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Integrated design of a gas separation system for the upgrade of crude SNG with membranes

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ABSTRACT

This paper investigates different design strategies and process layouts for upgrading crude synthetic natural gas (SNG) produced from lignocellulosic biomass to grid quality. The design problem is outlined by studying the involved key aspects with respect to the crude composition, purity requirements and process integration. A discussion of candidate technology identifies multistage membrane processes as a promising option, for which a multicomponent thermo-economic design model is coupled to a process model for SNG production. In a design study using multi-objective optimisation, the most promising membrane configurations are identified and optimised with respect to cost and efficiency. Comparing design strategies that consider different levels of process integration, it is shown that coupling the reactive and separation sections improves the process design. If process integration is not considered, the separation system is oversized by up to 60% and its investment cost is up to 46% too high. In the last part of the paper, the by-production of biogenic CO_2 at sufficient purity for storage is investigated, which would turn the process into a CO_2-sink for the atmosphere. The cost of captured CO_2 is assessed between 15 and 40 €/ton, which makes it potentially more advantageous than the capture at a fossil fuel power plant.

1. Introduction

The production of fuels from biomass and waste is considered as an important contribution for mitigating climate change by reducing greenhouse gas emissions. Contrary to biological processes like biomethanation or ethanol fermentation, thermochemical processes allow for a complete conversion of lignocellulosic materials by gasification. Among the candidate liquid and gaseous synthetic fuels, methane is one of the most promising options since the synthesis reaction approaches chemical equilibrium and its conversion efficiency is high and less exothermic than the one of liquid fuel. Distributed as synthetic natural gas (SNG) in the existing natural gas grid, it can be used as transport fuel in an increasingly dense network of fuel stations.

While several recent studies have investigated suitable technology and processes for SNG production [1–6], the issue of the gas upgrading to grid quality has not received much attention. The separation of carbon dioxide from methane is considered as a conventional operation in the removal of sour gas from natural gas in petrochemical applications, and relatively mature technology for the upgrade of biogas is available [7]. For SNG production, different candidate technologies have been identified and some basic performance data and simple phenomenological models have been reported for physical absorption [1,3], chemical adsorption [4], membrane permeation [2] and pressure swing adsorption (PSA) [3]. However, the detailed design of such a system needs to adapt not only to the quality and impurities of the crude product, but can also exploit the advantages of process integration. The present paper therefore investigates different design strategies for upgrading crude SNG to grid quality. Through the example of gas separation with membranes, it aims at showing in particular the benefits of a holistic design approach that considers a tight integration of the separation system with the reactive sections of the process. A suitable thermo-economic model of the membrane system is presented and coupled to a process model for crude SNG production. Multi-objective optimisation is used to compare different design approaches and optimal system layouts, and operating

Abbreviations: CC, countercurrent; CCS, carbon capture and storage; CMS, carbon molecular sieves; EOR, enhanced oil recovery; FICFB, fast internally circulating fluidised bed; PSA, pressure swing adsorption; rec, recycle; SNG, synthetic natural gas; TSA, temperature swing adsorption.

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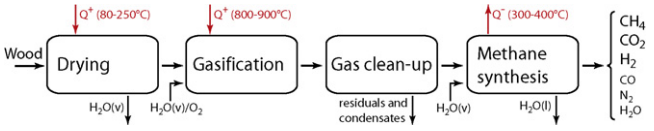

Fig. 1. Block flow diagram of crude SNG production from wood.

conditions for processes with and without CO_2-capture are proposed.

2. Process design problem

2.1. Production of crude SNG

Representing lignocellulosic biomass as a chemical molecule with the carbon atom as reference, the conceptual design of its conversion into methane is based on reaction (1):

$$CH_{1.35}O_{0.63} + 0.3475\,H_2O \rightarrow 0.51125\,CH_4 + 0.48875\,CO_2,$$
$$\Delta\tilde{h}_r^0 = -10.5\,\mathrm{kJ\,mol}_{\mathrm{wood}}^{-1} \quad (1)$$

Technically, the currently preferred route is to carry out the conversion in two reactive steps. As depicted on Fig. 1, the production of SNG from lignocellulosic biomass consists in gasifying the biomass and converting the producer gas into methane. Prior to gasification, the raw material is dried to below 20–25 wt% humidity in order to prevent excessive losses due to water evaporation. Due to the presence of dust, tars and catalyst poisons like sulphur compounds, the producer gas is then cleaned before entering the methane synthesis. The producer gas is then converted to methane in a catalytic fluidised bed reactor operated at 300–400 °C and requires upgrading before being fed to the grid. A comprehensive description and comparison of the different technological options for gasification and methanation are given by Mozaffarian and Zwart [1]. From the current state of research and process development [8], it is expected that the first installations will be based on indirectly heated fluidised bed gasification technology of FICFB-type that has been developed and commercialised by Hofbauer et al. [9].

Using a detailed process model developed in previous research [2,3], expected gas compositions of the producer gas and the crude methanation product for this technology are reported in Table 1. The corresponding properties of the feed and the process conditions considered for this base case are given in Tables 2 and 3, respectively. Depending on the operating conditions of the methanation reactor, cold, crude SNG contains around 50 vol.% of methane and 45 vol.% carbon dioxide. Especially at low pressure where the conversion is limited by thermodynamic equilibrium, a non-negligible residue of hydrogen and traces of carbon monoxide remain in the gas. According to Rauch [10], up to 5 vol.%[1] of nitrogen is furthermore present in the producer gas due to its use for the inertisation of the gasification feed and some slip from the adjacent combustion chamber. Since the methanation reaction reduces the volume of the reactants, the molar fraction of the inert species increases, which would prevent to meet the grid specifications for SNG without a special removal of nitrogen. In this work, it is assumed that this issue can be resolved by using CO_2 for feed inertisation and taking special care of nitrogen in an improved gasifier design, which allows for attaining a nitrogen content of 0.5 vol.% in the dry producer gas.

The stoichiometric design equation (Eq. (1)) shows that the overall conversion of wood to methane is exothermic and releases about 450 kJ/kg_{wood} of heat. The block flow diagram (Fig. 1) illustrates, however, that this net release is distributed over the gasification and methanation reactions. The first one is endothermic and requires heat at high temperature, whereas the second one is exothermic and releases heat at lower temperature. The process thus requires additional energy and the quality of the process integration will define the overall process efficiency. The grand composite curve of the process streams shown in Fig. 2 highlights that the process is pinched at the gasification temperature and that 200–250 kW of high temperature heat at 850–900 °C are typically required to convert 1 MW[2] of wood to crude SNG.

Table 1
Composition of producer gas and crude SNG as calculated by the process model (vol.%).

	C_2H_4	CH_4	H_2	CO	CO_2	N_2	H_2O
Gasification (FICFB, 1123 K)							
Hot	1.9	9.2	32.8	21.5	15.7	0.4	18.5
Cold	2.2	10.9	38.8	25.5	18.6	0.5	3.5
Methanation (1 bar, 593 K)							
Hot	–	26.7	3.6	0.1	27.1	0.6	41.9
Cold	–	44.8	5.9	0.1	45.1	1.0	3.1
Methanation (10 bar, 593 K)							
Hot	–	27.6	1.2	0.0	26.9	0.6	43.7
Cold	–	52.1	1.7	0.0	44.7	1.2	0.3

Table 2
Proximate and ultimate analysis of the feedstock.

Proximate analysis		Ultimate analysis	
$\Delta h^0_{\mathrm{wood,dry}}$ [a]	18.6 MJ/kg_{dry}	C	51.09 wt%
Φ_{wood}	50.0 wt%	H	5.75 wt%
		O	42.97 wt%
		N	0.19 wt%

[a] Δh^0 is defined on dry basis, and thus independent of the humidity.

Table 3
Nominal operating conditions of the process.

Section	Operating conditions	Value
Drying	Inlet temperature	473 K
	Outlet wood humidity	20 wt%
Gasification	Pressure	1 bar
	Gasification temperature	1123 K
	Steam preheat temperature	573 K
	Steam/dry biomass ratio	0.5
Methanation	Pressure	1 bar
	Inlet temperature	593 K
	Outlet temperature	593 K

2.2. Gas grid specifications

According to the new Swiss directive for the supply of biogas (also applying to synthetically produced gas) to the natural gas grid, unlimited amounts of gas can be fed in if its methane content is higher than 96 vol.% and the CO_2, H_2 and CO content less than 6, 4 and 0.5 vol.%, respectively [11]. Among other conditions, it is furthermore required that the dew point of the gas at grid pressure ($\leq$ 70 bar) is lower than −8 °C. The limit with respect to methane content is thereby based on the fact that biogas as a binary mixture of methane and carbon dioxide does not meet a Wobbe index between 13.3 and 15.7 kWh/Nm^3 if its methane content is below 96 vol.%, which is the usual norm for H-quality natural gas.

[1] From the given composition, a typical value of 2.9 vol.% is computed by difference.
[2] Based on the lower heating value of dry wood.