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Sorption enhanced gasification (SEG) of biomass with CO_2 capture for the production of Synthetic Natural Gas (SNG) and DME for transport sector with negative emissions

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Abstract

In this work, a process assessment of two synthetic fuel production plants based on a flexible Sorption Enhanced Gasification (SEG) of biomass is carried out, one focused on Synthetic Natural Gas (SNG) and the other on dimethyl ether (DME) production. Mass and energy balances of these two synthetic fuel production plants have been solved and their performance has been compared. The calculated biomass-to-fuel equivalent cold gas efficiency CGE_{eq} (i.e. accounting for energy credits/debits associated to electricity export/import) ranges from 53.8% of the bio-SNG plant to almost 57% of the bio-DME plant. The WTW greenhouse gases emission saving of a bio-DME vehicle in comparison with a conventional diesel vehicle is around 100 g CO_2-eq/km. The assessed plants are also able to capture 63-67% of the inlet biomass carbon, equivalent to 155 g CO_2-eq/km removed from the atmosphere and stored.

Keywords: biomass; gasification; enhanced; Dimethyl-ether (DME); synthetic natural gas (SNG)

1. Introduction

Synthetic biofuels production from renewable energy sources like biomass has an important role to play in the decarbonized energy mix scenario needed in the coming years for fulfilling the 1.5ºC target. Indirect gasification is one of the generally proposed technologies for biofuel production from biomass due to its advantage of producing a N_2-free syngas with inherent CO_2 separation [1]. Coupling the indirect gasification with *in-situ* CO_2 separation results in an intensified and flexible process, which will be developed in the H2020 EU project FLEDGED [2].

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2. Process description and assumptions

The block diagram of the DME and SNG production plants considered in this work is depicted in Fig.1. Biomass pretreatment and drying, sorption enhanced gasification (SEG), syngas cleaning and conditioning and steam cycle sections are common to the two cases. Main differences can be found on the synthetic fuel production island. Mass and energy balances of both plants have been calculated in Aspen Plus using the RKS-BM equation of state and following the assumptions discussed in the following sections. As a common basis for the calculations, 100 MW_{LHV} of biomass thermal input has been considered in both cases. A typical woody biomass has been considered with a moisture content of 45% and a LHV equal to 9.75 MJ/kg biomass a.r. (i.e., ‘as received’). The ultimate analysis (on weight dry basis) is 51.19% C, 6.08% H, 0.2% N, 0.05 % Cl, 41.3% O, 0.02% S and 1.16 % ash. Fixed carbon is $18.84\%_{dry}$.

2.1. Sorption enhanced gasification

Biomass is first dried in a tube bundle drier, where humidity content is reduced to 20 % using condensing steam at 6 bar (see Fig.1). Dried biomass at 80ºC is then introduced into the gasifier/carbonator of the SEG process. Slightly superheated steam at 1.5 bar is also fed into the gasifier with the biomass, with a steam-to-carbon (S/C) molar ratio of 1.5. A solid stream containing CaO and ashes from the combustor/calciner operating at 910 ºC also enters the gasifier, with the aim of reacting with the CO_2 formed from gasification and supplying the energy needed for the endothermic gasification, i.e. keeping the gasifier autothermal. Gasification and carbonation of CaO (i.e., $CaO_{(s)}+CO_{2(g)} \rightarrow CaCO_{3(s)}$) reactions occur in the gasifier. Balances in the gasifier have been solved following the 0-D model structure in [3]. The distribution of C, H and O atoms among CO, CO_2, H_2 and H_2O is calculated assuming a deviation from the equilibrium for the water gas shift (WGS) reaction, as proposed in [4]. CH_4 and higher hydrocarbons in the syngas and the unconverted char leaving the gasifier have been calculated through empirical expressions that depend on temperature (except for CH_4) that have been regressed from the SEG experiments carried out at the 200 kW_{th} pilot plant at the University of Stuttgart [5]. Solids circulation and therefore gasifier temperature are tuned to obtain the target syngas M module ($M=(H_2-CO_2)/(CO+CO_2)$) at the inlet of the fuel synthesis section, which is M=3 or M=2 for SNG and DME synthesis respectively.

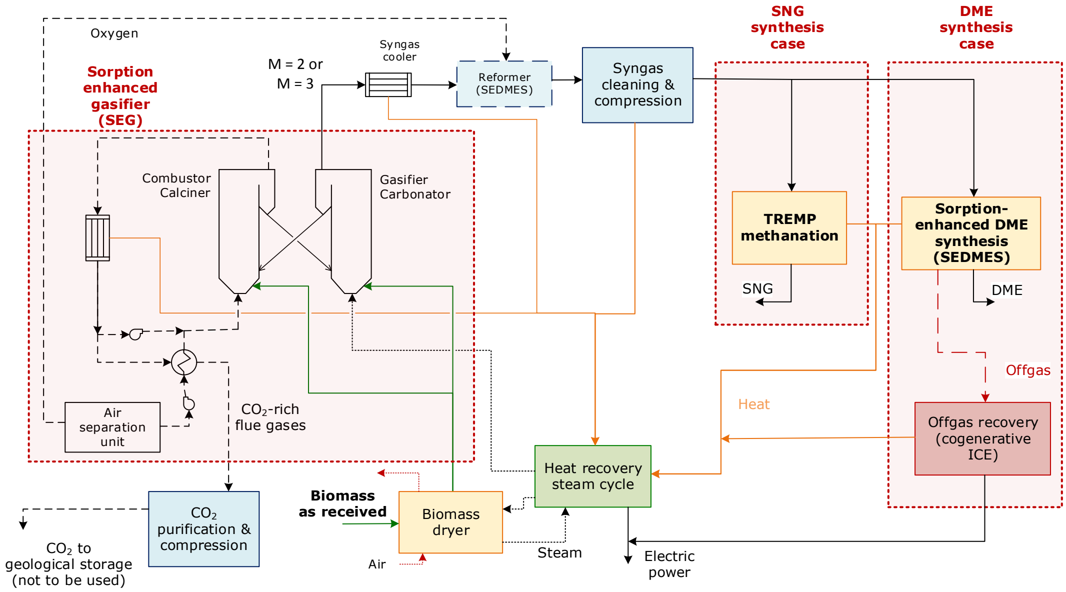

Fig. 1. Block diagram of the DME and SNG production plants assessed in this work.

Solids exiting the gasifier contain unconverted char, $CaCO_3$, CaO and ashes and are sent to the combustor/calciner where char is burnt and $CaCO_3$ calcined to CaO. When unconverted char is not enough for achieving the target combustor temperature, a fraction of the biomass is burnt in the combustor/calciner, keeping the total biomass input to the plant constant. Pure O_2 from an air separation unit (ASU) is used as oxidant and it is mixed with a cooled CO_2-rich gas recycle from the combustor/calciner outlet to achieve 30 %vol. O_2 at this reactor inlet. A specific consumption 240 kWh_{el}/ton_{O2} is assumed for the ASU. In this way, a CO_2-rich gas is generated which is cooled down, purified and compressed in a five stage intercooled compressor, with an assumed specific consumption of 110 kWh_{el}/ton_{CO2}.

2.2. Syngas cleaning and conditioning and biofuel production

In both plants, syngas cleaning and conditioning comprises solid particle removal, a caustic washing unit to remove soluble contaminants (i.e., HCl, NH_3) and a liquid redox process for bulk removal of H_2S. Syngas is then compressed and fed to activated carbon and ZnO bed for the removal of trace contaminants and of H_2S to ppb levels.

In the DME production plant, a high temperature filtration unit and a catalytic reformer are introduced downstream the gasifier to reduce the CH_4 content of the syngas that is an inert for the DME production process. This reformer is operated in auto-thermal mode feeding a fraction of the O_2 from the ASU to keep a reformer outlet temperature of 850 ºC. The reformer is also assumed to fully decompose tars, so that a dedicated tar removal unit is not needed in the SEDMES production plant. On the contrary, in the SNG production plant, tars are removed after partial syngas cooling by oil scrubbing in a regenerative OLGA process.

The DME synthesis process consists of a first conventional DME reactor followed by a sorption-enhanced DME synthesis (SEDMES) step, where DME is produced in the presence of a water adsorbent that removes the water formed from methanol dehydration, significantly increasing the DME yield per-pass [6]. This particular sorption enhanced DME synthesis process is also being developed within the FLEDGED project [2]. Both DME reactors operate at 25 bar and 240 ºC. A single pass DME yield (DME output moles referred to the inlet carbon) of 90% has been assumed. For the SNG synthesis step, the TREMP methanation process analyzed in [3] is chosen, which consists of four fixed bed catalytic reactors placed in series working at a pressure of 35 bar. The final purity reached for the CH_4 product is about $90\%_{vol}$ (the main impurity being N_2 used as sealing gas in the gasifier). In both plants the excess heat from gas and reactors cooling is recovered by generating steam at 480ºC and 40 or 28 bar (for the SNG and DME plants, respectively) and produce electricity.

3. Results discussion

Table 1 summarizes the results obtained for the synthetic fuel production plants assessed. The global cold gas efficiency (CGE) of the plant is calculated as the chemical energy content of the final product with respect to the input biomass on LHV basis. The SNG plant results in CGE of 60.1 %, which is larger than that of the DME plant (i.e., 53.7%) due to: (i) the presence of the reformer right after the SEG process, (ii) the need of operating the gasifier at higher temperature, which turns into a larger fraction of biomass sent to the combustor and higher sensible heat losses, and (iii) the heating value of the off-gas due to the limited carbon-to-DME yield (90%). Regarding the electric production, the use of the DME purification off-gas as a fuel in a CHP internal combustion engine for further electricity production reduces the net electricity import of the plant from 4.0 MW_{el} to 1.4 MW_{el}. This indirect fuel consumption for electricity production is taken into account by the CGE_{eq}, which is defined according to Eq.(1). The CGE_{eq} considers the fuel saving (or the additional biomass consumption) associated to the electric output (or input) of the plant (i.e., $P_{el,net}$, positive in case of net power output). To account for this indirect fuel consumption a reference biomass fed steam cycle with 34% of net electric efficiency is considered as power generation technology.

$$CGE_{eq} = \frac{\dot{m}_{DME/SNG} \cdot LHV_{DME/SNG}}{\dot{m}_{biomass} \cdot LHV_{biomass} - \frac{P_{el,net}}{\eta_{el,ref}}} \qquad (1)$$

Accounting for the electric power balance, the CGE_{eq} becomes 51.6% in the biomass to DME plant and 53.8% in the biomass to SNG plant.

Regarding the CO_2 capture ratio (i.e. fraction of inlet carbon separated in the flue gases and sent to compression and storage), the SNG production plant shows a higher one (67.7%) compared to the DME plant (63.0%) since the amount of CO_2 separated in the gasifier is larger to achieve the higher M module in the syngas. The remaining carbon is not directly emitted but it is chemically bound in the final fuel, and generates emissions when combusted in an engine.

Table 1. Main results obtained for the DME and SNG plants assessed.

	SNG-SEG production plant	DME-SEG production plant
	S/C = 1.5	S/C=1.5
Gasifier temperature [ºC]	703.4	716.2
Biomass input to the gasifier [%]	97.2	94.9
CGE_{SEG} [%]	69.0	63.5*
CGE [%]	60.1	53.7
Steam turbine electric output [MWe]	7.3	7.3
Off-gas engine electric output [MWe]	-	3.4
Net plant electric output [MWe]	-4.0	-1.4
CGE_{eq} [%]	53.8	51.6
CO_2 capture ratio [%]	67.7	63.0

* *calculated considering the syngas LHV at the reformer outlet*

The overall energy efficiency and emissions have been finally compared through a simple Well-to-Wheels balance. Data from the JRC study [7] are used for the energy and carbon balances of the different value chains. Resulting consumptions and emissions for fuel production and for 100 km driving range of the vehicle are presented in Table 2, in comparison with conventional diesel and gasoline. Despite the similar figures in terms of efficiency and primary energy consumption with respect to the conventional synthesis from biomass, the capability of carbon capture yields much stronger reduction in global carbon emissions.

Table 2. Well-to-Wheel performances of SNG and DME produced through SEG of biomass, benchmark values from [7].

	WtT (fuel production)				**TtW (fuel use)**			**WtW**				
								Primary energy	Emissions			
	Primary source	*MJ_P/MJ_F*	*gCO_2,eq/MJ_F emitted**	*gCO_2,eq/MJ_F stored*	*gCO_2,eq/MJ_F*	*MJ_F/100km*	*gCO_2,eq/km*	*MJ_P/100km*	*gCO_2,eq/MJ_F emitted*	*gCO_2,eq/MJ_F stored*	*gCO_2,eq/km emitted*	*gCO_2,eq/km stored*
Diesel	Oil	0.20	15.1	-	73.6	119	88	143	89	-	106	-
Gasoline	Oil	0.19	14.0	-	73.9	150	111	178	88	-	132	-
Conventional CNG	Natural gas	0.16	13.2	-	57.3	152	87	178	71	-	107	-
Bio-CNG from SEG	Woody biomass	0.83	-46.9	-119	55.0	152	84	278	8.1	-119	12.3	-181
Conventional bio-DME	Woody biomass	1.05	-64.9	-	67.3	122	82	250	2.4	-	2.9	-
Bio-DME from SEG	Woody biomass	1.01	-62.1	-127	67.3	122	82	245	5.2	-127	6.3	-155
* for biofuels, the value is the difference between the process emissions and the CO_2 removed from the atmosphere												

In fact, from the point of view of WTW emissions, fossil fuel based value chains are positive emitters, while the conventional DME one is almost carbon neutral. In addition, the processes investigated in this work have very low WTW emissions, to which, however, the potential to store about 155 g CO_2-eq/km is added. This lead to an overall negative balance of about -150 g CO_2-eq/km. Bio-CCS in fuel synthesis allows therefore not only decarbonized mobility, but also 'carbon negative' mobility, i.e. with a net carbon removal from the atmosphere. The energy efficiency of such processes is intrinsically lower than the efficiency of fossil fuel based ones, but most of the energy input is from a renewable source. Furthermore, compared to the conventional biomass-to-DME process, the additional CCS step in SEG processes does not lead to a significant increase of energy consumption, yielding similar WTW energy performance and making the processes energetically competitive.

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