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High temperature gas filtration with ceramic candles and ashes characterisation during steam–oxygen blown gasification of biomass

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ABSTRACT

Two experimental campaigns were performed with the aim to study the effect of two different bed materials, magnesite and olivine during steam–O_2 biomass gasification. The test-rig consists of a 100 kW_{th} atmospheric circulating fluidized-bed gasifier and a high temperature filter unit which contains 3 rigid ceramic candles with an outer diameter of 60 mm, 10 mm wall thickness and a length of 1520 mm. Tests were performed with different fuels (A-wood, B-wood, miscanthus and straw). Two types of filter elements were used, Dia-Schumalith[1] (DS3) and Dia-Schumalith[1] N (DSN1), which operated at 800 °C for 58 h and 50 h, respectively. The filtration performance was studied through continuous observation of the increasing pressure drop during the build-up of the dust cake. Gas face velocities ranged between 2.5 and 5 cm s^{-1}. DSN1 elements showed longer steady filtration compared to DS3 candles with filtration efficiencies equal to 100%. Formation of calcium and potassium silicates resulted from filter cake analyses. The filtration process influenced gas and tar composition of the incoming gas flow. Hydrogen content increased about 10% (dry basis) and the heavier tar compounds appeared to be broken into lighter chains, such as naphthalene whose concentration increased.

1. Introduction

Gasification of biomass is one of the promising routes of using this abundantly available and highly diverse renewable energy source. Advantages of this technology include high efficiency, low emissions, less waste and economic benefits even at comparatively small scales. The raw gasification product gas contains CO, CO_2, H_2, CH_4, H_2O and N_2 as main constituents. In addition to these main gas components, the gas contains organic (light hydrocarbons and tar) and inorganic (H_2S, HCl, NH_3, alkali metal) impurities and particulates [1]. An obstacle for widespread technology implementation and deployment is still formed by lacking economical solutions for the removal of these impurities. Tar species cause challenging issues in downstream equipment, such as filter plugging by coke formation and condensation on cold spots of equipment (e.g. engines, fuel cells), while deposition of solid particulate can cause deactivation of catalytic units and corrosion of turbine blades with deposition of alkali metals. Most equipment using biomass derived product gas requires a final tar content of 0.05 g Nm^{-3} or even less [1] while the limit set for particulate concentration for turbine applications is 2 ppmw [2]. Gas cleaning and conditioning are currently among the major areas of research in the field of biomass gasification. The main techniques involved in gas cleaning are mechanical methods like cyclones, hot gas filters, granular beds, catalytic and thermal cracking methods [3]. Hot gas cleaning is a more preferable technology because energy loss is minimised by avoiding cooling of raw product gas. Hot gas filtration is a core technology in this respect. The filtration systems currently under consideration for hot gas cleaning are fixed bed filters, ceramic filters, metal filters and barrier filters [4]. Candle filters manufactured with porous ceramics are very promising filtration systems with artificially elevated porosity and adjustable pore size and shape [5]. They offer the major advantage that they are resistant to high temperatures and can be operated at temperatures well above 400 °C. Other advantages include enhanced erosion and corrosion resistance, resistance to vibration and acoustic loads, and the possibility to remove multiple contaminants [6]. Ceramic filters are mainly used for removal of dust from hot gases produced by industrial processes such as combustion and gasification. They should be capable of operating for more than 10,000 h and maintain high particulate removal efficiencies, high flow capacity and low pressure drop [7,8]. First generation monolithic silicon carbide filter elements suffered of loss of strength and consequent mechanical failure when operated during pressurised fluidised bed combustion of coal. Thermal shock, fatigue, development of microcracking in the clay bonded silicon carbide as well as ash bridging caused loss of structural integrity of the candles [9,10].

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Advanced second generation filter elements with higher resistance to crack growth have been developed and studied for particulate removal at high temperature applications in pressurised fluidised-bed coal combustion (PFBC) [11] as well as in integrated coal and biomass based gasification combined cycles (IGCC) [12–14]. Major problems that can occur during a filtration process, hence causing the interruption of the operation, are the impossibility of maintaining stable pressure drop during the build-up of the filter cake and low filtration efficiency in dust removal. For this reason, new techniques are continuously being introduced such as the coupled pressure pulse (CPP) cleaning system [15], which guarantees higher efficiency in the regeneration of the candles, and the particulate control device (PCD) designed for enhancing the efficiency of a filter element in reducing particles load [16]. Nowadays, reliability of hot gas filtration systems towards their utilisation on industrial scale remains a major challenge.

Thus far, a detailed study on the filtration performance at temperatures as high as 800 °C during gasification conditions can be still considered original work. This paper presents new results from two experimental campaigns which were performed to continue an investigation started by the authors [17] on the behaviour of ceramic filter elements tested in a pilot scale filter unit at high temperatures during steam–oxygen blown gasification of different types of biomass. This study, performed at Delft University of Technology, was part of the "Chrisgas" project.

2. Methods

2.1. Experimental pilot scale facility

The test-rig (Fig. 1) consists of a steam/O_2 blown 100 kW_{th} atmospheric circulating fluidized-bed gasifier (CFBG) [18] with a primary cyclone included in the reactor design, and a high temperature filter unit (Pall filter) 404 cm long, with an internal diameter of 33 cm. The filter vessel contains three rigid ceramic candles, each one 1520 mm long, with an outside diameter of 60 mm and 10 mm wall thickness. The regeneration of the candles is done through the CPP, a novel method developed and patented by Karlsruhe Institute of Technology (KIT) in cooperation with Pall. With this system each candle is directly connected to the re-cleaning gas tank through a valve and nitrogen is used to pulse the candles. The frequency with which each candle is backpulsed, the opening time (pulse time) of the blow back (BB) valves (v_1, v_2 and v_3), and the pressure of the pulse are defined as cleaning parameters. The filter vessel and the N_2 BB reservoir are electrically heated with four heating elements (T_{max} = 1000 °C) and one heating element (T_{max} = 450 °C), respectively. The facility is also equipped with another filtration unit provided by Bayerische Wollfilzfabriken Textil GmbH & Co. KG (BWF) and connected downstream of the CFBG in parallel to the Pall filter. The BWF vessel contains four filter elements made of woven ceramic fibre material. The maximum temperature at which the BWF filter can operate is 450 °C. A more detailed description of the test-rig was given in [17].

Two types of filter candles were used during the filtration tests: Dia-Schumalith[1] (referred as DS3) and Dia-Schumalith[1] N (referred as DSN1). The filter material that constitutes the candles Dia-Schumalith[1] is a silicon carbide porous structure coated with a thin mullite membrane ($3Al_2O_32SiO_2$) that is sintered on the monolith structure and that behaves as the actual filtering element. DSN elements differ from DS elements only in the support's binder which is made of a material with higher temperature stability.

2.2. Gasification-filtration tests matrix

In 2009 two steam–O_2 gasification-filtration campaigns were performed, the first one with DS3 and the second one with DSN1 candles, and aimed at studying the effect of different bed materials on the gas composition, tar concentration and filtration behaviour. The selected bed materials were: olivine, a magnesium iron silicate ($(MgFe)_2SiO_4$), which was used fresh or namely untreated (untr.) and after being thermally treated (tr.) in a temperature range between 900 and 1200 °C; magnesite, a carbonate mineral ($MgCO_3$) whose calcined form is mainly constituted by MgO. Four fuels were tested: A-wood (clean wood), B-wood (recycled demolition wood), miscanthus (energy crop plant) and straw (agricultural residue). Their chemical composition was analysed by Forschungszentrum Jülich and is given in Table 1. Kaolin, a clay mineral ($Al_2Si_2O_5(OH)_4$), was used as additive in most tests with miscanthus and straw, fuels particularly rich in alkali compounds, but also in some tests with B-wood. Thanks to its property of "alkali getter", kaolin helps avoiding bed agglomeration in the reactor which can be caused by formation of ashes with low melting point when these ones are rich in alkali.

Once a new set of candles was positioned in the filter vessel, the unit was heated up while nitrogen was flowing through the system. The filter was operated at 800 °C during both campaigns. Main process conditions and filtration parameters of the two sets of experiments are given in Tables 2 and 3. The duration of a daily test could vary between 2 and 13 h, depending on the unpredictable occurrence of technical problems. DS3 and DSN1 candles were tested in total 58 h and 50 h, respectively. While the set temperature of the filter unit was kept constant at about 800 °C, the cleaning parameters were modified in order to find a cleaning strategy which could guarantee longer and stable filter operation. The pressure of the BB vessel varied between 170 and 350 kPa. The experience obtained with previous tests showed that higher BB pressure was necessary in order to have a more effective detachment of the filter cake. The longest BB frequency interval was set at 900 s and the shortest at 60 s. DS3 candles were tested mainly with a BB frequency of 300 s while with DSN1 candles a BB frequency of at least 600 s was applied. Opening time of the valves ranged between 150 ms and 200 ms. In general, a shorter valves opening time was coupled with a higher BB pressure.

2.3. Gas and tar analysis

Sampling points for gas and tar were located immediately downstream of the CFBG and of the filter unit, defined from this point on as upstream (UP) and downstream (DW) of the filter. The sampled gas, after being cooled down, was led to a micro gas chromatograph (Varian 4900 micro-GC) and an FTIR (Thermo Nicolet 5700 with a gas cell and an optical length of 2 m) for gas analyses. One column (type COX, length 1 m) of the micro-GC provided the separation of the main gas components (H_2, CO, CO_2, CH_4 and N_2), with subsequent quantification using a TCD detector, while a second column (type CP-Sil 5CB, length 4 m) was used for BTX separation and quantitative analysis using an FID detector. Gas composition was available every three minutes. Water concentration was determined gravimetrically while sampling the raw gas directed to the cascade impactor for particles measurements as well as during wet gas analysis done with the FTIR. Tar compounds (three samples at a time) were sampled according to the solid phase adsorption method (SPA) [19] and sent to KTH for analysis. Accuracy and precision of the above mentioned analysis techniques can be found in [18].

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[1] Dia-Schumalith is a trademark of Pall Corporation.