From wood to biogas – a small-scale demonstration

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1. Introduction

In Denmark, greening the natural gas grid is developing fast. Starting in 2012 with 0%, the share of biomethane (upgraded biogas to natural gas quality) in the grid is now (2019) about 10%. It is still increasing, but to end at the future goal of 100% it is necessary to include other sources, which could be gasification of biomasses followed by methanation.

2. Demonstration plant setup

Allothermal gasification of biomass coupled with ex situ tar removal was shown to be a viable method of producing tar-free syngas well suited for subsequent biomethanation. The gasification was performed in a fluid bed packed with olivine particles as bed material, operated at 1 barg and at isothermal conditions at 900°C furnace temperature corresponding to 800°C reactor temperature.

Steam is used as both the fluidization and gasification agent and is added from the bottom of the gasifier. A flow of wood pellets of 320 g/h, corresponding to a thermal input of approximately 1.55 kW based on the net heating value, and a CO₂ flow of 2 l/min at 1 barg are continuously added to the gasifier from the top. The gasifier uses 5.5 kW electricity, which covers the steam generation, furnace heating and losses.



Fig. 1: System overview of the gasifier (not in operation)

Figure 1 shows a system overview of the gasifier (not in operation) including fuel hopper, fluidized bed gasifier, exit filter, steam supply and CO_2 tank for filter cleaning and CO_2 flushing [1].

3. Steam reforming

the gasification furnace, Post the temperature is kept above 350°C in order to avoid tar condensation in the pipes. Ex situ tar removal was performed through catalytic steam reforming at a temperature of approximately 700 to 800°C reached by internal heat exchange and external electrical heating. The principle is based on an internal heat exchanger packed with three distinct catalysts catalyzing the endothermic steam reforming reactions, the exothermic methanation reactions and water gas shift, respectively. The internal heat exchanger is designed in a way that more energy is released through the exothermic methanation reactions than is consumed through the endothermic steam reforming reactions. Through this concept, it is possible to raise the temperature of the tar containing syngas from 350°C up to 700-800°C where the steam reforming reactions take place. A minor amount of electricity is needed to keep the max temperature at 800°C and to account for the heat losses of the tar reformer. The observed conversion rates were 100% for tar, 89% for ethylene and 62% for propane.

Figure 2 shows the combined steam reforming and pre-methanation unit. It includes a pre-cleaning part consisting of dolomite pellets only, which also starts a WGS process and heating up syngas from about 400°C to 700°C. Then follows a steam reforming part working at 800°C and reducing tar and methane to a minimum and exchanging heat with the incoming gas. The catalyst is the "TARGET" catalyst from NEXCERIS, USA. The last part of the heat exchanger consists of catalyst, which is a mixture of

dolomite pellets and methanation pellets. The methanation pellets consist of "Methanation Catalyst META-J103" from China, including >18% Ni. In this part, the concentration of H₂O is still high and there are two parallel reactions taking place here: WGS and methanation, while the temperature drops from 600°C to around 400°C by heat exchange. The gas from the tar cracker and pre-methanation then passes through an active carbon filter to remove any remaining pollutants and from there even through a water scrubber. We never found any tar in the water scrubber.



Fig. 2: Steam reforming and pre-methanation unit; the tar cracker

4. Biomethanation

The thermal fuel input of 1.55 kW coupled with the electricity consumption and allothermal operations results in a thermal output from the gasifier of 1.78 kW. As expected, most of the electricity is wasted as losses in this small lab-scale gasifier. When adding a biomethanation unit, which has an energy conversion efficiency of 80%, calculations indicate that it is possible to produce methane with an energy efficiency (based on thermal input to the gasifier) of approximately 100%. The biomethanation unit developed at the Technical University of Denmark (DTU) was installed, and the exit gas composition from the tar reformer is exactly suited for the subsequent methanation. It consists of a trickle bed reactor with a total electronic control system for addition of nutrients, pH-control, etc. A part stream of the syngas from the gasifier and tar cracker is added to the bioreactor, which converts all gasses to a biogas only containing CO_2 and CH_4 . The input gas to the biomethanation

unit contains around 48% CO₂, 11% CO, 31% H₂ and 10% CH₄, while the output gas contains around 64% CO₂ and 36% CH₄ (dry basis). Figure 3 shows the composition of the main components throughout the total system from the gasifier to the biomethanation output; all values were measured except the tar. Figure 4 shows the steam consumption, the tar reduction and the energy content in the gas flow throughout the system [2].



Fig. 3: The composition of the main components throughout the total reactor system



Fig. 4: The steam consumption, tar reduction and energy content in the gas flow

5. Tar modelling and mass balance

Tar from the gasifier is represented by Naphthalene ($C_{10}H_8$). According to literature, Naphthalene contributes to a large part of all the compounds in tar from gasification, and furthermore taking the average C/H ratio in tars it usually ends at 10/8 [3,4]. Hence, in this work we assume Naphthalene to represent all tars. We did not measure the tar, and we used the following method of modelling the tar content:

The mass balance was performed based on the fuel supplied to the gasifier and on measurements. The mass flow of fuel and CO_2 were continuously measured. N₂ and O_2 were always zero during the measurement, which is a test of correct

measurements as no air is supplied. The mass balance was performed based on the input and output sources of the gasification process. The gasification process input stream includes fuel material (wood pellets) and steam for fluidization (including moisture present in fuel) and CO₂ flushing to maintain the pressure in the fuel tank and prevent syngas backflow into the fuel tank. The output stream includes syngas, tar and steam. The elemental composition of input and output of gasification was balanced as shown below.

The empirical stoichiometric mass balance was done based on the stoichiometric molar flow of fuel at inlet compared to outlet, as shown in equation (1) and (2).

Fuel + flushing CO_2 + converted H_2O = measured gas components out + tar (1)

Equation (1) is developed as shown below:

 $x(C_{13}H_{19}O_8 + bH_2O + aCO_2) + yH_2O = \%CO + \%CO_2 + \%CH_4 + \%C_3H_8 + \%H_2 + \%C_2H_4 + zC_{10}H_8$ (2)

Equation (2) is explained as follows:

 $C_{13}H_{19}O_8$ represents wood that gives the correct relation of masses of C, H and O. The known amount of moisture in the fuel is represented by bH₂O and "b" is a calculated variable. The relation between flushing CO₂ and fuel is known, which determines variable "a". The inflow of CO₂ flushing was measured using a flowmeter. The fluid bed of the gasifier is fluidized by steam and yH₂O represents the amount of steam converted to other gases during the gasification process. Only a part of the H₂O is converted. "x" is the stoichiometric amount of fuel in inlet.

On the right side of the equation, all the measured molar concentrations of the output gases are assumed to be the

arbitrarily chosen stoichiometric amounts. As mentioned, in this work we assume Naphthalene to represent all tars. "z" is then the stoichiometric amount of $C_{10}H_8$ in the outlet. With these assumptions it is possible to calculate the three unknowns, x, y and z from the three elemental balance equations for C, O and H using Problem Solver in Microsoft Excel. The elemental material mass balance calculations were performed from volume basis to molar basis using the measured gas composition. Then the molar composition of dry gas and fuel was calculated with respect to its individual elements to form empirical equation stoichiometric for the gasification of wood. The calculations were made based on measurements of gas

compositions throughout the whole system from gasifier inlet to biomethanation outlet.

6. Conclusion

In a small-scale demonstration plant, we succeeded in converting wood pellets into biogas $(CO_2 + CH_4)$.

The gasifier was an electrically heated labscale fluid-bed and the conversion efficiency of the biomass was about 100% caused by the added electricity.

The tar was converted 100% to syngas in a steam reformer. The syngas from the gasifier and tar cracker was converted into biogas in a biomethanation unit.

We have shown that instead of measuring, it is possible to calculate the steam conversion and the tar content from the gasifier based on measurements of input mass flows and output composition of the gasses.

7. Future developments

The obvious future developments in connection with the gasifier are as follows:

- Testing new fuels, e.g. separated manure fibers, separated biogas fermented fibers, washed and dried straw, etc.
- Testing the present system, but using thermal methanation instead of biomethanation
- Testing more efficient biomethanation systems (more production/liter reactor)
- Investigating how to scale up all the good results from this laboratory-

scale system to a full-scale system keeping the high conversion efficiency and minimizing the electricity consumption.

The last point is going on now in a preproject, but the developments will depend on funding for the continuation.

8. Acknowledgements

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9. References

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