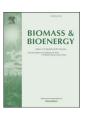
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Research paper

Hydrogen production within a polygeneration concept based on dual fluidized bed biomass steam gasification

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ABSTRACT

Dual fluidized bed biomass steam gasification generates a high calorific, practically nitrogen-free product gas with a volumetric H_2 content of about 40%. Therefore, this could be a promising route for a poly-generation concept aiming at the production of valuable gases (for example H_2), electricity, and heat. In this paper, a lab-scale process chain, based on state of the art unit operations, which processed a tar-rich product gas from a commercial dual fluidized bed biomass steam gasification plant, is investigated regarding H_2 production within a polygeneration concept. The lab-scale process chain employed a water gas shift step, two gas scrubbing steps, and a pressure swing adsorption step. During the investigations, a volumetric H_2 concentration of 99.9% with a specific H_2 production of 30 g kg⁻¹ biomass was reached. In addition, a valuable off-gas stream with a lower heating value of 7.9 MJ m⁻³ was produced. Moreover, a techno-economic assessment shows the economic feasibility of such a polygeneration concept, if certain feed in tariffs for renewable electricity and H_2 exist. Consequently, these results show, that the dual fluidized bed biomass at the production of H_2 , electricity, and district heat.

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1. Introduction

Today, the steam reforming of fossil fuels, mainly of natural gas, is the state of the art technology for H_2 production. After steam reforming, the generated gas passes through a water gas shift (WGS) unit and is finally fed into a pressure swing adsorption (PSA) unit, where H_2 with a purity higher than 99.999% can be produced [1]. H_2 from biomass within a polygeneration concept could be a CO₂ neutral alternative offering a high fuel utilization ratio considering all products.

The dual fluidized bed (DFB) biomass steam gasification generates a product gas with a volumetric H_2 content of about 40% using biomass (wood chips) as feed stock. Therefore, this product gas could be used for CO_2 neutral H_2 production as well as for electricity generation if it is burned in gas engines. Moreover, the process heat can be used for district heating. In addition, in previous experimental investigations, it was also shown that DFB biomass steam gasification is a suitable process for the production of Fischer-

http://dx.doi.org/10.1016/j.biombioe.2016.12.008 0961-9534/© 2016 Elsevier Ltd. All rights reserved. Tropsch fuels as well as synthetic natural gas (SNG) [2,3]. Consequently, DFB biomass steam gasification offers a route for the simultaneous production of valuable gases (e.g. H_2 or CH_4), liquids, electricity, and district heat. This concept is referred to as polygeneration.

Several polygeneration processes have been comprehensively investigated by different authors. For example, Gassner et al. extensively investigated the polygeneration of SNG, power, and district heat [4]–[6]. Moreover, Gale et al. [7] investigated Fischer-Tropsch polygeneration concepts and Gao et al. [8], Li et al. [9], and Liu et al. [10] investigated polygeneration concepts aiming mainly at methanol production. In contrast, Tock and Marchal [11] investigated a polygeneration concept for H₂ production based on biomass and Kyriakarakos et al. [12] investigated a process aiming the production of H₂, potable water, and power based on wind energy and solar energy.

Former experiments carried out by this research group have investigated different process chains, which aimed at H_2 production from the product gas generated by DFB biomass steam gasification technology. The investigations were conducted at the site of the commercial DFB biomass steam gasification plant in Oberwart, Austria, which is a plant for heat and power generation. However, in

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these investigations, the product gas was extracted after the tar scrubber of the gasification plant [13,14].

In contrast, recent research activities have shown that it is even possible to operate a lab-scale WGS unit with tar-rich product gas from the gasification plant without experiencing catalyst deactivation [15]–[17]. Processing the tar-rich product gas gives advantages regarding the energy efficiency because it can be extracted at a higher temperature (about 150°C) and with a higher volumetric steam content (about 35%). Therefore, less heating and steam addition is required before feeding the gas into a WGS unit.

This paper presents the investigations of a lab-scale process chain for the simultaneous production of pure H_2 and a valuable off-gas, which can be used for heat and power generation. The process chain was operated with tar-rich product gas extracted from the commercial DFB biomass steam gasification plant in Oberwart and employed a WGS step, two gas scrubbing steps, and a PSA step. Furthermore, a techno-economic assessment of the presented polygeneration process based on real gasification plant data and the experimental results was carried out.

2. Materials and methods

This section describes the process chain which was used for the experimental investigations. It covers the gasification plant and the lab-scale process chain with a WGS step, gas scrubbing steps, and a PSA step. A parameter study of the lab-scale process chain was conducted prior to this investigation in order to find suitable operating parameters for the different units. In addition, the methodology, which was used for the techno-economic assessment, is described.

2.1. The gasification plant

The lab-scale process chain processed tar-rich product gas from the industrial scale combined heat and power (CHP) gasification plant in Oberwart (fuel power 8 MW). Fig. 1 shows a simplified flowchart of the employed process.

The plant is based on the dual fluidized bed (DFB) steam gasification technology described in detail in Ref. [18]. The CHP plant generates district heat and electricity with biomass (wood chips) as feed stock. Information about the average feed stock composition can be reviewed in Ref. [19].

Biomass is fed into the biomass dryer and, subsequently, into a screw conveyor which transports the biomass into the gasifier. In the gasifier, the biomass is in contact with steam (steam to fuel ratio, d.a.f., of about 0.5) and the bed material (olivine) at about 850°C. The resulting product gas has a volumetric composition of about 40% H₂, 25% CO, 22% CO₂, 10% CH₄, as well as minor amounts of C_xH_y, N₂, and sulfur components. Furthermore, the product gas has a lower heating value (LHV) of up to 14 MJ m⁻³ (d.b.) and a dust content of about 26 g m⁻³ [19].

In the subsequent process steps, the product gas is cooled and passes through a bag house filter. The bag house filter has a particle separation efficiency of more than 99%. Therefore, a practically dust free product gas can be assumed after the filter [20]. In addition, in the following rapeseed methyl ester (RME) gas scrubber, the product gas is cooled from about 150 to 40°C, therefore, tar, steam, and other condensable fractions of the product gas are removed before the product gas is fed into the gas engines for electricity generation. Heat from the flue gas line is mainly recovered for the process and for district heating. Fly ash is removed before the flue gas is released into the atmosphere.

It is possible to extract a partial flow of the product gas for experimental work from an extraction point located before the RME gas scrubber (see Fig. 1). The tar content of the gas extracted before the RME gas scrubber is significantly higher (about 9 g m⁻³ GCMS tar [19]) than after the RME gas scrubber (about 3 g m⁻³ GCMS tar [19]), which is a challenge for the reliable operation of the subsequent process chain. However, the tar-rich product gas has a higher temperature and a higher volumetric steam content (about 150°C and 35%) compared to the gas extracted after the RME scrubber (temperature about 40°C and volumetric steam content of about 7%). Therefore, the tar-rich product gas shows advantages regarding the energy efficiency because the gas needs less heating and less additional steam before processing in a WGS reactor. A detailed description of the process and the CHP plant can be found in Refs. [13,20,21].

2.2. The lab-scale process chain

This section describes the lab-scale process chain which processed the tar-rich product gas extracted from the gasification plant in order to generate H_2 and a valuable off-gas stream. Fig. 2 shows a simplified flowchart of the process chain where I to V indicate the gas sampling points. The process chain employed a WGS unit, an RME scrubber, a glycol scrubber, and a PSA unit for the final gas separation. These different units are discussed below.

2.2.1. The water gas shift unit

The aim of the water gas shift unit was the conversion of CO to H_2 according to Equation (1).

$$CO + H_2 O \rightleftharpoons H_2 + CO_2 \quad \Delta H = -41.1 \text{kJ} \cdot \text{mol}^{-1} \tag{1}$$

The WGS unit consisted of three fixed bed reactors (A, B, and C) in series filled with a commercially available Fe/Cr based catalyst. Each catalyst bed had a diameter of about 9 cm and a bed height of about 40 cm resulting in a catalyst volume of approximately 2.5 dm³ in each reactor. The activation procedure of the catalyst before the operation is described in detail in Ref. [21].

Along the height of each reactor, seven type J thermocouples were installed in order to record the temperature profile along the reactors. At the inlet and outlet of each reactor, the gas stream could be heated or cooled in order to achieve the desired gas inlet, respectively, outlet temperatures.

Before the inlet (I, Fig. 2) of the first reactor, steam was added to assure a high steam content in the processed gas in order to avoid coking and carbon deposition on the catalyst surface. The WGS unit was operated at ambient pressure and the gas inlet temperature was set to about $350^{\circ}C$ (I, Fig. 2).

In previous experiments, the WGS unit was operated with a tarrich product gas for more than 2250 h in order to prove the stability of the catalyst under the conditions of a tar-rich product gas. During that investigation, no catalyst deactivation could be observed [16].

2.2.2. The gas scrubbing units

Two gas scrubbing units were employed after the WGS unit in order to remove tar and NH_3 from the processed gas. In addition, steam should be condensed.

The first scrubbing device, which is referred to as the RME scrubber, used RME as the scrubbing fluid. In past investigations, it turned out that RME is a good solvent for tar [22]. The RME scrubber was operated at ambient pressure and at a temperature of about 15°C. Therefore, the tar-rich gas stream was cooled and, consequently, tar was condensed and dissolved in the RME. Furthermore, steam contained in the gas stream was condensed and most of the NH₃ was dissolved in the condensed steam. Some RME in the scrubber was continually removed and replaced with fresh RME in order to avoid saturation with tar.

After the RME scrubber, the processed gas passed through a

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