Integration study for alternative methanation technologies for the production of synthetic natural gas from gasified biomass

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This paper analyzes the integration of two different methanation technologies – fixed bed adiabatic and fluidised bed isothermal - in a SNG production process and the consequences for the overall process energy conversion performance. The different operating conditions of the two methanation technologies lead to a change in temperature levels and quantities of recoverable heat, respectively, but also to differences in the overall processes' power consumption. Using pinch methodology for optimal internal heat recovery in combination with flowsheeting software (ASPEN Plus), the two methanation alternatives are fitted into the SNG production process. The potential power production from recovered process heat is analysed based on the Carnot efficiency and compared to the overall power consumption within the SNG process. Both methanation alternatives perform equally within the given boundary conditions, resulting in an output of SNG of 63.3 MW_{LHV} per 100 MW_{LHV} dry fuel input and a ratio of about 1.22 between theoretical power production and overall power consumption.

1. Introduction

1.1 SNG production as transportation fuel

Synthetic natural gas (SNG) from biomass is among the promising alternatives of second-generation biofuels, mainly aiming at an effective reduction of CO_2 emissions within the transport sector. SNG can be readily blended with natural gas and there are a number of off-the-shelf technologies it can be used with. Its potential advantages compared to other synthetic transportation fuels such as DME or ethanol are the use of the existing natural gas infrastructure allowing for a smooth transition from fossil to synthetic natural gas, as well as the large number of existing end-use applications. The predicted overall thermal efficiency from biomass to SNG on a lower heating value basis ranges between 55-75% for different studies (Mozaffarian and Zwart, 2003, Duret et al., 2005, Heyne et al., 2008, Gassner and Maréchal, 2009).

1.2 Methanation technologies

A major process step within the production of SNG from biomass – besides gasification – is the conversion of the product gas to methane. Basically, two reactor principles have been developed for methanation; a comprehensive review of the development has been

recently published by Kopyscinski et al. (2010). Methanation is either carried out in a series of adiabatic fixed bed reactors with inter-cooling and optional product recycle (e.g. TREMP or Lurgi methanation), or in a single fluidised bed reactor at isothermal conditions (e.g. COMFLUX methanation). Commonly used catalysts are Ni-based.

Due to the recent interest in biomass gasification for production of SNG the methanation technology has been adapted to this new feedstock material, accounting for smaller process scales, different syngas composition and other impurities (e.g. organic sulphur) compared to coal-based SNG generation (Kopyscinski et al., 2010). The Paul-Scherrer-Institute (PSI) in Switzerland has adopted the isothermal fluidised bed methanation technology for atmospheric operation in a once-through reactor and successfully proven operation at pilot-scale in the Güssing biomass gasification plant (EU project, 2009).

1.3 Objectives of this study

Within this study, the influence of choice of methanation technology on the overall SNG production process heat balance is analysed using pinch technology. The theoretical potential for the conversion of excess process heat to power is depicted using Carnot efficiency curves for the overall process depending on the methanation technology. The potential for power generation is compared to the overall SNG process power consumption for each alternative.

2. Methodology

2.1 Methanation modelling

The two methanation technologies have been modelled and simulated with ASPEN Plus at different conditions. The flow sheets for the two technologies are represented in Figs. 1 and 2. The unit modelling assumptions are detailed in Tab. 1.



Figure 1: Adiabatic fixed bed methanation with inter-cooling and recycle (TREMP technology); H1 & H2: heater, C1-C4: cooler, R1-R3: adiabatic methanation reactor; (Flow sheet adapted from Haldor Topsøe (2009) and Harms et al. (1980)).

For the fixed bed methanation, the reactor inlet temperatures are set to 300°C for R1 and 250°C for R2 and R3, respectively. The recycle ratio is set to limit the outlet temperature

of R1 to 650°C in order to avoid catalyst sintering. The CO conversion for the methanation step is required to be above 99.9% in order to obtain a low CO content in the methane rich gas. The minimum pressure prior to the methanation achieving the required CO conversion was determined to be 18 bar. At lower pressures, the CO conversion is not satisfactory and the amount of high temperature heat recoverable from coolers C1 and C2 lower. Higher pressures in contrast are penalised by an increased compression work. The pressure was therefore kept at 18 bar in this study.

The fluidised bed methanation is assumed to operate at 300°C according to experimental data from Seemann (2006). The inlet pressure for the methanation reactor is set to 2.5 bar. The lower pressure is possible due to the isothermal operation reaching higher CO conversion values fulfilling the required CO conversion of 99.9% according a sensitivity analysis conducted.



Figure 2: Isothermal fluidised bed methanation (COMFLUX technology); H1 & H2: heater, C1: cooler, R1: isothermal methanation reactor; (Flow sheet adapted from Friedrichs et al. (1982)).

Table 1: Unit modelling assumptions for methanation.

Unit	Modelling assumptions	
Methanation reactors	- Gibbs equilibrium reactor (both technologies very close	
	to equilibrium in reality (Kopyscinski et al., 2010))	
	- $\Delta P = 0.5$ bar, $Q_{loss} = 0$ kW	
	- stoichiometric amount of steam added in order to obtain	
	H ₂ /CO ratio of 3 via water-gas-shift reaction	
Compressor	- single $(P_{out}/P_{in} < 5)$ or multi-stage compressor with	
	intercooling to 100 – 120 °C, last stage no cooling	
	- stage isentropic efficiency: 0.72	
Heat exchanger/Flash tank	- $\Delta P = 0$ bar, $Q_{loss} = 0$ kW	

2.2 Process integration

The two methanation technologies are analysed for their thermal integration with a SNG process via indirect gasification of biomass. The process flow sheet is presented in Fig. 3. The basic modelling assumptions for the process can be found in Heyne et al. (2008). The proposed SNG production process used an equilibrium model with temperature approach at isothermal conditions for the methanation modelling resembling the isothermal methanation presented in this study.



Figure 3: Simplified SNG production process flowsheet. C and H indicate syngas coolers and heaters, respectively. The heat demand/excess of the different process steps is not indicated in the figure.

The second methanation step indicated in Fig. 3 actually only serves for ensuring removal of eventual traces of CO left in the methane rich gas and its influence on the overall process heat balance is negligible. The integration study conducted here therefore focuses on the first methanation step.

The implementation of the two alternative technologies is evaluated using the resulting heat stream balance from the overall SNG process modelled in the flowsheeting software ASPEN Plus as basis for a pinch analysis. The potential for power production is evaluated based on the Carnot representation of the resulting grand composite curves – as developed by Linnhoff (1989) – serving as a measure of the maximum amount of power that could theoretically be produced from the excess process heat. This potential then is related to the actual power consumption of process alternatives that differs due to the different pressure levels the methanation technologies are operated at. The final delivery pressure for SNG is set to 60 bar, representing an average value for natural gas distribution network pressure.

3. Results

For both processes the absolute amount of heat available from the methanation process including the cooling of the methane rich gas is about 12.7 MW. However, the temperature level for the heat release differs to some extent as illustrated in Fig. 4, showing the Grand Composite Curves on a Carnot efficiency base ($T_{ref} = 20^{\circ}$ C). The shaded area in both curves represents the maximum amount of power that theoretically can be extracted from the heat streams via a Carnot process.



Figure 4: Carnot representation ($T_{ref} = 20^{\circ}C$) of the resulting grand composite curves for the SNG processes. Left: isothermal fluidised bed methanation. Right: adiabatic fixed bed methanation.

This amount can be related to the actual power consumption of the SNG production process with the respective methanation technology. Table 2 gives some performance indicators for the two alternative SNG production processes. For the adiabatic fixed bed methanation technology, both the potential for power production as well as the actual power consumption are higher compared to the isothermal methanation alternative. This results in about the same ratio between power production potential and actual power consumption for the given boundary conditions.

Parameter	Isothermal	Adiabatic fixed bed
	methanation	methanation
Dry biomass fuel input [MW _{LHV}]	100	100
SNG production [MW _{LHV}]	63.3	63.3
Carnot-based power production potential [MW]	6.57	8.28
Process power consumption [MW]	5.45	6.75
ratio power production potential/power consumption [-]	1.21	1.23
specific power production potential [kW/MW _{SNG,LHV}]	103.8	130.8
specific power consumption [kW/MW _{SNG,LHV}]	86.1	106.6

Table 2: Performance data for the SNG process using the two alternative methanation technologies.

4. Discussion

The higher operation pressure of the fixed bed methanation technology requires a compression of a larger gas flow prior to methanation, thereby resulting in a higher power consumption of this SNG production process alternative. This though is compensated for by better opportunities for power generation due to a heat release at higher temperature levels compared to the isothermal fluidised bed methanation operating at 300 °C. Both the SNG production and the ratio between theoretical power production and actual power consumption do not differ significantly for both

technology alternatives. The simpler process setup of the isothermal fluidised bed methanation process combined with its good resistivity to carbon deposition (Seemann, 2006) makes it more suitable for smaller scale applications (up to 100 MW_{th} biomass fuel input), while at large scale both techniques are applicable with fixed bed methanation already being commercially applied for coal-to-SNG conversion processes.

5. Conclusions

Two methanation technologies have been compared for their performance within the production process of SNG from biomass via gasification. The analysis showed a similar SNG production and ratio between the theoretical power production from process heat and the actual power consumption of the overall process. This makes both techniques equally applicable from a process integration perspective within the given boundary conditions.

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