

Research article

Tar abatement for clean syngas production during biomass gasification in a dual fluidized bed



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ABSTRACT

Syngas obtained from biomass gasification needs to fulfil strong purity requirements before being used as raw material in power energy generation or chemicals manufacturing. The use of hot catalytic filter candles inside the freeboard of fluidized bed gasifiers allows obtaining clean syngas without dust and low tar content. The tar removal efficiency of four different catalytic filter designs was evaluated with real biomass tar produced in situ in a dual fluidized bed gasifier (DFBG). The tar conversion reached at the outlet of the fluidized bed gasifier was larger for the candles with catalytically active layer design. If a monolith is also incorporated, the tar conversion increases up to 95% which is one of the highest values obtained up to date. In this case, the tar content at the outlet of the catalytic filter was as low as 0.2 g/Nm³ (N₂ free, d.b.).

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1. Introduction

Biomass gasification represents a promising technology to produce energy from a renewable source with zero CO₂ emissions. Gasification allows transforming biomass in a gas with high content of H₂ and CO which account for more than 70% of the energy stored in the biomass. Among the available technologies for biomass gasification, dual fluidized bed gasifiers (DFBG) allow reaching high gasification efficiencies [1], as it has been shown in some operating gasification plants in Austria [2] and Sweden [3]. In a DFBG, steam gasification takes place in a bubbling fluidized bed (BFB) where biomass is converted to syngas. Following this, the residual char is transferred to a circulating fluidized bed (CFB) which acts as a combustor, where the char is oxidized and therefore heat is generated to be used in the subsequent gasification process.

Nevertheless, other gasification products also present in the gasification gas can lead to operational problems in the further use of the syngas generated as raw material in power energy generation or chemicals manufacturing. One of these products is the solid particles leaving the fluidized bed. In recent years, the use of ceramic and metallic filters for particle filtration at hot conditions has been investigated [4–6]. Another product is tar, composed by those organic compounds with a molecular weight larger than benzene [7]. In order to prevent tar condensation and therefore fouling, it is desirable that the tar content is decreased down to 30 mg/Nm³ or even lower if the gasification gas is to be used in downstream units such as gas engines or turbines [8].

If the gas is intended for syngas or methanol production or for use in a fuel cell, then more severe restrictions are applied and the tar content should be further reduced to values between 0.1 and 1 mg/Nm³ [9].

In recent years, catalytic hot gas filters for tar abatement have been developed as a cost-effective way to upgrade biomass gasification gas [10–12]. A catalytic filter candle is normally placed in the freeboard of a fluidized bed where gasification takes place. The incorporation of a catalytic filter inside the gasifier presents several advantages. On one hand, it contributes to maintain the thermal efficiency of the biomass conversion process and on the other hand, particle entrainment is avoided. Therefore, a hot and clean gas is obtained at the outlet of the gasifier with reduced investment costs. Three different types of manufacture processes for catalytic filters have been described in literature [13,14].

- i. Incorporation of a catalytic component in the ceramic grain and binder mixture during the ceramic filter manufacture process
- ii. Modification of the design of the ceramic filter by including a porous inner tube fixed at the head of the filter candle to allow the integration of a catalyst fixed bed
- iii. Catalytic coating on the porous support of a conventional hot gas ceramic filter.

The first process was early discarded due to the low surface area of the catalytic filters produced. The high temperatures used in the manufacturing process led to grain sintering and therefore to losses in the active surface of the catalyst [13]. Catalytic filters produced under the other two processes have been optimized and tested under different

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conditions. Fig. 1 presents a scheme of the different configurations of these two catalytic filters.

Catalytic filters with a catalyst fixed bed (FB) are shown in Fig. 1A. They present a high catalytic potential given their flexibility to integrate a custom-made tar reforming catalyst and their capability to integrate high amount of this catalyst in the hollow cylindrical space inside the filter candle considering the limitations imposed by the total weight of the candle and the price of the catalyst incorporated. However, face velocities referred to the outer surface of the catalytic filter candle should be limited to allow enough residence time for the catalytic tar reforming reaction [15].

Studies about the optimum composition of the fixed bed catalyst can be found in literature. Nacken et al. [13] tested several tar reforming catalyst systems of different NiO loadings. They evaluated the effect of the variation of the catalyst support material, the preparation conditions, the NiO loading and the effect of doping with ruthenium on the catalytic activity of the tar reforming catalyst. The catalytic activity tests were conducted using naphthalene as model tar compound. The highest catalytic reforming activity was found for a MgO supported Ni catalyst with a NiO loading of 6 wt%. With this catalyst, complete naphthalene conversion at 800 °C during 100 h operation even in the presence of H₂S was reached. Therefore, this catalytic filter was tested in a larger scale. A catalytic filter candle of adequate dimensions was manufactured and inserted in the freeboard of a bubbling fluidized bed gasifier where crushed almond shells were used as feedstock [17]. Gas and hydrogen yields were notably increased with the use of this catalytic filter and tar content at the outlet of the catalytic filter was between 0.7 and 0.95 g/Nm³ (N₂ free, d.b.). Besides, stable performance of the filter was observed after 22 h of gasification.

The catalytic filters with catalytic coating on the porous support of the conventional hot gas ceramic filter are denoted as catalytic layer filters (CL) (Fig. 1B). This design of catalytic filter had already been tested for combined particle separation and NO_x removal from laboratory [18] to pilot scale [19]. The advantages of the catalytic layer filters is that higher face velocities can be used compared to the fixed bed catalytic filters and therefore the size and weight of the catalytic filter could be reduced. This also implies that for the same outer diameter and superficial velocity higher residence time can be achieved when compared to the fixed bed catalytic filters. Besides a simplification of the manufacture process compared to the fixed bed filters is also expected [15]. The possibility of integration of a tar reforming catalyst as a catalytic layer by catalytic activation of 10 mm thick alumina based filter disks was first

demonstrated [10,20,21]. Then, several studies were carried out aiming at finding suitable catalytic systems for the pore walls of ceramic filters which combine high surface support materials and active catalysts [11, 15]. In these studies, MgO and CaO–Al₂O₃ were used as supports as well as MgO–CaO and MgO–Al₂O₃. In some cases, they were doped with La₂O₃, olivine or ZrO₂. In all cases, the coated filters were catalytically activated by impregnation with the appropriate aqueous solution of nickel nitrate hexahydrate to adjust the NiO loading amounts of 6 and 60 wt% related to the amount of catalyst support [15]. The catalytic activity was evaluated in all the cases using naphthalene as model tar compound. Promising materials were selected to manufacture filter candles to be tested in the freeboard of a bubbling fluidized bed [22]. In these experiments, tar conversion extent obtained by means of the catalytic filter was around 58% with final tar contents in the gas around 0.8 g/Nm³ (N₂ free, d.b.). Methane was also partially converted (28%). As a result, a significant increase in the gas yield (15%) and in hydrogen concentration was reported.

Modifications and improvements of the first design of catalytic layer filters (CL) have been recently presented. First, the replacement of SiC as filter material with another material which could withstand the high gasifier freeboard temperatures (between 800 and 850 °C) was accomplished. SiC was initially used due to the high heat conductivity and good thermal shock resistance for cyclic back pulse cleaning of the catalytic filter [16]. However, it was replaced by Al₂O₃ which allowed long operating times at 850 °C [23]. One of the new configurations for catalytic layer filter candles included an additionally integrated catalyst: a catalytically activated Al₂O₃-based hollow-cylindrical monolith integrated in the hollow cylindrical space of the catalytic filter candle [24]. The incorporation of the monolith increased the Ni load of the catalytic filter. Promising results were obtained with this new design of catalytic layer filter candles (CL + M). Using the same experimental apparatus at the same operating conditions [23], tar conversion of 93.5% was reached with the use of the catalytically activated monolith in comparison with the 58% tar conversion of the catalytic layer SiC candle. Results were also better than those obtained for SiC candle of fixed bed design, where 79% tar conversion was obtained. The final tar content of the clean gas was around 0.25 g/Nm³ (N₂ free, d.b.). A catalytically activated ceramic foam as additional reforming step for integration into the hollow-cylindrical space of the catalytically activated filter candle was used (CL + Foam). The catalytic activity of this combination at different superficial velocities was first examined using naphthalene as model tar compound [25]. Based on these results, a catalytic filter of combined

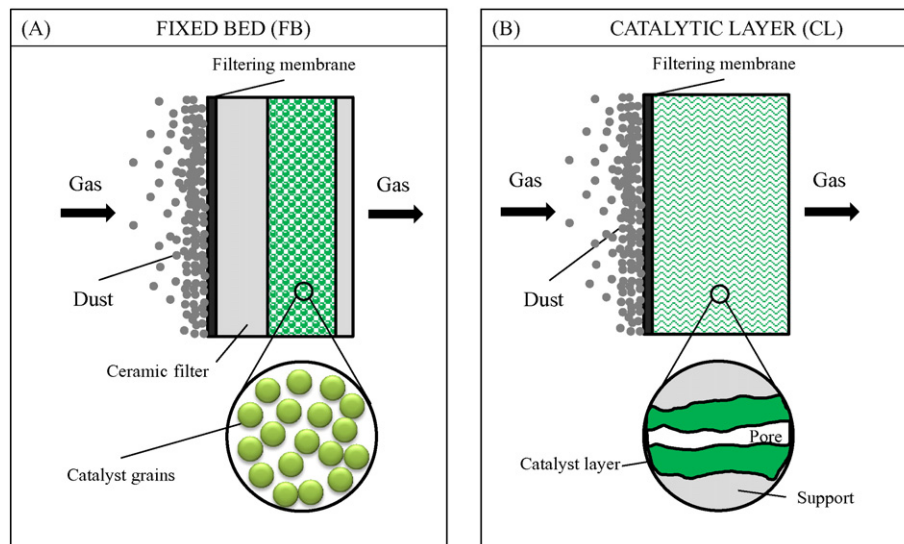


Fig. 1. Scheme of configurations of catalytic filters: (A) fixed bed and (B) catalytic layer. Adapted from Hackel et al. [16] and Nacken et al. [15].

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