaining the possible best quality of the product gas, in terms of heating value and composition. The reaction temperature is one of the most important parameters affecting the syngas quality. Several experiments show a close link between the air mass injected within the reactor (the equivalence ratio) and the temperature reached.

In the proposed paper we investigate the effect of varying the equivalence ratio in the case of an externally heated reactor, on the temperature distribution within the reactor, and on the syngas quality (i.e., composition and low heating value).

Keywords: equivalence ratio, gasification, agricultural residue, biomass.

1 INTRODUCTION

The use of biomass as energy source represents nowadays 10% of the world total primary energy demand (Figure 1), about 87% of which consists of solid biomass [2]. This kind of biomass is mostly used for heat generation and power production.

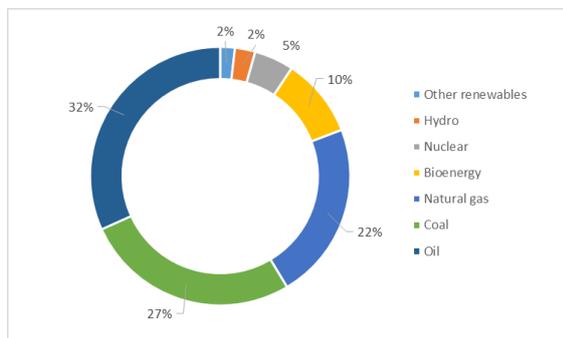


Figure 1: World primary energy demand by source (*data source: WEO-IEA, [1]*).

Gasification is one of the thermochemical conversion processes for solid biomass that has been developing more and more in recent years. It produces a gaseous fuel starting from a solid fuel by a thermal degradation process. The heat required for gasification is usually supplied by partially oxidizing the fuel through an oxidant agent, which can be air, pure oxygen or steam [3]. However, part of this heat could also be supplied from outside the reactor, using renewable sources such as the solar one. In particular, we can provide through a concentrating solar power (CSP) system the energy needed by the endothermic reactions of the gasification process, thus resulting in a better use of the fuel energy content.

Coupled CSP-gasification technology has been proposed many times over the last years. Kaniyal *et al.* [4] coupled CSP and biomass gasification to produce biofuels. Tanaka his coauthors [5] studied a system in which biomass steam gasification produces syngas, which is sent to a gas turbine and the steam, is produced via a CSP plant. Ravaghi-Ardebili *et al.* [6] analyzed an unified process consisting of a CSP plant, which supplies the produced steam to the biomass gasification process as

well as to the downstream conversions to chemical commodities and energy carriers for methanol/dimethyl ether production. Palumbo *et al.* [7], tested an indirectly heated hybrid co-feed system at high temperatures, using also the CSP technology, with biomass, methane, and steam as reactants, to produce H₂-rich syngas. Despite these latter cases, so far, it was often proposed to feed the gasification process by the solar radiation directly concentrated on the reactor surface or inside, via a glazed area and several experimental campaigns were carried out (see for instance [8]-[13]).

In order to couple a CSP with a gasification reactor, it is important first selecting an appropriate reactor. Gasification reactors are classified in three main categories [14]: entrained bed (usually for power larger than 50 MW_{th}), fluidized bed (ranging in 5-100 MW_{th}) and fixed bed (10-10000 kW_{th}). The former category requires a very high temperature (up to 2000 °C), pure oxygen and steam as oxidant agents as well as high-quality fuels, thus it does not seem the best solution for coupling with CSP systems, which cannot reach such high temperature. Fluidized bed reactors have an operating temperature ranging in 700-800 °C along the whole reactor, and it would not be ideal to have them coupled with a CSP plant because it can only provide heat up to a maximum temperature of 550 °C (using molten salts as thermal fluid). On the contrary, fixed bed reactors seems a good option, since they operate at temperatures ranging in 800-1400 °C but with reaction zones well separated. Moreover, they are the most widespread (78% of the total amount of gasification plants in the world) [14]. Gasification in fixed bed reactors consists of three steps, taking place in different zones of the reactor: drying (25-250 °C), pyrolysis (250-600 °C), oxidation and reduction (more than 700 °C). The first two are endothermic reactions thus can be partially or totally heated from outside.

With the general aim of optimizing the coupling between CSP and biomass gasification, here we are interested in studying the gasification process. In a conventional gasification, temperature within the reactor is governed by partial oxidation of the fuel, thus by the amount of oxidant injected. On the contrary, in an externally heated reactor, since the needed temperature may be guaranteed by the external source, one could optimize the amount of oxidant injected within the reactor to improve

the syngas quality. In this framework, and starting from the findings of De Bari *et al.* [15], we performed an experimental campaign to study the effect of the amount of oxidant used in gasification of residual agricultural biomass, on syngas composition.

2 EXPERIMENTAL SETUP

Reactor used for the tests (Figure 2) consists of a stainless steel cylinder (8.3 cm diameter), and a reaction chamber of 59 cm height. The upper part is removable for direct accessing the feeding system. At the bottom, a metal plate works as fixed bed for biomass. Beneath, there is the “wind box”, namely the air (or steam, or N₂) heating system. The insulation consists of glass wool and aluminum. The electric heating system is a Watlow 96 series. Gas analysis was performed through a gas chromatograph VAR-IAN® CP-4900 micro GC and Galaxie

SW. During data processing N₂ sent to the reactor was then subtracted to make all the tests uniform and comparable.

Hazelnut shells were used as fuel. Hazelnut is an important food crop in Italy, especially in central regions. The production is mostly located in Cuneo, Roma, Avellino, Napoli and Messina [16]. Hazelnut shells are a waste byproduct currently used in some cases as fuel for household heating system. However, it could be better used in larger and more efficient plants, i.e. gasification plants, in a circular economy perspective.

Table 1 shows the fuel “proximate” and “elemental” analysis, obtained by thermogravimetry Mettler Toledo®, and an analysis of the elements (CHNO) carried out using a Leco 2000 CHN® analyzer. Fuel LHV was assessed by calorimeter Parr 6200® of Thermo Fisher®. Its value equals 17.23 MJ/kg.

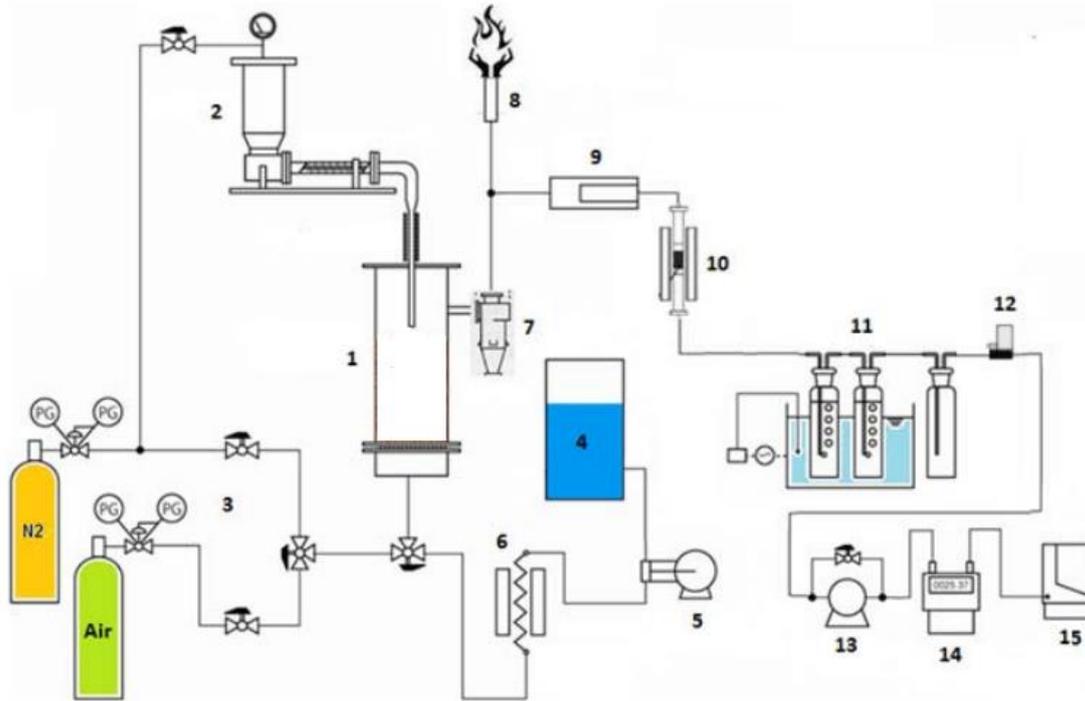


Figure 2: Scheme of the test plant. 1) updraft reactor; 2) feeding system; 3) air and nitrogen feeding systems; 4) steam generating unit; 5) pump for extract the distilled water; 6) water heater; 7) cyclone for particulate removal; 8) torch; 9) high temperature dedusting system; 10) secondary reactor; 11) bubblers; 12) flow control system; 13) vacuum pump; 14) flowmeter; 15) gas-chromatograph.

Table 1: Composition of raw hazelnut shells.

Proximate analysis	wt%	Elemental Analysis	wt%
Humidity	7.90	Carbon	46.65
Ashes	1.16	Hydrogen	5.55
Volatile substances	72.45	Oxygen	38.74

Four different ER values (0.0, 8.0, 15.0, 23.0 and 30.0 %) were tested. Each test took about 40 minutes and for each of them at least eight measurements were performed. Therefore, those presented here are mean results.

3 RESULTS AND DISCUSSION

The amount of oxidant injected in a gasification reac-

tor can be measured by the *equivalence ratio* (ER), that is the ratio between the actual air/fuel ratio and the stoichiometric one, eq. (1).

$$ER = \frac{\left(\frac{m_{air}}{m_{fuel}} \right)_{real}}{\left(\frac{m_{air}}{m_{fuel}} \right)_{stoch}} \quad (1)$$

The effect of ER on syngas quality can be evaluated, for instance, through its composition and heating value, and by computing the *cold gas efficiency* η_g (2). This quantity is the ratio between the energy content of syngas produced and that of the biomass used to produce it; therefore, by definition, η_g is a measure of syngas quality.

$$\eta_g = \frac{LHV_g \cdot V_g}{LHV_b \cdot M_b} \cdot 100\% \quad (2)$$

In equation (2) V and M indicate volume and mass flow rate respectively, LHV is the low heating value, and subscripts g and b indicate syngas and biomass respectively.

In the rest of this section we present the results of the tests performed at Sapienza University of Roma.

Temperature distribution within the reactor is reported in Figure 3. As shown, temperature distribution in the upper part of the reactor is almost independent from ER, while at the bottom it varies in a wide range depending on ER value. In general, temperature at the bottom increases as ER increases, because of the increase of oxygen available for combustion reactions. The large difference between top and bottom temperatures is due to two main effects: first, lower part of the reactor is not electrically heated; hence, there temperature is determined by partial oxidation and possible heat loss. Second, oxidant enters the reactor at 450 °C from the bottom, and thus it cools down the system in that region.

Figure 4 reports volume flow rates of the main syngas components. Hydrocarbons lighter than methane are not measured but they represent at maximum 3% in volume; moreover, tar content is not accounted for since it is entrapped within the bubblers and, in a real application, it should be removed from the syngas stream. As expected, volume flow rates of the main compounds increase as ER increases. This is due to the increasing amount of air entering the reactor with ER, and its interaction with the fuel. By subtracting airflow rate from syngas flow rate it can be estimated the effect of ER on the amount of syngas produced. As reported in Figure 5, it increases with ER. In the case of ER=8.0%, the amount of syngas produced is 31% larger than that of ER=0.0% (pyrolysis test), which is taken as reference. Then, syngas flow rate increases almost linearly with ER, reaching 63% at ER=30%.

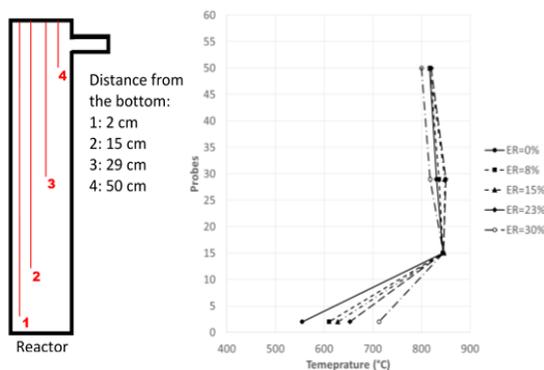


Figure 3: Temperature distribution within the reactor.

To better analyze the effect of ER on syngas composition, we removed the nitrogen content, and considered the sum of the remnant compounds as 100%. Results are shown in Figure 6 and Figure 7. Contents of hydrogen and methane are strictly related: CH_4 reacts with O_2 through two different reactions, (3) and (4), forming CO and H_2O or CO and H_2 respectively. These products directly or indirectly (CO and H_2O can react together through the *shift reaction*, (5), producing CO_2 and H_2) increases the hydrogen content of syngas.

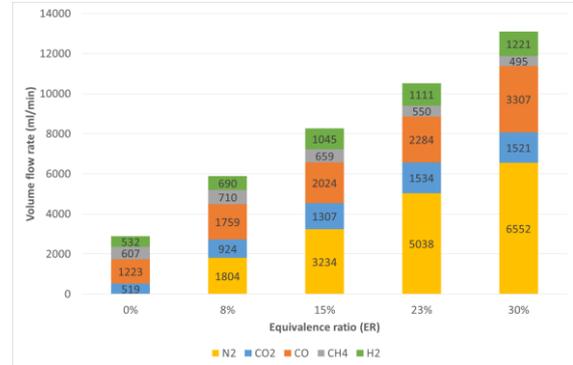
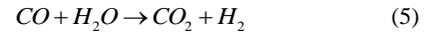
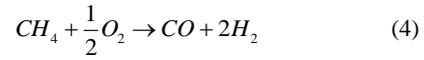
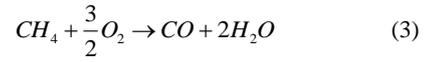


Figure 4: Volume flow rates (in ml/min) of syngas compounds at different ER values.

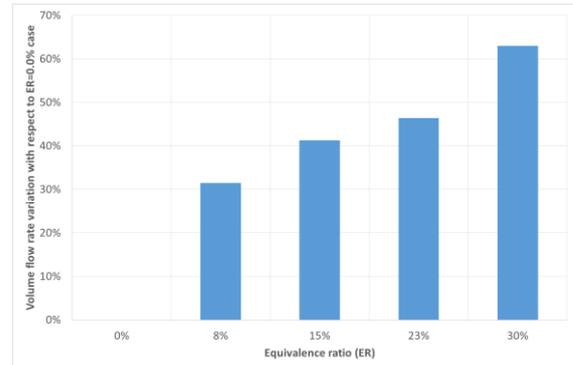


Figure 5: Variation of volume flow rate of syngas at different ER values.

Therefore, as ER increases the amount of methane involved in reactions with oxygen increases, leading to a decrease of CH_4 content with ER. On the contrary, hydrogen content increases with ER because secondary reactions producing H_2 are faster than hydrogen oxidation, thus leading to a net increase of H_2 content with ER. However, at about ER=15% hydrogen oxidation starts to become relevant, hence H_2 content decreases as ER increases.

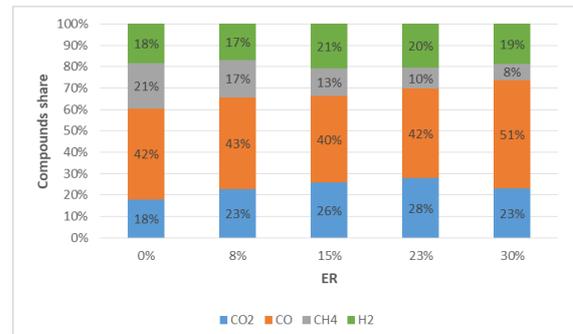


Figure 6: Syngas composition at different ER N_2 free.

CO concentration is almost constant up to ER=23% then it increases. On the contrary, CO_2 increases up to ER=23% and then decreases. This could be due to the

high temperature measured at the bottom of the reactor when ER=30%. Indeed, in this case the temperature ranges in 700-725 °C, and at this temperature endothermic reactions are favored, resulting in a larger content of CO and H₂, and smaller CO₂.

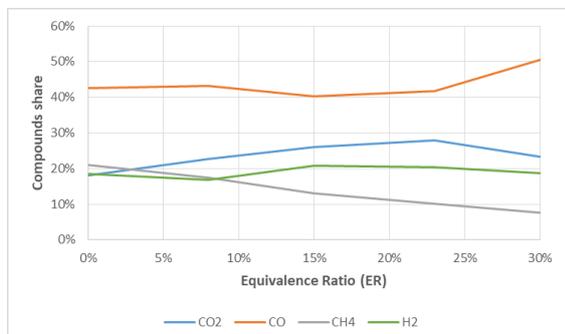


Figure 7: Trend of CO₂, CO, CH₄ and H₂ as a function of ER.

According to Guo *et al.* [17], we computed the low heating value (LHV) of the syngas as

$$LHV = 107.98 \cdot H_2 + 126.36 \cdot CO + 358.18 \cdot CH_4 \quad (6).$$

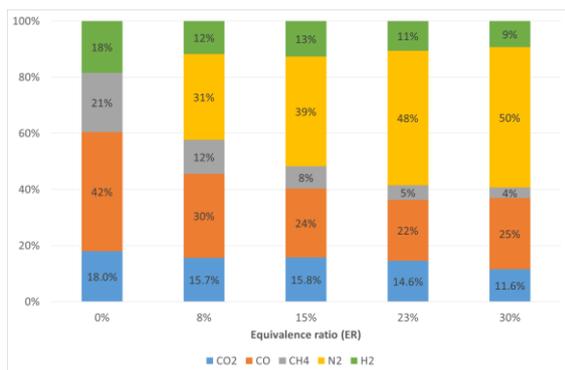


Figure 8: Syngas composition with N₂.

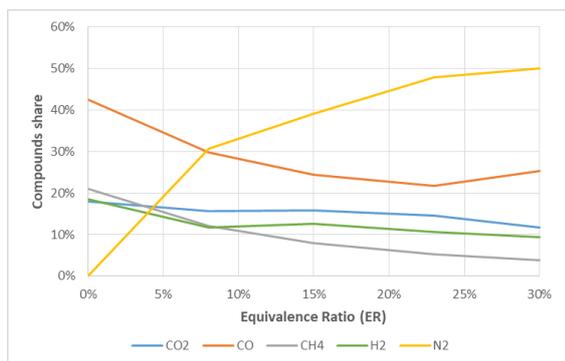


Figure 9: Trend of CO₂, CO, CH₄, N₂ and H₂ as a function of ER.

Since the heating value of a gas mixture should consider the whole composition of the mixture, to evaluate syngas LHV we have to refer to its actual composition, as reported in Figure 8. Since nitrogen is an inert gas, increasing its amount within the syngas decreases the syngas LHV. Therefore, it is obvious expecting a decrease of syngas LHV with ER. However, trying to identify an optimal ER from the syngas quality point of view, the presence and concentration of other components (Figure 8)

should be accounted for. For this reason, beside LHV we compute a nitrogen-free low heating value (LHV*). LHV and LHV* are shown in Figure 10. Both of them decrease as ER increases, but the reduction is more evident in the case of LHV, as expected. In this case indeed, the combination of an increasing content of N₂ with ER, and a reduction of other fuel gas (Figure 9), results in a reduction of the syngas LHV by about 62%. LHV* trend with ER is again decreasing, but reduction is less relevant than that of LHV, showing a decrease of about 25%. Reduction of both LHV and LHV* decreases with ER, and this is due to the increase in CO content when increasing ER from 23 to 30 %, thus balancing the increase of nitrogen mass content within syngas.

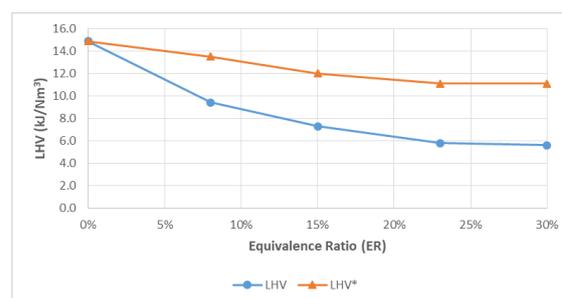


Figure 10: Low heating value of syngas as functions of ER.

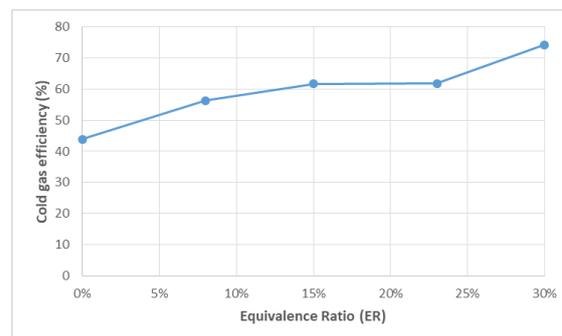


Figure 11: Cold gas efficiency as functions of ER.

Cold gas efficiency as a function of ER is reported in Figure 11. As shown, it increases from about 44% at ER=0% to about 74% at ER=30%. This means that as ER increases, the reduction of syngas quality is compensated by the increase of syngas flow rate.

4 CONCLUSIONS

In this work, we studied the effect of the amount of oxidant entering the reactor on the syngas composition and quality. We have seen that increasing ER the syngas produced increases accordingly, but at the same time its LHV decreases. This is due to the increase of nitrogen content, and to the increasing amount of fuel gases involved in partial oxidation. Progress of combustion with ER is also measured by temperature variation on the reactor bed, ranging from about 555 °C at ER=0% up to 713 °C with ER=30%.

All this aspects can be useful when one wants to use an external heat source, such as solar energy (i.e., a CSP plant), to provide part of the heat required by gasification. Adopting an external heat source allows to optimize ER with the aim of maximizing both the syngas flow rate and its quality (LHV).

5 REFERENCES

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