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Steam–oxygen gasification of forest residues and bark followed by hot gas filtration and catalytic reforming of tars: Results of an extended time test

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Steam–oxygen gasification in a Circulating Fluidized-bed (CFB) reactor was developed for producing transportation fuels from different wood residues. This article presents the results of a two week test campaign, in which crushed forest residues and industrial bark mixture were used as the feedstocks. The aim of the work was to carry out extended time testing of the developed gasification and hot gas cleaning process and to determine the fate of different gas contaminants and trace components of wood. In the test runs, wood fuels were gasified in the CFB reactor at a 0.2–0.25 MPa pressure using a mixture of steam and oxygen as the gasification agent. A mixture of sand and dolomite was used as the bed material in order to maintain stable fluidization and to catalyse in-situ tar decomposition before hot filtration. Raw gas was filtered at ca. 550 °C and the filtered gas was then led into a two-stage catalytic tar reformer. The gasifier performance and the concentrations of different gas contaminants were determined at four different operating variable set points during a total of 215 h of operation. The results for carbon conversion efficiency, raw gas composition and the fate of fuel nitrogen, chlorine and trace metals are presented in this paper. The concentrations of gas contaminants were determined after the ceramic filter unit and after the catalytic reformer. The conversion efficiencies for hydrocarbon gases, tars and ammonia in the reformer are also presented. The test run was carried out as a continuous operation without any interruptions or operational problems.

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1. Introduction

Advanced 2nd generation biofuels can be produced from a wide variety of biomass feedstocks utilising many different biotechnical or thermochemical conversion pathways [\[1\]](#page--1-0). Gasification-based alternatives have two principal advantages over most other conversion routes. Firstly, a wide variety of biomass qualities as well as many waste streams can be used as feedstock. Secondly, various different end products (e.g., methanol, DME, Fischer Tropsch Diesel and Synthetic Natural Gas) can be produced from a mixture of hydrogen and carbon monoxide (syngas).

Different types of fluidized-bed gasifiers have been industrially applied in power and heat applications already since the 1980s. At present several air-blown CFB gasifiers as well as dual fluidized-bed steam gasifiers operating at atmospheric pressure are in industrial use for example in Finland, Austria and Germany [1–[3\].](#page--1-0) Atmospheric-pressure dual bed steam gasification has also recently been demonstrated in a semiindustrial scale production of Synthetic Natural Gas in Sweden [\[4\]](#page--1-0).

Europe and in the USA [\[5,6\]](#page--1-0), but without commercial breakthrough. In addition, the first industrial scale pressurised fluidized-bed gasification plants using brown coal and peat as the feedstock were operated in Germany and Finland in the late 1980s and early 1990s [\[7](#page--1-0)–9]. However, these gasification systems were initially developed for coal and not for biomass feedstocks, which have significantly higher volatile matter content and different ash composition. Very good operation experiences were obtained at the High Temperature Winkler demonstration plant in Germany with Rhenish brown coal [\[7\],](#page--1-0) whereas the operation with peat at the Oulu ammonia plant in Finland already suffered from problems related to the high tar content of syngas and sintering of the peat ash [\[8,9\]](#page--1-0). At the Oulu gasification plant ash sintering problems limited the operation temperature of the gasifier to below 970 °C, resulting in 0.5–1 g/m^3 naphthalene concentrations. This led to deposit formation problems in the final gas cooler and naphthalene condensation in the cooler of the syngas compression system [\[9\].](#page--1-0) In addition, ash sintering and melting created deposits in the gas outlet pipe and in the cyclone and blocked the return leg of the recycling cyclone.

Pressurised oxygen-blown biomass gasification based on bubbling fluidised-bed reactors was also developed already in the 1980s in

Previous studies on pressurised air-blown gasification of coal, peat and wood [\[10\]](#page--1-0) have shown that tar yields in wood gasification were

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still almost an order of magnitude higher than those of peat gasification. Furthermore, biomass ash usually has high concentrations of alkali metals and silica, resulting in more problematic ash behaviour than that ob-served with peat or coal [\[11\]](#page--1-0). These previous experiences were taken as a starting point in 2005, when the development of an optimal biomass gasification process for syngas applications was started again in Finland [\[12\].](#page--1-0) The target of the development was a process concept for intermediate scale production of transport fuels (fuel production ca. 100 kton/a) with good heat integration to forest industries or other heat-consuming industries [\[12](#page--1-0)–15]. The commercialization of CFB gasifiers in the Finnish power and heat market had also created valuable industrial experience and a good technical basis for designing gasifiers for more demanding syngas applications. In 2007–2012, pressurised steam–oxygen-blown Circulating Fluidised Bed (CFB) gasification was intensively developed at VTT [\[12,16\]](#page--1-0). The R&D impetus was related to industrial plans for the production of liquid transportation fuels from forest residues. In the developed process, wood feedstocks are first converted into raw gasification gas, also containing high concentrations of tars and hydrocarbon gases. This hot raw gas leaves the gasifier at ca. 900 °C and is cooled to ca. 550 °C and then filtered before catalytic reforming of tars and hydrocarbon gases. After the reformer the product gas is suitable for final gas cleaning and conditioning using industrially proven gas treatment processes such as shift conversion and acid gas removal processes. Economically promising processes have been designed [\[12,14\]](#page--1-0) on the basis of this concept, with special emphasis on maximising the overall biomass utilisation efficiency by using the by-product heat of the gas cooling and synthesis process, as well as off-gases from the synthesis, for generating heat for process industry or district heating.

Our previous study [\[16\]](#page--1-0) focused on the initial development of the process concept and described the effects of the gasifier operating pressure, used bed material and other main gasifier operating variables on the gasifier performance. The main focus was on the gasifier operation, whereas the performance of the filter unit and the reformer was not yet studied in detail. In these studies we observed that CFB gasification is a stable and easily controlled process when the gasifier is operated at pressures up to 0.4 MPa, whereas operation above 0.5 MPa is challenging due to overheating of the bottom part of the gasifier bed, resulting in ash sintering problems. At lower gasification pressures, the bed material calcium as well as the inherent wood calcium is in the form of CaO, which has many positive effects on gasification as discussed in more de-tail in [\[11,16\].](#page--1-0) At higher pressures calcium is in the form of $CaCO₃$, which does not have similar positive effects on tar decomposition and ash chemistry. Most of the previous tests were carried out with pelletized wood fuels, which are easier to handle at the test rig, whereas the extended time test of this article was carried out using the most potential Finnish biomass feedstocks in their original physical form, without expensive pelletizing.

This paper presents the results of the extended-time gasification and gas cleaning tests, in which the performances of the gasifier, hot filter unit and the catalytic reformer were determined during a 215-hour test run. The gasification process was operated according to the basic process concept identified in the previous R&D stage [\[16\]](#page--1-0) to be most reliable from the operation point of view. In this extended-time test run, the gasifier was operated at 0.2–0.25 MPa and at 910 °C, filtration was carried out at ca. 550 °C and the filtered gas was reformed in a two stage reformer, in which three types of catalysts were used in order to achieve stable tar decomposition without soot formation problems. Background studies on hot gas cleaning described in [\[17\]](#page--1-0) had created the basis for the design of the raw gas cleaning process. The main objective of the extended time test of this article was to demonstrate stable and problem-free operation of the three key unit operations of the process with the two most potential Finnish wood feedstocks, bark and forest residues. In addition, the fates of different gas contaminants in the gasification, filtration and reforming processes were determined in order to create data for the design of the final gas cleaning train before the synthesis processes.

2. Experimental

The 215-hour gasification test run was carried out using three different wood types as the feedstock. Different batches of similar feedstocks had already previously been tested for shorter test periods as described in [\[16\].](#page--1-0) Wood pellets of 8 mm diameter (later referred to as P-W) were made in Finland from clean wood sawdust (pine and spruce) and they did not contain any bark. The pellets were crushed to below 5 mm sieve before loading into the gasifier feed hopper. The crushed forest wood residues (FWR) were collected from Eastern Finland and they contained mainly residues from forest thinning from pine and spruce forests. The crushed bark (Bark), which contained 20–30% of stem wood, was a mixture of softwood bark and birch bark and it originated from an industrial pulp and paper mill in Eastern Finland. The forest residues and bark were first dried during the summer in open piles, assisted by warm air blowing. Then the dried material was crushed to below 10 mm sieve. Table 1 presents the proximate and ultimate analyses and elemental ash compositions of the feedstocks as averages for several individual set point samples. Photographs of the feedstocks are presented in [Fig. 1](#page--1-0). Five representative samples of 3–5 l were taken from each tested feedstock, and then the samples were divided into smaller analytical samples for the different analyses. The results in Table 1 are given as the average of five replicate feedstock samples. The analytical samples and the feedstock analysis results were considered to represent the fuel batches satisfactorily.

A mixture of 70 wt.% dolomite and 30 wt.% sand was used as the bed material in this test run. This bed material mixture was found in previous tests [\[16\]](#page--1-0) to be a good choice when the gasification pressure was below 0.4 MPa. Dolomite originated from Sweden and was sieved to 0.1–0.6 mm particle sizes. The silica sand material was Finnish and 90 wt.% of the particles were within the range of 0.1–0.4 mm. This sand is also used as a bed material at several industrial gasification and combustion plants in Finland. The chemical composition of the bed materials is given in reference [\[16\]](#page--1-0).

The tests were carried out using the Process Development Unit called UCG-PDU illustrated in [Fig. 2](#page--1-0). The figure also shows the sampling points of gas analysis and for collecting different gas contaminant samples. This PDU is based on a pressurised circulating fluidised-bed reactor using steam and oxygen as the primary fluidising agents, which are pre-

Table 1

db: dry basis.

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