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Integration of Biomass Gasification and Hot Gas Cleaning Processes

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In the near future, most of the world population will live in the developing countries, and it is plausible that biomass will be one of the main renewable energy sources of the future. Fluidized bed reactors should be the best solution to transform biomasses having different physical properties in energy and chemical vectors through gasification, due to its possibility to operate continuously, at high temperatures, and to utilize catalysts into the reactor and downstream of it, in order to reduce tar, NH_3 etc. in the product gas.
Catalytic filter candles are an innovative solution for hot gas cleaning & conditioning. It has been demonstrated that catalytic ceramic filters inserted in the freeboard of a fluidized bed gasifier, allow the complete removal of particulate by means of their anisotropic porous filtering structure, and furthermore act as catalyst to remove tar and ammonia, thanks to the Ni based catalyst contained into the filter itself (Rapagnà, Gallucci and Foscolo, 2017).
To by-pass the preparation of the catalytic ceramic filters and to render the overall process more feasible in practice, ceramic filters filled with commercial steam reforming catalyst pellets have been located in the freeboard of a 0.1 m ID fluidized bed reactor, where olivine particles act as bed inventory.
Experimental tests were carried out to check the values of the gas pressure drop through the plain filter and through the filter containing catalyst pellets, at different temperatures and gas filtration velocities. It has been found that at the highest temperature and filtration velocity (T 800°C and v 140 m/h) the pressure drops are around 48 mbar, while at the operating conditions (T 800°C and v 100 m/h) the total pressure drops are only 35 mbar. Furthermore it was observed that there is an almost negligible difference between the pressure drops in the case of empty and filled candle; it was thus deduced that the catalyst pellets do not cause relevant additional losses compared to the empty filter. The trends of pressure drops were fitted with the empirical Darcy-Forchheimer relation and with the Ergun equation, and it was found that the major contribution is given by the viscous term of the relation. Furthermore an additional contribution of concentrated pressure drops was identified and ascribed to the outer Al_2O_3 membrane of the candle characterized by very fine pores.

1. Introduction

In the next quarter of century it has been estimated that the energy consumption will increase of 28 % due to population growth and the rise in living standards of the people of the developing countries (EIA, 2017). With the present energy production system, it will be difficult to maintain the current emission levels of greenhouse gases, and it is not plausible to expect a CO_2 reduction of 50% of their current values by 2050 needed to limit the predictable temperature rise of 2 °C. Since the increase of energy consumption will take place mainly in the developing countries, it is necessary to increase the use of biomass as a renewable energy source, also because their current contribute to the whole energy production is significant. Since biomass is a resource diluted throughout the territory, it is necessary to consider small processing plants, widespread throughout the territory in order to reduce biomass transport costs. This solution makes possible the use of the total resulting heat for district heating, enhancing the contribution of the renewable sources to the whole energy requirement of the territory itself. The technology of the biomass transformation plants should be flexible in order to utilize all the biomass present in the territory, which may have different size, humidity and composition. The plant

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should operate at ambient pressure, to be easy to manage, and must respect the increasingly stringent environmental regulations. The CO_2 emitted by the biomass transformation plants, can be drastically reduced, by using new technologies such as carbon capture and storage (CCS). These last technologies use Mg and Ca sorbent solid particles that are transformed in carbonates in the Carbonator reactor at temperature ranging from 600 to 700 °C. These carbonated particles are transported in a Calciner reactor where CO_2 is released at temperature >900 °C (Zhenissova *et al.*, 2015). These solid particles, being porous, are subject to fragmentation during their transport between the Carbonator and the Calciner reactors, with consequent formation of fines.

Fluidized bed reactor should be the best solution to transform biomasses having different physical properties in energy and chemical vectors through gasification. With this type of gasifier it is possible to operate continuously at high temperatures, to utilize catalysts into the reactor and downstream of it, in order to reduce tar, NH_3 etc. in the product gas, and to permit the Carbon Capture and Storage CCS by using appropriate solid materials as a bed inventory.

In order to meet the increasingly stringent environmental regulations, and increase the efficiency of biomass transformation in useful products, such as synthesis gas and/or hydrogen, it is necessary to carry out conversions and cleaning of the product gas at a high temperature

The elimination of dust contained in the gas is therefore imperative before subjecting it to subsequent processing such as the transformation of high molecular weight hydrocarbons and methane in light gases.

Catalytic filter candles are an innovative solution for hot gas cleaning & conditioning. The powder contained in the gas adheres to the outer surface of the ceramic candle. As previously reported (Milazzo *et al.*, 2013), excessive solid deposits on the surface of the filtration unit can cause clogging or damage of the filter itself, in addition to an increase of the gas pressure drop. It is thus necessary to remove this layer of powder periodically (each 20-30 minutes), by feeding a pulse of clean gas (pre-heated nitrogen) from the inside of the ceramic candle, known as back pulsing system (Heidenreich and Foscolo, 2015).

It has been demonstrated that catalytic ceramic filters inserted in the freeboard of a fluidized bed gasifier, allow the complete removal of particulate by means of their anisotropic porous filtering structure, and furthermore act as catalyst to remove tar and ammonia, thanks to the Ni based catalyst contained into the filter itself (Rapagnà and Spinelli, 2015).

To by-pass the preparation of the catalytic ceramic filters and to render the overall process more feasible in practice, ceramic filter filled with commercial naphtha steam reforming catalyst has been located in the freeboard of a 0.1 m ID fluidized bed reactor, where olivine particles act as bed inventory.

Air pressure drop through the empty and filled ceramic filter were measured and an empirical equation has been used to fit the experimental data.

2. Experimental

The measurements were carried out in an experimental apparatus composed of a bubbling fluidized bed gasifier with an internal diameter of 0.10 m and 0.85 m of height, externally heated with a 6 kW electric furnace (Rapagna *et al.*, 2014). The bed material consists of 3 kg of olivine with an average particle diameter of 360 µm and density 3000 kg/m^3 (Rapagnà *et al.*, 2017). The upper part of the reactor was modified in order to insert the ceramic candle inside the freeboard; the ceramic filter, provided by PALL Schumacher GmbH, has an external diameter of 60 mm, an internal diameter of 40 mm, and a total filtration length of 440 mm (see Figure 1a). The ceramic candle has an Al_2O_3 based asymmetric structure, with new improved candle support type (UHT) with coarse pores and a fine Al_2O_3 outer membrane layer on top of the support materials to collect small particles due to its fine pores.

The measurements of pressure drops through the candle were carried out for different values of temperature and filtration velocity. The pressure drops were investigated through the empty candle and through the candle partially filled with commercial naphtha reforming catalyst pellets, in order to study, besides the plain ceramic filter, also the configuration of the candle made catalytic, for the simultaneous abatement of particulate and tar from the raw syngas (Rapagnà *et al.*, 2010). For the measurements through the candle filled with catalyst, the cavity of the filter (diameter 40 mm) was partially filled with the catalyst pellets, leaving a smaller cavity of diameter 20 mm for the gas to flow towards the exit of the candle. The catalyst pellets are kept in the peripheral zone of the cavity by means of a steel tube with several slits to ensure the passage of the gas (see Figure 1b).

The temperature was measured by means of two thermocouples placed in the olivine bed (T1) and in the upper part of the candle (T2); the operating temperature of the candle was calculated as average of the values measured in T1 and T2.

The pressure drops through the candle were calculated by the difference of the pressure values measured in the freeboard of the reactor and at the outlet of the candle, respectively.

The tests were carried out feeding air as fluidization medium from the bottom of the reactor and measuring the pressure drops through the ceramic candle. The air flow, between 15 and 50 Nml/min, was measured and controlled by means of a mass flow controller, in order to obtain the desired filtration velocities through the ceramic candle, between 20 and 140 m/h (0.005 and 0.039 m/s) depending also on the operating temperature.

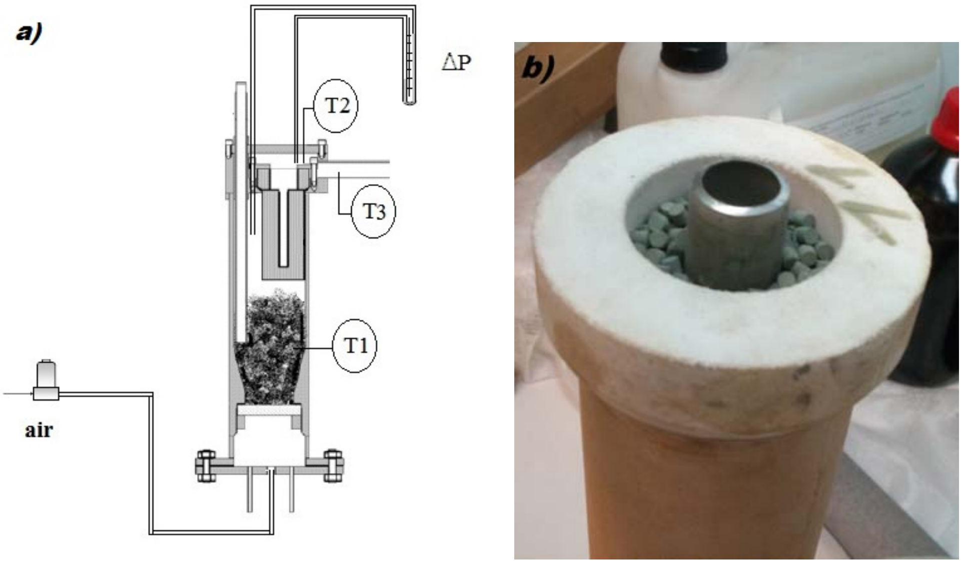


Figure 1: a) Experimental apparatus used for the measurement of the pressure drops through the candle; b) Candle partially filled with catalyst pellets

3. Results and discussion

The pressure drops for different values of filtration velocity measured at 800°C through the empty candle (EC) and through the candle filled with catalyst (CC) are shown in Figure 2.

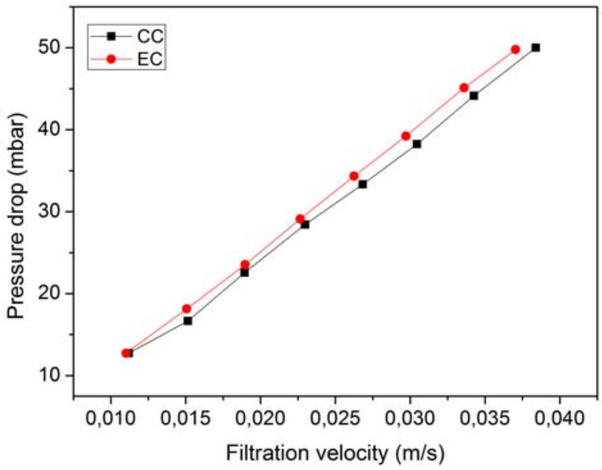
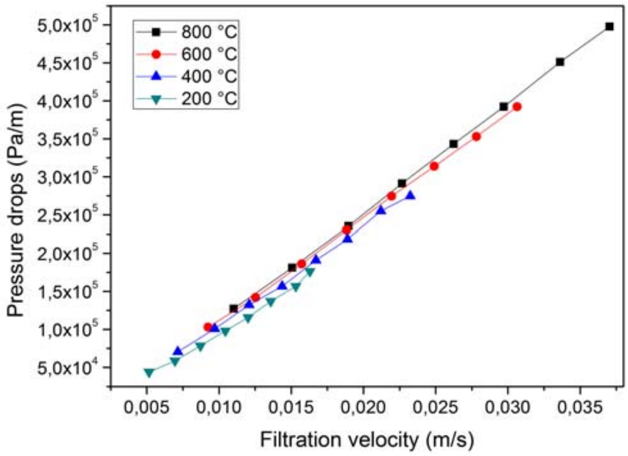


Figure 2: Pressure drops through the empty candle (EC) and the candle filled with catalyst (CC) measured at 800°C

From the diagram it is possible to observe that the pressure drops in the two configurations EC and CC are almost identical; the maximum difference between the pressure drops measured at the same velocity in the two configurations is lower than 2% and could be due to intrinsic measurement errors of the instruments.

The contribution of the catalyst pellets layer can thus be considered definitely negligible compared to the total pressure drop through the candle.
Furthermore, it is observed that the pressure drop measured at 800°C in the CC configuration for the higher filtration velocity investigated, 0.04 m/s (140 m/h), is equal to 48 mbar, and that measured for the filtration velocity recommended by the manufacturer of the ceramic candles, i.e. 0.028 m/s (100 m/h), is 35 mbar. These values can be considered acceptable for the process, because lower than/or comparable to the typical pressure drops throw the fluidized bed of industrial reactor (Kraft *et al.*, 2017).
Being the results in the two configurations (EC and CC) nearly the same, the attention was focused on the case of the empty candle. Figure 3 shows the pressure drops per unit length for different filtration velocities at the operating temperatures tested: 800, 600, 400 and 200 °C.



	Fitting curves	R^2
800 °C	$1.13e7 \cdot v + 5.99e7 \cdot v^2$	0.99986
600 °C	$1.08e7 \cdot v + 6.79e7 \cdot v^2$	0.99985
400 °C	$9.49e6 \cdot v + 1.09e7 \cdot v^2$	0.99968
200 °C	$7.07e7 \cdot v + 2.21e7 \cdot v^2$	0.99972

Figure 3: Pressure drops for unit length through the empty ceramic candle (EC) measured at 800, 600, 400 and 200°C

Figure 3 shows that the experimental results can be fitted with quadratic curves with very high coefficients of determination, which represent the goodness of the fitting.
The trends of the measured pressure drops were thus correlated with a known expression through porous media as functions of the filtration velocity: the Darcy-Forchheimer equation (Bear, 1972).

$$\frac{dP}{dr} = \frac{\mu}{\alpha} v + \frac{1}{2} \rho C v^2$$

Being α the permeability and C the inertial resistance factor, both depending on the configuration of the system.
Since in our case the surface perpendicular to gas flow direction is cylindrical and therefore variable, the radius changes from an initial value r_0 to a final value r_1 and the expression of the filtration velocity, no more constant, becomes $v = \frac{v_0 r_0}{r}$, being v_0 the filtration velocity through the external surface of the candle (at r_0). Including this expression in the relation of Darcy-Forchheimer and integrating between r_0 and r_1, the final expression of the pressure drops per unit length becomes the following:

$$\frac{\Delta P}{\Delta r} = \frac{\mu v_0 r_0}{\alpha \Delta r} [ln(r_0) - ln(r_1)] + \frac{(v_0 r_0)^2}{2 \Delta r} \rho C \left[\frac{1}{r_1} - \frac{1}{r_0}\right]$$

The expression is composed of a viscous term, depending on the viscosity μ, and an inertial term, depending on the density ρ of the fluid.
The experimental pressure drops measured through the filter candle have thus been fitted with the relation of Darcy-Forchheimer and the values α and C have been calculated for the EC configuration, taking in consideration the measurements carried out at all the operating temperatures (800, 600, 400 and 200 °C). The values of permeability and inertial resistance calculated are displayed in Table 1.

Table 1: Permeability (α) and inertial resistance factor (C) for the configuration EC

	EC
α (m^{-2})	4.38e-12
C	2.88e8

The experimental values of pressure drops fitted with the expression of Darcy-Forchheimer showed that for all temperatures and filtration velocities, the viscous term largely prevails over the inertial term, being between 70 and 95% of the total pressure drop.

To further evaluate the results obtained, the experimental data were fitted with the Ergun Equation, assuming the porous ceramic candle as a packed bed of small particles. The Ergun equation is expressed by the following formula:

$$\frac{dP}{dr} = \frac{150\mu(1-\varepsilon)^2 v}{\varepsilon^3 d_p^2} + \frac{1.75(1-\varepsilon)\rho v^2}{\varepsilon^3 d_p}$$

Being d_p the particle diameter or pore size and ε the void fraction of the porous medium, 0.35 in the case of the filter candle used for the tests (Pall Corporation, 2000). Similarly to the changes made in the Darcy-Forchheimer equation because of the cylindrical surface of the porous media, the Ergun relation was modified as reported below to be adapted to this specific case.

$$\frac{\Delta P}{\Delta r} = \frac{150\mu(1-\varepsilon)^2}{\varepsilon^3 d_p^2}\frac{v_0 r_0}{\Delta r}[ln(r_0) - ln(r_1)] + \frac{1.75(1-\varepsilon)\rho}{\varepsilon^3 d_p}\frac{(v_0 r_0)^2}{\Delta r}\left[\frac{1}{r_1} - \frac{1}{r_0}\right]$$

Fitting the experimental pressure drop data with the Ergun equation, a particle diameter of approximately 70 μm was deduced; this value is comparable to what observed from SEM analysis by (Heidenreich, 2013) and to the value measured by (Simeone *et al.*, 2011) on similar filter candles. The viscous and inertial terms of the pressure drops obtained from the experimental data and from the fitting with the Ergun equation are reported in Figure 4.

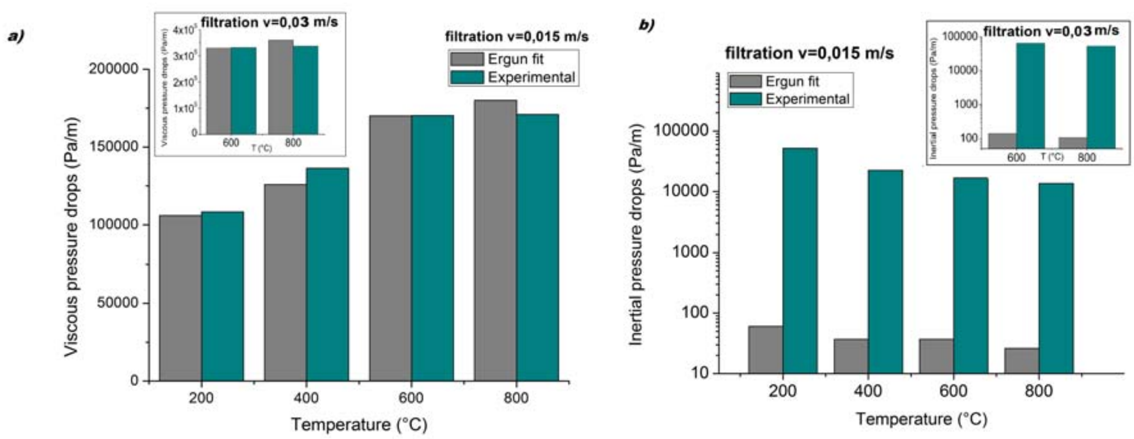


Figure 4: Comparison between the Ergun fitting and the experimental values of the viscous term (a) and the inertial term (b) of pressure drops for unit length through the empty ceramic candle (EC) at different temperatures

As shown in the Figure 4, the viscous term of Ergun equation fairly predicts the same term obtained from experimental data while, since Re_p << 1, the inertial term calculated by the Ergun equation is almost negligible and thus the corresponding experimental value is not well predicted. An explanation of this result can be due to the presence of the outer Al_2O_3 membrane layer (of few 100 μm) with extremely fine pores, that give an extra term as a concentrated pressure drop and therefore proportional to ρv^2.

4. Conclusions

The present study investigated the pressure drops through an empty ceramic candle (EC) and through a filter candle filled with catalyst for tar steam reforming (CC) integrated in the freeboard of a fluidized bed gasifier. It was observed that the pressure drops in the configuration of candle filled with catalyst pellets at 800°C and filtration velocity of 100 m/h, typical operating conditions during biomass gasification tests, are about 35 mbar, which is a definitely acceptable value for the process, being this value lower than the pressure drops of industrial fluidized bed reactors (Gobin *et al.*, 2003). Furthermore, the experimental values showed that the pressure drops through the EC and CC configurations are almost identical, meaning that the catalytic layer does not cause relevant pressure drops.

The experimental values were then fitted with the empirical Darcy-Forchheimer equation, which identifies two contributions, one ascribable to viscous losses and the other to inertial losses. The fitting showed that the major contribution to the total pressure drop is given by the viscous term.

Finally the measured values were recalculated with the Ergun equation; the comparison between the experimental and the calculated data brought to the hypothesis that the total pressure drops through the candle can be described with the Ergun equation plus an additional term relative to concentrated pressure drop due to the external layer of the filter candle. This external layer (of few 100 μm) has in fact very small diameter pores in order to perform efficient particulate filtration on the external surface of the candle, and therefore can act as a further resistance to the flow.

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