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Original scientific paper

BIOMASS TO BIOSNG – METHANATION OF SYNGAS FROM BIOMASS GASIFICATION

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Abstract

The conversion of biomass into synthetic natural gas from biogenic sources (bioSNG) via thermochemical routes is a promising approach for integrating renewable energy into existing gas infrastructure. A key step in this pathway is the catalytic methanation of synthesis gas (syngas), typically consisting of H_2, CO and CO_2. This study focuses on the experimental investigation of syngas methanation using a fixed-bed reactor cooled by thermal oil, with the aim of assessing the influence of various process conditions on reactor performance and methane concentration. The experimental setup comprises a tubular reactor operating with a commercial Ni-based catalyst under steady-state conditions. External temperature regulation is achieved via a thermal oil cooling, enabling effective control of the exothermic reaction. A series of experiments were conducted to examine the effects of gas hourly space velocity (GHSV), reactor pressure, and thermal oil temperature on the methanation process. Higher pressures and lower GHSV values resulted in enhanced methane concentrations and more pronounced exothermic behavior, while the thermal oil temperature played a crucial role in heat removal and reactor stability. In addition, the study explored the effects of higher hydrocarbons, specifically ethene (C_2H_4) and the water content in the feed gas, as well as the impact of increased hydrogen/water content on the methanation reaction. To assess the latter, a feed gas mixture containing 15% H_2O, 55% H_2 and the rest of the original gasification product gas was used. These feed gases were again evaluated under varying pressures, GHSV values, and oil temperatures. The presence of ethene was found to positively influence methane formation, leading to higher methane concentration. Furthermore, the H_2/H_2O mixture was shown to improve methane concentration likely due to enhanced hydrogenation reactions, while an increase in water content alone led to reduction in methane formation. These findings provide valuable insights into the complex heat and mass transfer phenomena occurring during catalytic methanation and highlight the importance of feed composition, as well as thermal management, in reactor design and operation. This work contributes to a better understanding of methanation processes in thermally regulated systems and offers data for the further development and optimization of bioSNG production technologies based on biomass gasification.

***Keywords**: biomass gasification, methanation, bioSNG, thermal oil-cooled reactor, catalyst performance*

1. INTRODUCTION

The production of synthetic natural gas (SNG) from biomass is a promising pathway for renewable storage and decarbonization of the gas grid. [1] This process involves several key steps: biomass gasification, gas cleaning, methanation, and final conditioning of the gas for the injection into the natural gas network. [2]

In the first step, biomass is converted to a raw synthesis gas (syngas) through thermal gasification, typically in a dual fluidized bed (DFB) gasification reactor. [3] This raw gas contains a mixture of hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), steam, nitrogen (N_2), as well as impurities such as tars, sulfur compounds, and particulates. [2] The typical composition of the dry product gas following gasification is presented in Table 1.

Table 1. Typical composition of dry product gas after biomass gasification [4]

Component	Volume fraction [vol. %]
H_2	30-45
CO	20-25
CO_2	20-25
CH_4	6-12
N_2	0-1

After extensive gas cleaning to remove contaminants, the syngas is subjected to methanation, where CO and CO_2 react with H_2 to form CH_4 and water. The main reactions involved are [2]:

$$CO + 3H_2 \rightleftarrows CH_4 + H_2O, \Delta Hr^\circ = -206\ \frac{kJ}{mol} \quad (1)$$

$$CO_2 + 4H_2 \rightleftarrows CH_4 + 2H_2O,\ \Delta Hr^\circ = -165\ \frac{kJ}{mol} \quad (2)$$

These reactions are strongly exothermic, making temperature control within the reactor essential to avoid catalyst deactivation and ensure selectivity towards methane [2].

The resulting product gas – often called bioSNG – must meet strict specifications regarding Wobbe index, calorific value, and methane content before injection into the natural gas grid. [5]

A simplified process flow diagram of the related bioSNG production process investigated within the research project BioHeat is shown in Figure 1.

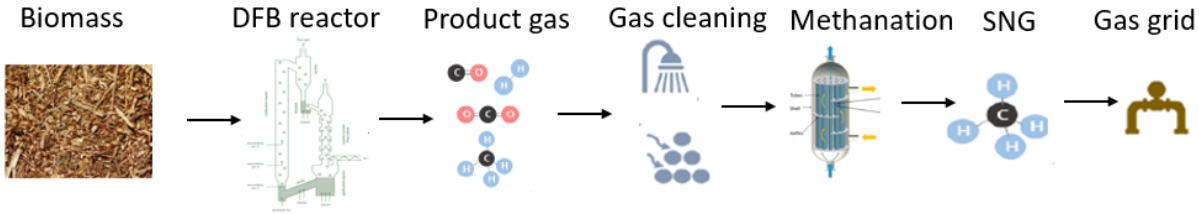

Figure 1. Schematic overview of the bioSNG production process from biomass including dual-fluidized bed gasification and methanation

2. EXPERIMENTAL

This work focuses on the methanation step of the bioSNG process, using a laboratory-scale oil-cooled fixed-bed reactor operated with synthetic gas mixtures that mimic cleaned gasification products. To illustrate the overall setup used for the methanation experiments, a simplified process flow diagram of the test rig is shown in Figure 2. The setup includes the gas supply system, heating and cooling loops, fixed-bed reactors, and analytical equipment for gas composition monitoring. The gas mixture is precisely adjusted using mass flow controllers, allowing for flexible operation under different feed compositions. The thermal oil loop ensures almost isothermal conditions along the reactor wall, which is critical for understanding reaction kinetics and thermal behavior under various operating scenarios. Data acquisition and control are managed through an automated system, enabling reliable and reproducible experimental conditions. This setup provides a robust platform for studying key influences on methane formation and for evaluating potential process optimizations for SNG production.

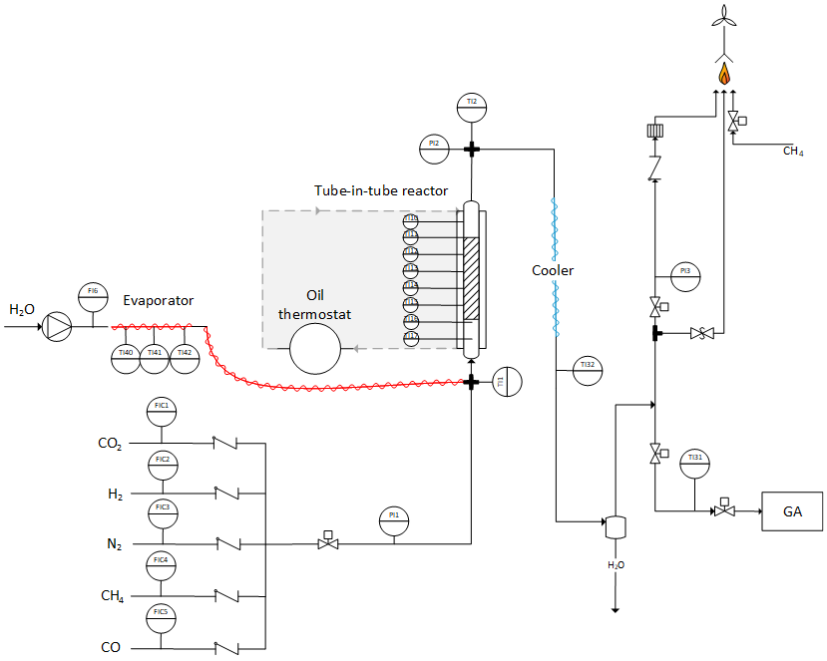

Figure 2. P&I diagram of methanation laboratory-scale pilot plant

The reactor is a single tube stainless steel reactor, filled with a commercial nickel-based catalyst. Eight thermocouple elements are inserted along the reactor axis to monitor the axial temperature profile. The reactor is thermally regulated using a surrounding oil jacket to ensure reactor cooling.

The influence of varying gas hourly space velocity (GHSV), oil temperature, and pressure was investigated at a constant inlet gas composition, as well as under different feed gas types containing water vapor, ethene, or hydrogen/steam mixtures to simulate various operating scenarios.

Gas compositions were selected to match typical outputs of DFB gasifiers after tar and sulfur removal including different amounts of steam or hydrogen addition to counter carbon formation in the reactor. Table 2 summarizes the tested gas compositions in 4 different feed gases.

Table 2. Compositions of tested feed gases (in vol.%)

	50 vol.% H_2O	40 vol.% H_2O	50 vol.% H_2O + ethene	H_2/H_2O mixture
H_2O	50.0	40.0	50.0	15.0
H_2	20.5	24.0	20.5	55.0
CO	12.0	14.5	12.0	12.2
CO_2	11.5	14.0	11.5	11.7
CH_4	6.0	7.5	5.0	6.1
C_2H_4	0.0	0.0	1.0	0.0

3. RESULTS AND DISCUSSION

The experimental investigation focused on how specific components in the feed gas influence methane formation in an oil-cooled fixed-bed methanation reactor. In particular, the addition of higher hydrocarbons – specifically ethene (C_2H_4) – was found to significantly enhance methane production. As illustrated in Figure 3, introducing C_2H_4 into the feed led to a pronounced increase in CH_4 concentration, especially at lower gas hourly space velocities (GHSV). This behavior can be attributed to the rapid and highly exothermic hydrogenation of ethene, which not only boosts local reaction rates but also raises the reactor temperature. The elevated temperatures in turn promote CO and CO_2 methanation, contributing further to the overall methane production.

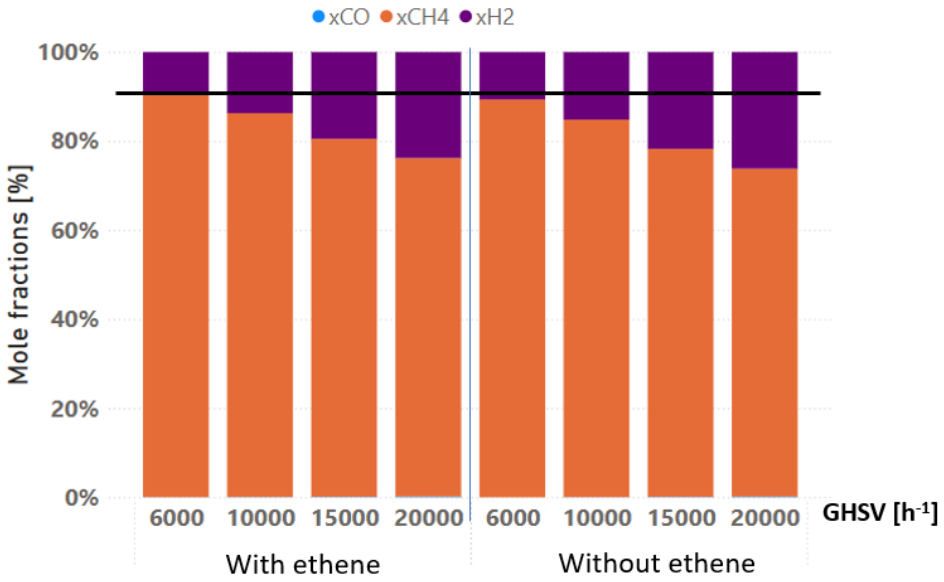

Figure 3. Influence of ethene on methane production under varying process conditions

In addition to this case, the influence of steam concentration (40 vol.% vs. 50 vol.% H_2O) and hydrogen-rich H_2/H_2O mixture on the methanation performance was also investigated. While the detailed diagrams will be presented in a separate publication, it can already be stated that steam content has a strong effect on thermodynamic limits, and that the use of hydrogen-rich mixture enables the production of injection-grade bioSNG under appropriate process conditions.

4. CONCLUSION

The experimental results emphasize the crucial impact of feed gas composition on the efficiency of the methanation process. While high water content alone can limit methane formation due to equilibrium constraints, hydrogen-rich mixtures with added steam (H_2/H_2O) demonstrate a favorable effect when applied under suitable conditions. Additionally, the introduction of higher hydrocarbons, particularly ethene (C_2H_4), significantly enhanced methane production through rapid hydrogenation. These findings highlight the potential of optimizing feed gas composition not only to avoid negative effects but also to actively enhance methane production. As such, careful management of the feed gas composition is essential for maximizing the performance of industrial methanation systems.

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