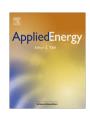
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Biomass steam gasification for production of SNG – Process design and sensitivity analysis

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ABSTRACT

A process design for small-scale production of Substitute Natural Gas (SNG) by steam gasification of woody biomass is performed. In the course of this work, thermodynamic models for the novel process steps are developed and implemented into an already existing model library of commercial process simulation software *IPSEpro*. Mathematical models for allothermal steam gasification of biomass as well as for cleaning and methanation of product gas are provided by applying mass balances, energy balances and thermodynamic equilibrium equations. Using these models the whole process is integrated into the simulation software, a flowsheet for an optimum thermal integration of the single process steps is determined and energy savings are identified. Additionally, a sensitivity study is carried out in order to analyze the influence of various operation parameters. Their effects on amount and composition of the product gas and process efficiency are evaluated and discussed within this article.

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1. Introduction

In increasingly industrialized societies the energy consumption has risen steadily in the last decades. Besides, the dependence on fossil primary energy sources is very strong. Because of limited fossil fuel resources and environmental aspects the share of renewable and sustainable energy of the overall energy demand must be increased. Therefore political strategies have been developed and targets have been defined by the European Commission [1]. It is commonly agreed, that one promising option to reach these targets is the use of biomass as a renewable energy carrier. Biomass is carbon neutral, which means that during growth process as much CO₂ is captured via photosynthesis as is released during thermal utilization and consequently net carbon emissions equal zero. Furthermore, biomass is independent from short-term availability fluctuations, unlike hydro, wind and solar power and is therefore suitable for stabilization of renewable energy systems [2]. In recent years, research and development strongly focuses on gasification of solid biomass for production of so called secondary generation fuels (SGF), like Substitute Natural Gas (SNG), Fischer-Tropsch fuels (BtL) or bio hydrogen (BioH2). Especially the production of SNG seems to be a reasonable way because the possibility to feed the SNG into an already existing natural gas infrastructure is given. Further the SNG process is characterized by high conversion efficiency [3].

The production of SNG via gasification of solid biomass and subsequent methanation of the synthesis gas requires a gasifier generating a producer gas with a suitable composition (low nitrogen, high hydrogen). Therefore, mostly allothermal steam gasification technologies are used. Due to the spatial separation of endothermic gasification and exothermic combustion there will be no mixing of the combustible producer gas with the flue gas respectively atmospheric nitrogen. Thus, a virtually nitrogen free, medium calorific value producer (respectively synthesis gas) with high hydrogen content, well suited for SNG production, can be generated using a cheap and technically simple gasification agent without the need for an air separation unit. In principle, also autothermal gasification using pure oxygen as gasifying agent would be suitable. However, air separation for generation of oxygen is expensive and therefore in general economically viable only for large-scale applications. Allothermal steam gasification therefore constitutes the technology of choice for small to middle scale applications, which are mainly considered for biomass due to its local distribution and availability limitations. Several projects deal intensively with the production of SNG, e.g. the EU-project "BioSNG" [4]. In the combined heat and power (CHP) plant in Güssing (Austria) [5,6] a 1 MW pilot and demonstration unit for the production of BioSNG was installed and successfully proven. The process of the Güssing CHP plant, as well as other present concepts, is based on atmospheric gasification and commercial cold gas cleaning. Thus, it has to be operated in the power range of 10-25 MW to become economically profitable.

The combination of pressurized gasification and hot gas cleaning entails a clear process simplification and significant efficiency

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Nomenclature filter surface (m²) R universal gas constant ([mol⁻¹ K⁻¹) Α isobaric heat capacity (J $kg^{-1} K^{-1}$) filter thickness (m) S \vec{d}_p $h_{f,298}^0$ relative sensitivity coefficient (-) sauter-diameter (m) $S_{i,rel}$ standard enthalpy of formation at 298,15 K (J kg^{-1}) Ť temperature (K) V flow rate $(m^3 s^{-1})$ variable for the operation parameters (-) K_p thermodynamic equilibrium constant (-) viscosity coefficient (m2) α inertia coefficient (m) length of the packed bed (m) ΔG_R^0 LHV lower heating value (kJ kg⁻¹) Gibbs free enthalpy of reaction at standard pressure total carbon input into the reformer (kg s^{-1}) $(J \text{ mol}^{-1})$ $\dot{m}_{C,fuel}$ residue carbon leaving the reformer $(kg s^{-1})$ $\dot{m}_{C,res}$ ⊿p pressure drop (Pa) pyrolysis char input into the reformer (kg s^{-1}) dynamic viscosity (Pa s) $\dot{m}_{pyr,waf}$ η residue char leaving the reformer (kg s⁻¹) $\dot{m}_{res.waf}$ η_{chem} cold gas efficiency (%) standard pressure (bar) overall thermal utilization rate (%) η_{ges} $P_{el,C}$ electric power consumption (kW) thermal process efficiency (%) η_{th} heating rate of excess steam (kW) air excess ratio (-) P_{EH} λ $p\delta_{eq}$ logarithmic deviation from thermodynamic equilibrium stoichiometric coefficient of species i (-) v_i (-)fluid density (kg m⁻³) ρ_f steam excess ratio (-) partial pressure of species i (bar) σ p_i partial pressure of species i at thermodynamic equilibν superficial velocity (m s⁻¹) p_i^* rium (bar) porosity (–)

increase by which a decentralized SNG production is allowed in small arrangements [7]. Furthermore, the pressurized operation is favorable for subsequent methanation. Pressurized gasification is enabled by the application of the Biomass Heatpipe Reformer (HPR), an innovative allothermal steam gasification technology [8,9]. Main parts of the HPR are a combustion chamber and a reformer, both designed as bubbling fluidized bed reactors. Heat is transferred from the combustor into the allothermal reformer using so called Heatpipes. Heatpipes are enclosed metal pipes containing an alkali metal working fluid (e.g. Na, K). This working fluid is evaporated in the area of the exothermic combustion chamber fluidized bed and then condensed in the area of the endothermic gasification fluidized bed. The resulting elevated heat transport density enables complete decoupling of combustion and gasification and therefore pressurized operation of the reformer. This allows compact reactor design and integration, what leads to economic and energetic efficiency comparable to the performance of large systems [10]. A 500 kW_{th} demonstration plant of the Biomass Heatpipe Reformer has been in operation in Pfaffenhofen (Germany) over the past two years showing stable operation [10]. Further, a commercial sized CHP application with a thermal power input of 1.3 MW_{th} is currently under construction in Grassau (Germany).

The objective of the work presented in this article is the performance of a process design for production of SNG using a combination of pressurized gasification with the HPR, hot gas cleaning and methanation. Therefore, mathematical models for the single process steps are required. Mathematical models can be divided into thermodynamic equilibrium and kinetic rate models. Kinetic rate models provide detailed information on the analyzed process and can give accurate results. But however, they are computationally intensive and always contain parameters which make them applicable only for one particular reactor design. In contrast, thermodynamic equilibrium models are computationally less intensive, independent of reactor design and therefore more convenient for process studies. Moreover, equilibrium models predict thermodynamic limits of performance under different conditions. This makes these models a useful tool for process design and optimization. Hence, many applications of equilibrium models are reported in literature [11-15]. A detailed review and analysis of biomass gasification models is given in [16].

The presented article is organized in three parts. In the first part, thermodynamic equilibrium models developed for the SNG process will be described. A validation of the newly developed HPR model is performed within the second part. In the third part, results of process simulation are shown. Energy savings have been identified and an optimum thermal integration for the overall process has been determined. Furthermore, a comprehensive sensitivity analysis has been carried out to show the influence of the most important operation parameters.

2. Process scheme

A flow chart of the SNG production process is shown in Fig. 1. Gasification occurs in the Biomass Heatpipe Reformer (HPR), which is a patented development of the Technical University of Munich [17]. As gasification agent superheated steam is used. Based on the allothermal operation, the HPR produces a nearly nitrogen-free, high calorific value syngas. Because of the ideal $\rm H_2/CO$ ratio and the pressurized operation, the generated gas is well suited for subsequent methanation.

Methanation catalysts are very sensitive against catalyst poisoning. Thus, an intensive gas cleaning is required. In a first step, solid particles are separated with the help of a cyclone. The separated particles are fed back into the combustion chamber of the HPR. In the next step, tars are removed from the syngas by catalytic reforming on a nickel based catalyst. Before the alkali elements can be removed, it is necessary to cool down the syngas below the condensation temperature of the alkali elements. After cooling the alkalis are removed in a filter cartridge. Sulfur leads to poisoning and deactivation of the methanation catalyst and therefore it is captured by chemical adsorption on a copper-oxide sorbent. The methanation itself is carried out in a fixed-bed reactor with a nickel based catalyst. Since the methanation reaction is strongly exothermic, cooling of the reactor is inevitable. After methanation, the raw-SNG has to be dried and cleaned from carbon dioxide and hydrogen to comply with the demands of the natural gas grid.

While gasification of the biomass is endothermic, syngas and flue gas cooling as well as methanation are exothermic. Therefore, an optimal process design and thermal integration plays a major role in the performance and efficiency of the overall process.

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