Thermodynamic Investigation of SNG Production Based on Dual Fluidized Bed Gasification of Biogenic Residues

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Abstract

Natural gas is an important commodity in the European energy market. A promising concept for the production of synthetic natural gas on a carbon neutral basis is presented by the gasification of biogenic residues and the further reaction to a methane-rich gas. This paper investigates the thermodynamics of methanation for different product gas compositions of the dual fluidized bed gasification technology. A complete methanation of the carbon oxides is possible by the utilization of a product gas from the sorption enhanced reforming process. For product gases from conventional or carbon dioxide gasification, only partial methanation of carbon monoxide occurs. Additionally, proper handling of carbon depositions through adjustments of feed gas composition and operational parameters in the methanation reactor are essential. Temperature and pressure variations allow a thermodynamically optimized operation, which can reduce energy costs for compression or lower the amount of gas upgrading for grid feed-in. Vice versa, it is shown that the feed gas can be optimally adjusted to the operational parameters of the methanation via the sorption enhanced reforming process.

1. Introduction

Increasing greenhouse gas emissions and the limited availability of primary energy carriers directed the energy policy of the European Union towards sustainable and innovative energy technologies [1]. Natural gas is one of the most important primary energy carriers in Europe, but its availability is heavily dependent on the non-European market. The production of synthetic natural gas (SNG) from biogenic residues offers a promising alternative to the utilization of fossil fuels and represents a novel concept to support the current energy strategy of the European Union [1, 2].

One possible process route is the dual fluidized bed (DFB) gasification, which allows the utilization of locally available residual biogenic or waste resources and offers possibilities for the production of highly valuable secondary energy carriers on a carbon neutral basis [3, 4]. In combination with sorption enhanced reforming (SER) this technology enables the production of a nitrogen-free product gas with adjustable hydrogen to carbon monoxide and hydrogen to carbon dioxide contents [5].

Before the product gas from the DFB process can be fed to the methanation unit, rigorous gas cleaning is required in order to protect the downstream equipment and the methanation catalyst [6–8]. For the methanation reactor itself, several concepts have been utilized. Adiabatic or cooled fixed bed reactors, fluidized bed reactors, three-phase reactors or micro-reactors. However, only adiabatic fixed bed methanation is commercially available as of today. This variety of reactor types
also explains the wide range of operation conditions. Temperatures from 250 °C to 700° C and pressures from 1 bar to 87 bara have been applied. From a thermodynamic point of view the methanation is favored at low temperatures and high pressures [9, 10]. In order to feed the generated gas into the Austrian gas grid, the feed-in regulations must be satisfied [11]. Alternatively, a mixture of CH₄ and H₂, also referred to as hythane, can be generated as a substitute for natural gas in industrial applications [12].

The main chemical species, which are involved in the methanation reaction system, are CH₄, H₂, CO, CO₂ and H₂O. The corresponding reaction equations are the CO-methanation,

\[
2CO + 3H_2 \rightleftharpoons CH_4 + H_2O \quad \Delta H^\circ_{R,298} = -216 \frac{kJ}{mol} \quad (1)
\]

the reverse water gas shift reaction, and

\[
CO_2 + H_2 \rightleftharpoons CO + H_2O \quad \Delta H^\circ_{R,298} = 39 \frac{kJ}{mol} \quad (2)
\]

the CO₂-methanation (combination of (2) and (1)).

\[
CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O \quad \Delta H^\circ_{R,298} = -177 \frac{kJ}{mol} \quad (3)
\]

Additionally, the reaction enthalpies at 300 °C (\(\Delta H^\circ_{R,300}\)) are given. Besides these species, the product gas of the DFB gasifier also contains higher hydrocarbons. As the main component ethylene (C₂H₄) is identified and is thus included here [13]. The hydrogenation to methane can be written as:

\[
C_2H_4 + 2H_2 \rightarrow 2CH_4 \quad \Delta H^\circ_{R,298} = -209 \frac{kJ}{mol} \quad (4)
\]

A deactivation mechanism of the catalyst, which cannot be prevented by gas cleaning steps, is the formation of solid carbon on the catalyst. While adsorbed carbon on the catalyst surface is a necessary reaction intermediate during methanation, the formation of stable deposits leads to catalyst fouling [14]. Thermodynamically, this deposition can be accounted for by the Boudouard reaction.

\[
2CO \rightleftharpoons CO_2 + C(s) \quad \Delta H^\circ_{R,298} = -174 \frac{kJ}{mol} \quad (5)
\]

The deposited surface carbon can also be hydrogenated to methane,

\[
C(s) + 2H_2 \rightleftharpoons CH_4 \quad \Delta H^\circ_{R,298} = -82.1 \frac{kJ}{mol} \quad (6)
\]

or undergo gasification with steam [15].

\[
C(s) + H_2O \rightleftharpoons CO + H_2 \quad \Delta H^\circ_{R,298} = 134 \frac{kJ}{mol} \quad (7)
\]

These reactions show that increased amounts of H₂, H₂O or CO₂ in the product gas might prevent the carbon deposition. A different form of deposition can occur through the adsorption of higher hydrocarbons like C₂H₄ on the catalyst surface. Between 500 and 600 °C, this can lead to coke deposits [16].

If kinetic models are considered, all of the above mentioned reaction pathways have to be taken into consideration. The catalytic methanation of syngas is, however, mostly limited by heat and mass transfer and not by kinetics [10]. For temperatures down to 320 °C the gas composition is close to the thermodynamic equilibrium [17, 18]. A thermodynamic calculation thus provides a good estimation of the expected gas composition. Because of the broad variety of possible carbon species, deviations from thermodynamic equilibrium for carbon deposition have to be expected [10]. Nevertheless, graphitic carbon has previously been used to elucidate this issue, since kinetic models are often only valid for specific reaction conditions and catalysts [19].

Extensive studies have been performed on the thermodynamics of methanation [14, 19, 20]. However, for systems with multiple simultaneous reactions, the effect of different product gas mixtures as well as temperature and pressure is not straightforward. Thus, the thermodynamic calculations in this paper are applied to
different product gas mixtures, which have been obtained by the gasification of different biogenic residues with the 100 kWth DFB gasifier at TU Wien. The chosen feed gas compositions for the methanation aim at covering the broad range of product gas compositions, which can be produced by the DFB gasifier.

2. Concept and methodology

In order to calculate the thermodynamic equilibrium, only four of the seven reaction equations (Eq. (1) to Eq. (7)) need to be considered. Otherwise, the system would be overdetermined, because only four equations are linearly independent of each other. For example, the CO₂-methanation reaction can be seen as the reversed water gas shift reaction followed by the CO-methanation. Thermodynamic calculations are performed with HSC Chemistry and MATLAB. The main focus of this investigation is a low temperature methanation (300 °C) at ambient pressure. These parameter settings result from the current efforts in the design and construction of a lab-scale fluidized bed methanation test rig at TU Wien for the given parameters. Nevertheless, also a temperature variation from 200 °C to 500 °C and a pressure range from 1 bar to 10 bar are carried out. Graphite is chosen as the prevailing carbon species, since Frick et al. [19] found that the Gibbs free energy is lower than for amorphous carbon and is thus preferentially formed.

In order to classify the feed gas composition the stoichiometric number (SN) is defined.

$$ SN = \frac{y_{H_2}}{3y_{CO} + 4y_{CO_2} + 2y_{C_2H_4}} $$

(8)

SN gives the ratio between the molar fraction of H₂ ($y_{H_2}$) to the molar fractions of the carbonaceous species in the feed gas which react to CH₄. If SN is equal to one, there is a stoichiometric amount of H₂ available according to Eqs. 1, 3 and 4. Because the regarded pressures in this study are relatively low, it is safe to assume ideal gas behavior. Molar fraction are thus equal to volume fractions. This definition of SN is not unambiguous, because the chemical equilibrium is influenced by all available species and therefore also by CH₄ and H₂O. Nevertheless, it allows an approximate classification of the feed gas mixture. Typical product gases from the DFB gasification show similar CH₄ concentrations. Water concentrations in the feed gases are assumed zero. This is attributed to the required gas cleaning which is conventionally carried out at low temperatures [21]. If similar CH₄ concentrations and a water free feed gas are assumed, the implementation of SN is justified.

Additionally, the CH₄ yield ($Y_{CH_4}$),

$$ Y_{CH_4} = \frac{n_{CH_4,eq}}{\Sigma_i n_{i,feed}} \times 100 $$

(9)

the carbon yield ($Y_C$),

$$ Y_C = \frac{n_{C,eq}}{\Sigma_i n_{i,feed}} \times 100 $$

(10)

the CO conversion ($X_{CO}$), and

$$ X_{CO} = \frac{n_{CO,feed} - n_{CO,eq}}{n_{CO,feed}} \times 100 $$

(11)

the CO₂ conversion ($X_{CO_2}$) are defined.

$$ X_{CO_2} = \frac{n_{CO_2,feed} - n_{CO_2,eq}}{n_{CO_2,feed}} \times 100 $$

(11)

Index $i$ refers to the carbonaceous species in the feed ($i = CH_4, CO, CO_2, C_2H_4$). Gas cleaning is not within the scope of this study. The feed gas mixture for the methanation is assumed free of impurities and other minor components. Besides, kinetics or heat and mass transfer phenomena are not considered.
Tab. 1: Investigated feed gases

| Parameter                      | Unit | Feed gas number |
|--------------------------------|------|-----------------|
| Source                         | -    | [5]             |
| Gasification agent             | -    | [3]             |
| Feedstock                      | -    | [22]            |
| Bed material                   | -    | [23]            |
| Gasification temperature       | °C   | [24]            |
| Combustion temperature         | °C   | [25]            |
| Source                         | -    | [5]             |
| Gasification agent             | -    | [3]             |
| Feedstock                      | -    | [22]            |
| Bed material                   | -    | [23]            |
| Gasification temperature       | °C   | [24]            |
| Combustion temperature         | °C   | [25]            |

Feed gas composition (water free feed)

| H₂     | vol.-% | 68.3  | 52.4  | 42.6  | 35.6  | 25.8  | 71.1-47.6 |
|--------|--------|-------|-------|-------|-------|-------|-----------|
| CO     | vol.-% | 6.5   | 18.4  | 12.0  | 11.7  | 7.3   | 7.3-21.6  |
| CO₂    | vol.-% | 8.9   | 21.3  | 21.8  | 36.5  | 33.7  | 4.1-23    |
| CH₄    | vol.-% | 14.5  | 7.3   | 12.0  | 11.7  | 7.3   | 17.4-8.8  |
| C₂H₄   | vol.-% | 1.9   | 0.6   | 2.4   | 2.5   | 1.1   | 1.9-0.5   |

*CO₂/H₂O = 68/32 vol.-%  
b O/L = 80/20 wt.-%

3. Results and discussion

In Tab. 1 the investigated feed gas compositions for the methanation are shown. Feed gas no. 1 shows a typical SER gas with high hydrogen content. Limestone (L) is used as bed material at lower temperatures and bark (BA) is chosen as the feedstock. Feed gases no. 2-4 present product gases from conventional gasification. With feed gas no. 2 the same fuel and bed material is used but the gasification temperature is higher which results in lower H₂ and higher CO and CO₂ contents. For feed gas no. 3 lignin (LI) is used as fuel and olivine (O) as bed material. Sewage sludge (SS) and an olivine/limestone mixture (O/L) are the basis for feed gas no. 4, which results in low H₂ and high CO₂ contents. For feed gas no. 5, a CO₂/H₂O mixture is used as gasification agent and rapeseed cake (RSC) and O as fuel and bed materials, respectively. This results in even lower H₂ and high CO and CO₂ concentrations. Feed gas no. 6 shows a temperature variation for SER gasification. This is included to demonstrate the adaptability of the DFB gasifier to the requirements of the methanation process (also see Fig. 4). Data for this variation is only available for softwood (SW) as feedstock.

In Fig. 1 results of chemical equilibrium calculations at 300 °C and 1 bar are shown for feed gas nos. 1-5. The volume fractions of the dry gas components after methanation (referred to as raw-SNG) and the water content of the raw-SNG are depicted. C₂H₄ is not displayed in any of the figures, because it is completely converted under all investigated conditions. CO is not shown in Fig. 1, because only trace amounts remain in the raw-SNG. Additionally, Tab. 2 lists some key figures as defined in Eqs. 8-11.

Almost complete conversion of CO and CO₂ to CH₄ can be reached with the SER feed gas (feed gas no. 1). The CH₄ yield is

![Fig. 1: Raw-SNG gas composition for feed gas nos. 1-5 at 1 bar, and 300 °C](image-url)
Tab. 2: Key figure results of the equilibrium calculations

| Par.       | Unit | Feed gas number |
|------------|------|-----------------|
| $SN$       |      | 1   | 2   | 3   | 4   | 5   |
| $Y_{CH_4}$ | %    | 1.16| 0.37| 0.27| 0.19| 0.11|
| $Y_C$      | %    | 99.9| 29.4| 24.6| 14.4| 5.2 |
| $X_{CO}$   | %    | 0.27| 46.5| 54.5| 52.8| 50.9|
| $X_{CO_2}$ | %    | 100 | 99.9| 99.9| 99.8| 99.9|

99.9% and no carbon formation is thermodynamically expected. However, 22 vol.-%db of H$_2$ are still in the raw-SNG and would have to be separated before grid feed-in below 4 vol.-%. Feed gas nos. 2 and 3 show similar results to each other. About 50 vol.-%db CH$_4$ can be expected. CO is almost completely transformed, whereas less than 50% of the CO$_2$ is converted. Thermodynamically, severe carbon deposition can occur with about half of the feed carbon being separated from the gas stream. The raw-SNG from the SS product gas (feed gas no. 4) shows a CH$_4$ yield of only 14.4%. The rest of the gas is mainly CO$_2$. If a product gas from CO$_2$ gasification is used (feed gas no. 5), 86.5 vol.-%db of the raw-SNG consists of CO$_2$ and only 10.4 vol.-%db are CH$_4$. This is equal to a methane yield of 5.2% and a CO$_2$ conversion of 2.2%, respectively.

For feed gas nos. 2-5 the $SN$ is below one. Since all feed gases are dry, this correlates with the carbon deposition. CO is almost completely converted for all feed gases. However, even small amounts of CO might exceed the allowed threshold level for grid feed-in on the one hand. On the other hand, CO$_2$ methanation is found to be kinetically hindered even for very low CO concentrations [26]. For feed gas no. 1 7 ppm$_{v,db}$ of CO remain in the raw-SNG in the thermodynamic equilibrium. At least 580-750 ppm$_{v,db}$ need to be expected for feed gas nos. 2-5.

Investigation of the sewage sludge product gas

In the following section a more in-depth discussion of the SS product gas follows (feed gas no. 4). Because of the expected carbon deposition for this feed gas composition, H$_2$O should be added if a long catalyst lifetime and a high conversion efficiency are aimed at. Fig. 2 depicts the raw-SNG gas composition after the addition of H$_2$O for temperatures from 200-500 °C and pressures of 1, 5 and 10 bara (Fig. 2b). The amount of water added corresponds to the minimum amount needed to prevent carbon deposition. This minimum volume fraction of H$_2$O in the feed gas ($H_2O_{feed}$) as well as $Y_{CH_4}$ are also displayed (Fig. 2a). With increasing temperature, less CH$_4$ and CO$_2$ and more CO and H$_2$ are present. Accordingly, the CH$_4$ yield decreases from 41% to 26% with increasing temperature.
at 1 bar. $H_2O_{feed}$ decreases from 55 vol.-% to 40 vol.-% within the displayed temperature range. Nevertheless, the methanation would be preferred at low temperatures from a thermodynamic point of view, if the additionally required steam is not seen as the decisive factor. Especially the strongly rising CO content at higher temperatures makes low temperature methanation attractive. Pressure only has a significant influence on the gas composition at higher temperatures. At 500 °C $Y_{CH_4}$ can be substantially elevated if the pressure is increased to 5 bar. A further pressurization only allows a minor improvement of $Y_{CH_4}$. Whereas, at 200 °C $Y_{CH_4}$ is almost constant for all pressures. For $H_2O_{feed}$, hardly any influence of pressure can be observed.

A comparison of Fig. 1 and Fig. 2 clarifies the influence of the added feed water at 300 °C and 1 bar for the SS product gas. By the addition, the CH$_4$ content is elevated from 29 to 37 vol.-%db. At the same time, the CO$_2$ content is lowered from 66 to 56 vol.-%db. H$_2$ slightly increases from 5.6 to 6.6 vol.-%. The CO concentration is marginally lowered from 714 to 667 ppmv,db. Despite the addition of 48 vol.-% of H$_2$O to the dry feed gas, the H$_2$O concentration in the raw-SNG only increases slightly. This can be visualized by the steam gasification reaction (Eq. 7). Most of the steam is needed to gasify the carbon. Because no carbon is present after feed water addition, $X_{CO_2}$ is negative under all displayed conditions. In other words, more CO$_2$ is formed during methanation than converted.

For grid feed-in the raw-SNG needs to be freed of CO$_2$. A maximum of only 2 vol.-% is allowed. A H$_2$ content below the allowed threshold level of 4 vol.-% after CO$_2$ separation could be achieved by increasing the pressure at 260 °C to 10 bar. If the desired commodity is hythane only CO$_2$ separation is necessary.

**Investigation of the SER product gas**

Feed gas no. 1 is a typical SER product gas with a high H$_2$ content. SN is greater than one, which allows a practically complete methanation of the carbon oxides (CO+CO$_2$) at temperatures below 300 °C with a CH$_4$ yield of nearly 100 % (Fig. 3). Pressure again only has significant influence on the gas composition at higher temperatures. With pressurization, the decreasing trend of CH$_4$ and the increasing trends of H$_2$, CO and CO$_2$ at higher temperatures can be counteracted. In addition, above 440 °C at 1 bar carbon formation is thermodynamically possible. As is shown in Fig. 3a, H$_2$O needs to be added. At higher pressures, this can be prevented. Below 300 °C there is practically no influence of pressure or temperature on the gas composition.

![Fig. 3: Temperature and pressure variation for feed gas no. 1 in the thermodynamic equilibrium: 1 bar (full line), 5 bar (dashed line) and 10 bar (dash-dotted line). a) CH$_4$ yield, b) Raw-SNG gas composition](image-url)
Methanation around 300 °C and 1 bar shows a favorable raw-SNG composition without the need of compression. Lower temperatures would not improve the gas composition, but increase the challenge of employing an active catalyst. For grid feed-in, only H₂ would need to be separated from the raw-SNG. For the application as hythane on the other hand, no further upgrading step is necessary except water condensation. Besides these advantages, the methanation of SER product gas also comes with drawbacks. The high H₂ content can only be reached by the increased transport of carbon from the fuel to the flue gas. However, the excess carbon (in the form of CO₂), which is still in the raw-SNG in case of conventional gasification, is already removed within the gasification process for the SER product gas. The overall process chain thus shows a low carbon utilization factor. But this lower carbon utilization factor lies primarily in the gasification process and not in the methanation process. Whereas the addition of H₂ from external sources (e.g. electrolysis) would allow the methanation of the leftover CO₂ and yield a high carbon utilization factor for conventional gasification. Another suggestion would be the installation of a water gas shift reactor prior to the methanation reactor, which was demonstrated successfully for hydrogen production from DFB derived wood gas [27].

Investigation of variable product gas compositions of the SER process

Fuchs et al. [25] already described the adaptability of the SER process with regard to the product gas composition. In Fig. 4 the evolution of the product gas components over the gasification temperature of the 100 kWth DFB gasifier at TU Wien is depicted. By temperature variation, the product gas can be adjusted to the required feed gas for methanation. However, this also adds an additional parameter to the modelling of the methanation reactions.

Fig. 5 displays the composition of the raw-SNG in the thermodynamic equilibrium for all data points of Fig. 4 over SN. Temperature and pressure are again set to 300 °C and 1 bar for the methanation process, respectively. In order to assess carbon formation \( Y_C \) is given. There is a decreasing trend for CO₂, H₂O and the amount of carbon formed for an increasing SN. CH₄ has a maximum at a SN slightly above one. At the same point carbon formation declines to zero and the small incline in H₂ turns into a sharp increase for higher SN. CO is only present in trace amounts (0.14-614 ppmv,db) and is not displayed here. From a thermodynamic point of view, the feed gas with a SN of 1.09 generates a raw-SNG with the most favorable composition for the methanation at 300 °C and 1 bar. A SN of 1.09 corresponds to a gasification temperature of about 680 °C.
The associated compositions for the feed gas and the raw-SNG as well as the key figures are depicted in Tab. 3. Different operation conditions of the methanation might favor other feed gas compositions from Fig. 4 and vice versa. In order to find the most suitable feed gas composition for deviating methanation conditions, reiterations of the thermodynamic equilibrium calculations would have to be carried out.

Tab. 3: Feed gas and raw-SNG composition and key figures for the feed gas with a SN of 1.09 at 300 °C and 1 bar in the thermodynamic equilibrium

| Par. | Unit | Feed gas | Raw-SNG |
|------|------|----------|---------|
| CH₄  | vol.-%<sub>db</sub> | 13.3     | 86.1    |
| H₂   | vol.-%<sub>db</sub> | 67.8     | 12.8    |
| CO   | vol.-%<sub>db</sub> | 7.3      | 0.005   |
| CO₂  | vol.-%<sub>db</sub> | 9.8      | 1.1     |
| C₂H₄ | vol.-%<sub>db</sub> | 1.7      | 0       |
| H₂O  | vol.-%<sub>db</sub> | 0        | 40.2    |

| Y₈CH₄ | % | 98.8 |
| Y₈C   | % | 0    |
| X<sub>CO</sub> | % | 100  |
| X<sub>CO₂</sub> | % | 95.8 |

4. Conclusion and Outlook

In this work, the suitability of various product gases from the 100 kW<sub>th</sub> DFB gasifier for methanation has been evaluated from a thermodynamic point of view. It has been shown, that complete methanation is only possible for SER product gases. For all other gases, only the partial methanation of CO is possible, whereas CO₂ might even constitute the main raw-SNG component. Additionally, gases from conventional steam gasification or gasification with CO₂ admixture to the gasification agent (H₂O+CO₂) are subject to carbon deposition. Therefore, H₂O needs to be added for a stable operation. Furthermore, the influence of different operation conditions of the methanation on the raw-SNG composition have been visualized. By the careful choice of operation conditions, energy savings and/or less effort for further gas upgrading can be accomplished. A further investigation of the SER product gases has revealed that it is also possible to adapt the gasification process to suit certain methanation conditions optimally. However, if H₂ is available from an external source, conventional gasification can be beneficial because of a higher carbon utilization factor.

It should be noted that all investigations in this paper are based on thermodynamic equilibrium calculations. There are many other parameters, which can influence the performance of the methanation. Catalyst poisoning due to insufficient gas cleaning, the choice of the reactor concept or kinetic limitations necessitate experimental investigations.

Acronyms

BA  bark
DFB  dual fluidized bed
L  limestone
LI  lignin
O  olivine
O/L  olivine/limestone mixture
raw-SNG  synthetic natural gas after methanation/before gas upgrading
RSC  rapeseed cake
SER  sorption enhanced reforming
SNG  synthetic natural gas
SS  sewage sludge
| SW | softwood       | $Y_C$    | carbon yield in % |
| vol.-% | volumetric percent | $Y_{CH_4}$ | methane yield in % |
| wt.-% | weight percent   | $y_i$     | molar fraction of species $i$ |

### Symbols

- $\Delta H_R^{300}$: molar reaction enthalpy at 300 °C
- $H_2O_{\text{feed}}$: volume fraction of H$_2$O in the feed in vol.-% $\hat{n}_i$: molar flow of species $i$ in mol/s $N_i$: number of carbon atoms in species $i$ $S_N$: stoichiometric number $X_{CO}$: carbon monoxide conversion in % $X_{CO_2}$: carbon dioxide conversion in %

### Indices

- $a$: absolute
- $C$: carbon
- $CH_4$: methane
- $C_2H_4$: ethylene
- $CO$: carbon monoxide
- $CO_2$: carbon dioxide
- $db$: dry basis
- $eq$: equilibrium
- $feed$: in the feed gas
- $H_2$: hydrogen
- $v$: volumetric

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