



Research paper

Product gas composition for steam-oxygen fluidized bed gasification of dried sewage sludge, straw pellets and wood pellets and the influence of limestone as bed material



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ABSTRACT

This is a study on gasification of low cost biogenic fuels for synthesis gas production with assessment of the gas composition and impurity concentrations. Experiments were carried out in a 20 kW fuel input bubbling fluidized bed facility with steam and oxygen as gasification agents. Dried sewage sludge, straw pellets and, for reference, wood pellets were used as fuels. The influence of limestone as low cost bed material acting as in-situ tar cracking catalyst and sulfur sorbent was analyzed. In terms of product gas quality and the influence of limestone as bed material, it can be summarized: (i) Regarding permanent gases, all fuels yielded comparable gas composition and output on water and ash free basis. The gas contains 40% H₂ and 20% CO (dry mole fraction) and is thus suitable for synthesis. (ii) The gravimetric tar concentration of the product gases varied between 15 and 28 g m⁻³ for experiments with inert silica sand or ash bed, but could be reduced significantly by addition of calcined limestone to below 6 g m⁻³. As a novel approach, also the elemental composition of the gravimetric tar was analyzed. (iii) Since straw and sewage sludge contain significant amounts of nitrogen, sulfur and chlorine, the concentrations of H₂S, NH₃ and HCl in the syngas have been measured by wet chemical methods for these fuels. Limestone acts as sulfur sorbent and it was shown, that the H₂S content of the product gas could be lowered significantly, reducing the effort of downstream gas cleaning.

1. Introduction

Steam-oxygen fluidized bed gasification is a promising high temperature atmospheric process for conversion of solid fuels to a nitrogen free product gas, that is rich in hydrogen and carbon monoxide and thus can be used for synthesis of fuels (e.g. synthetic natural gas, dimethyl ether, kerosene) and chemicals (methanol, plastic monomers) [1]. As gasification agent, a mixture of steam and oxygen is used, while the necessary heat for the endothermic gasification process is provided through partial fuel oxidation. Therefore only a single fluidized bed reactor is needed, in contrast to the allothermal dual fluidized bed (DFB) steam gasification (e.g. GobiGas [2], Güssing [3]), which simplifies the process layout and saves capital costs concerning the gasifier. In addition to that, oxygen is an effective reactant for enhancing the gas yield, char conversion and tar reforming [4]. On the other hand, the on-site production of oxygen requires energy and investment. Oxygen can be supplied by air separation with an energy demand of 720 kJ kg⁻¹ [5], which corresponds to approximately 1% of the GCV of the fuel input at ER = 0.25 and has thus no major influence on the overall

energy balance. If a power-to-gas-process is also installed on-site, oxygen is produced as a by-product of water electrolysis with abundant renewable power and can be stored and used for the gasification. Compared to conventional air gasification, the steam-oxygen gasification delivers a nitrogen-free and thus higher quality product gas, as required for downstream synthesis.

The gasifier is operated at high temperatures of about 850 °C. Since the fluidized bed provides very good material and heat exchange, the fuel undergoes fast pyrolysis. It decomposes into permanent gases, tars, char and ash. The char and primary tars are subsequently gasified or respectively reformed by oxygen and steam. The raw product gas, leaving the gasifier, consists mainly of H₂O, H₂, CO, CO₂, CH₄, light hydrocarbons (C₂, C₃, C₄), and larger hydrocarbons, e.g. aromatics and polyaromatics, which are referred to as tars.

For economic and ecological reasons the use of biogenic residues as low-cost regenerative fuels is favorable. Where this work's focus is set on dried sewage sludge as fuel, because of its abundant and constant seasonal availability, e.g. on dry basis over 10 Tg a⁻¹ in Europe [6], and its negative fuel costs (disposal fees). Additionally, wheat straw is

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considered as fuel, as it is an agricultural side product and has thus a great potential in Germany [7] and Europe [8].

As common for most thermochemical gasification processes, the product gas contains tars and - especially for low quality fuels like sewage sludge - other impurities such as H_2S , HCl and NH_3 , which are not tolerated by downstream synthesis catalysts [9]. Therefore, the product gas needs to be cleaned before synthesis. Efficient and low cost gas cleaning is considered to have a huge impact on the economic feasibility of the process [10]. For the design and development of the entire process chain, comprising of gasification, gas cleaning and synthesis, detailed information on the product gas composition, including tars and impurities, is needed. Reduction of tars and impurities by primary measures such as active bed materials and optimized operation parameters can be economically and technically favorable [11,12]. A limestone based, and thus CaO containing, bed material or bed additive can provide several functions in fluidized bed gasification: (i) it supports the catalytic cracking and steam reforming of tars [9,13,14]; (ii) it acts as sorbent to capture unwanted product gas components such as H_2S ($\text{CaO} + \text{H}_2\text{S} \leftrightarrow \text{CaS} + \text{H}_2\text{O}$) [12]; (iii) it increases the ash melting point and therefore decreases the risk of bed agglomeration. CaO is far more active for H_2S capture [15] and tar cracking [16] than CaCO_3 . Hence, in order to gain high tar and H_2S reductions, the conditions in the gasifier (temperature, CO_2 partial pressure) should be kept at calcining conditions ($\text{CaO} + \text{CO}_2 \leftrightarrow \text{CaCO}_3$), which is the case in this work.

Compared to other gasification processes, the atmospheric bubbling fluidized bed steam-oxygen gasification was less studied in literature and especially for residue gasification and syngas impurities, there is little data. The studies, considered most relevant to this work, are referred to in the following. Steam-oxygen gasification of sewage sludge was conducted in a small scale bubbling fluidized bed at a gasification temperature of 816°C and an ER of 0.25 [17]. The reported gas mole fractions on dry and N_2 -free basis were 35% CO_2 , 30% H_2 , 21% CO , 9% CH_4 , 5% C_2 . Also activated carbon and CaO were used in a second fluidized bed stage with promising tar reduction. This study shows the potential of the process and CaO as bed additive, but lacks H_2S and HCl measurement. Also the facility is rather small and has a special design with two serial fluidized beds. In other studies [18] [19], sewage sludge was used in steam air gasification, where the main focus was set on the influence of WHSV and different bed materials: dolomite, olivine, alumina. Dolomite, where the active component is CaO , was found the most active bed for tar reduction. However, since air-steam gasification leads to a significant dilution of the product gas, it is not applicable for synthesis gas production. Sewage sludge steam oxygen gasification with rotary kilns [20] or screw reactors [21], as well as steam gasification [22,23] has also been investigated in literature, but cannot be directly compared to this work. At IFK, preliminary work [24] has been done on dual fluidized bed (DFB) steam gasification with fuels and bed materials comparable to this study. Also the same gasification reactor was used. By comparison of the results of both IFK studies, the differences between the process layouts can be assessed.

A 1000 kW internally circulating bubbling fluidized bed pilot plant was used by Ref. [25] to perform steam gasification of almond shells with an ER of 0.23 and a dry gas composition on mole basis of 30% H_2 , 30% CO , 25% CO_2 , 10% CH_4 and 2% other light hydrocarbons as well as impurities of $40 \cdot 10^{-6}$, $50 \cdot 10^{-6}$ H_2S , $40 \cdot 10^{-6}$, $50 \cdot 10^{-6}$ HCl , $60 \cdot 10^{-6}$, $70 \cdot 10^{-6}$ NH_3 was reported. Also gravimetric and GC-MS tar concentrations of $7\text{--}10 \text{ g m}^{-3}$ and $12\text{--}18 \text{ g m}^{-3}$ respectively were reported referring to dry, STP and N_2 -free conditions. Gil et al. [26], conducted steam oxygen-gasification of wood chips in a silica sand bed in a facility that is comparable in size and setup to the one used in this work. Ref. [27,28] conducted steam oxygen gasification of wood at ambient pressure and absolute pressures up to 600 kPa. Increasing pressure lead to higher CH_4 yields, which is beneficial when SNG is the desired synthesis product. However, elevated pressure leads to higher facility investment costs and is not the focus of this work but can be an option for industrial application. Also

dolomite was used as bed material in that study and a promising catalytic effect of the bed material on tar cracking was observed. Circulating fluidized bed reactors have also been used in literature: In Ref. [29] peat was gasified in a demonstration plant at an absolute pressure of 1100 kPa, in Ref. [30] wood chips were gasified at 350 kPa absolute pressure and in another study distillers grain [31].

For assessment of the steam-oxygen gasification for syngas production in respect of gas quality, results from sufficient large research facilities are needed. For wood, as stated above, this is the case in literature. For sewage sludge only smaller scales and other process layouts have been investigated in literature. Very little could be found on steam-oxygen gasification of straw. Also a comparison of different fuels in the same facility could not be found. Therefore, the goal and the novelty of this work is the assessment of the product gas quality, including impurities, as derived from steam-oxygen gasification of the low cost biogenic fuels wheat straw and sewage sludge in relevant conditions with wood pellet gasification serving as a reference. Also the specific properties of sewage sludge as fuel and its consequences on the process, namely the use of ash as bed material and the different properties of sewage sludge tars, are addressed in this work.

2. Materials and methods

2.1. Experimental facility

The 20 kW fuel input fluidized bed facility used in this work is shown schematically in Fig. 1. The bubbling fluidized bed reactor has a total internal height of 3.5 m with an internal diameter of 0.15 m in the gasification zone and 0.2 m in the freeboard above. The facility is equipped with various thermocouples and pressure transducers. Electrical heating allows to control and adjust the temperature inside the gasifier's fluidized bed as desired. The fluidized bed contains between 5 and 20 kg bed material. Preheated steam is injected through six bubble cap nozzles to fluidize the bed. Another two bubble cap nozzles are used to introduce oxygen. For safety reasons, 25% nitrogen was added to the oxygen stream. It is assumed, that this nitrogen is inert. To overcome the utilization of nitrogen in an industrial plant, steam and oxygen would be mixed and introduced together into the gasifier, which was not possible in this work due to technical limitations. Fuel is dosed gravimetrically with a screw feeder into the lower part of the fluidized bed. Due to the high ash content in sewage sludge, the bed material is constantly removed during the gasification process by an overflow port to maintain a constant bed height. The overflow port was closed for wood and straw gasification. Downstream of the gasifier, the product gas is cleaned from particles by two cyclones and a candle filter, which

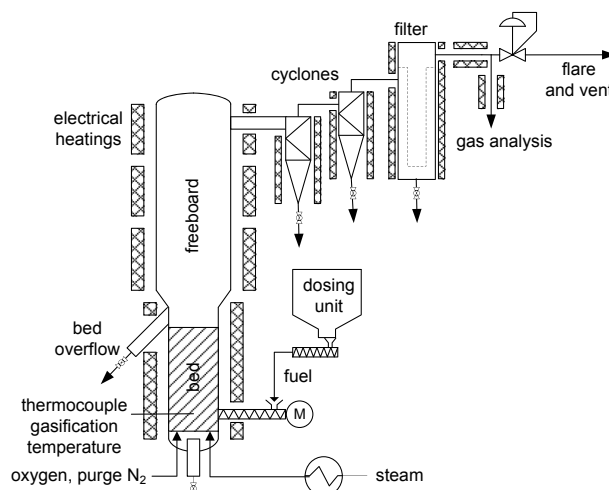


Fig. 1. 20 kW bubbling fluidized bed facility.

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