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# Tar abatement in a fixed bed catalytic filter candle during biomass gasification in a dual fluidized bed

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ABSTRACT

Catalytic filters are a novel technology for tar conversion in biomass gasification processes. Both particle elimination and tar abatement can be achieved in a single step at high temperatures minimizing energy efficiency penalties. This paper analyses the performance of a fixed bed catalytic filter candle in the reduction of biomass tar generated *in situ* in a dual fluidized bed gasifier (DFBG). In this study, the temperature of the filter was limited to 800 °C. Several variables affecting the performance of the filter were tested. Experiments at different gasification temperatures in the range 750–850 °C were performed so that the amount of tar reaching the catalytic filter was varied. The amount of tar at the catalytic filter inlet did not affect the tar conversion achieved which was around 75%. The major tar compound in the gasification gas at the outlet of the catalytic filter was naphthalene. At the highest temperature tested (850 °C), the tar content in the clean gas was 0.65 g/Nm$^3$. The effect of the face velocity in the filter on the tar conversion reached was evaluated. Tar conversion decreased when the face velocity increased from 40 to 90 m/h as the residence time of the gas in the catalytic filter was lower. The ratio $H_2O$ to dry biomass was also varied (0.5–0.9) in order to produce tars of different nature. Higher conversions were achieved for higher values of this ratio.



## 1. Introduction

Biomass as fuel represents an increasingly interesting option to decrease the $CO_2$ emissions associated to fossil fuel combustion. One of the possibilities is biomass gasification. Gasification is a thermal process in which biomass is converted to valuable gases, mainly $H_2$ and CO, in the presence of a gasifying agent (air, oxygen, steam, $CO_2$ or mixtures of these gases). Dual fluidized bed gasifiers (DFBGs) operated with steam as gasifying agent have demonstrated their advantages in terms of improving gasification efficiency and minimize the generation of by-products such as tar [1]. A dual fluidized bed gasifier consists of two separate reactor zones: a steam gasifier where biomass is converted to nitrogen-free syngas and a combustor where air oxidizes the residual char therefore providing the heat needed to gasify biomass. Typically, the gasifier is a bubbling fluidized bed (BFB) and the combustor is a circulating fluidized bed (CFB). Examples of operating plants are the 8 $MW_{th}$ Güssing gasifier (Austria) [2] and the 2 $MW_{th}$ gasifier at Chalmers University of Technology [3].

Depending on the composition of the gasification gases, the product gas can be burned to produce heat, coupled to gas turbines or fuel cells to produce electrical power or used for the synthesis of hydrogen, methanol and liquid fuels [4]. However, during the gasification process tar appears as by-product of biomass conversion. Tar is defined as all organic compounds with a molecular weight larger than benzene [5]. The formation of tar represents one of the main drawbacks for the commercialization of this technology [6] as it can condense easily and therefore be the cause of downstream pipeline blocking and engine or turbine fouling. In order to become a competitive technology, tar concentration in the product gas of a biomass gasifier should be reduced to levels that can simultaneously comply with environmental regulations and be compatible with the end application [7]. Some indicative specifications for tar content in the gasification product gas can be found in literature. For gas engines, less than 50 mg/Nm$^3$ are required. In the case of fuel cells or syngas or methanol synthesis, the limits are quite lower, less than 1 and 0.1 mg/Nm$^3$, respectively [6].

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Tar abatement measures have been traditionally classified into two different categories: primary and secondary methods. A primary method implies tar reduction inside the gasifier bed while secondary methods include cleaning downstream the gasifier. As primary measure, non-metallic catalysts are added to the gasification bed to reduce *in situ* the tar produced in biomass gasification. Dolomites are the most widely in-bed catalysts for tar conversion in biomass gasification processes. Olivine, another naturally occurring mineral, has also demonstrated tar conversion activity similar to that of calcined dolomite and is a much more robust material than calcined dolomite. In a previous paper from the authors, a new developed Fe/olivine material [8] was tested as in-bed catalyst for tar conversion [9]. The capacity of this material for primary in-bed reduction of biomass tar in a dual fluidized bed system was assessed. The Fe/olivine material led to an important decrease in the amount of produced tar. As secondary measure, commercial Ni steam reforming catalysts have also been widely used during the last decades for tar conversion downstream the gasifier bed. Most of them were designed to be used in a fixed bed [10]. They showed activity for tar destruction. Besides complete reforming of methane water-gas shift activity was observed which allowed to adjust the $H_2$/CO ratio of the product gas. Nevertheless, they required a product gas without particulates despite gasification gases are a high particle-loaded stream [10]. A hot gas filter would be then needed upstream of the catalytic bed, but the possibility of pore blocking in the filter due to thermal tar conversion was also a potential problem. In this case, the use of catalytic filters would be more advisable [11]. Catalytic filters allow integrating in a one-step cleaning process removal of particles and tar abatement at high temperature. Moreover, catalytic filter candles can be directly integrated into the freeboard of a fluidized bed biomass gasifier called the UNIQUE gasifier concept [12].

In the last years, the performance of these hot gas catalytic filters has been evaluated showing a very good tar and methane reforming activity. Different filter configurations have been tested [13–20]. This paper is focused on fixed bed catalytic filter candles, which consist of a porous inner tube fixed at the head of the filter candle to allow the integration of a catalyst bed. This type of catalytic filter presents the highest potential to provide high catalytic capacity [16], mainly due to two reasons: the possibility to integrate a high amount of catalyst material in the hollow cylindrical space of the filter candle and the possibility to integrate different types of tar-specific reforming catalysts. Nacken et al. [16] evaluated different tar reforming catalytic systems to be integrated in a filter element of the type DIA-SCHUMALITH®. Calcined dolomite, CaO-$Al_2O_3$ and MgO were used as catalyst support and different NiO loadings were evaluated. The authors found an important effect of the structural and chemical properties of the support material on the catalytic properties for tar reforming. After testing with naphthalene as model tar compound, they selected an integrated MgO supported Ni catalyst (6 wt.% NiO loading) to be further evaluated in a bench-scale fluidized bed biomass gasification plant, operating at atmospheric pressure and temperature within the range 800–820 °C [13]. The experimental tests were performed using a segment of a full industrial size filter candle placed in the freeboard of the gasifier, which was a bubbling fluidized bed. Crushed almond shells were utilized as the biomass feedstock. Notable improvements in gas quality when using this type of catalytic filter candle were reported. Hydrogen yield was increased and tar content in the gas was reduced. The main compounds found in the tar were toluene and naphthalene. Pressure drop measurements were carried out with time and they increased with time until a plateau was approached after 22 h of continuous operation.

In the present work, a Fe/olivine guard bed followed by a hot catalytic filter at the outlet of the biomass gasifier has been used for tar abatement. Therefore, the catalytic filter was tested with real biomass tar generated *in situ* in a DFBG which adds value to the results obtained. Moreover, the evaluation of the influence of different operational parameters on the filter performance was considered. The tar amount and composition at the inlet of the catalytic filter as well as the face velocity were varied and their effect on tar conversion studied.

**Table 1**
Proximate and ultimate analysis of pine wood (a.r).

| Composition (wt.%) | |
|---|---|
| Moisture | 6.3 |
| Ash | 1.1 |
| Volatiles | 77.2 |
| Fixed carbon | 15.4 |
| C | 46.6 |
| H | 6.0 |
| N | 0.2 |
| S | 0.004 |
| Cl | 0.002 |
| High heating value (kJ/kg) | 18235 |

## 2. Experimental

### 2.1. Bed material and biomass

Fe/olivine is used as bed material in the fluidized bed gasifier with a particle size in the range 0.1–0.25 mm. Details about the characteristics of this material can be found elsewhere and only a brief information is given here [9]. The Fe/olivine was prepared by impregnation so that the final iron percentage in the olivine was 16%. The biomass feedstock used in this work was pine wood with an average particle size of 0.5–2.0 mm. Table 1 shows the proximate and ultimate analysis of biomass as received (a.r).

### 2.2. Dual fluidized bed gasification plant

The catalytic filter for tar abatement has been tested with biomass tar produced *in situ* in a bench scale dual fluidized bed gasification plant located at ICB-CSIC. Details about the configuration of this unit can be found elsewhere [9]. Fig. 1 shows a scheme of the gasification plant. The gasifier was a bubbling fluidized bed where biomass was fed in the bed and gasified with steam. Solids exiting the gasifier flowed through a fluidized bed acting as a loop seal to avoid mixing the atmospheres in the gasifier and the combustor. In the combustor, the ungasified char was burnt. The hot solids were transported along a riser, collected by a cyclone and then kept in a solid reservoir which also acted as loop seal. The solid flow returning to the gasifier was controlled with a solid valve. Gas product streams were connected to on-line gas analyzers which determined CO, $CO_2$, $CH_4$, $O_2$ and $H_2$. In some specific experiments, off-line gas analyses were carried out in a gas chromatograph (HP 5890) fitted with a Porapack N column to analyze the presence of $C_1$-$C_3$ hydrocarbons in the gas outlet stream of the gasifier.

In order to test the effect of the catalytic filter elements in tar reduction, a new reactor and furnace were located downstream the gasifier prior to the tar measurement, as it is shown in Fig. 1. The filter was placed inside this new reactor and then heated by the furnace which allowed controlling the temperature inside the filter. It was possible to by-pass the catalytic filter in order to determine the tar content at the inlet of the catalytic filter.

### 2.3. Catalytic filters

A catalytic filter with dimension $70 \times 50 \times 30 \times 16$ has been tested in the present work. It was prepared by Pall Filtersystems GmbH from a commercial hot gas filter candle of the type DIA-