

Co-production of green, high quality hydrogen and bio SNG using electrochemical hydrogen compression



CO-PRODUCTION OF GREEN, HIGH QUALITY HYDROGEN AND BIO SNG USING ELECTROCHEMICAL HYDROGEN COMPRESSION

A.J. Grootjes¹, P.J. Bouwman², L.F.J.M. Raymakers³, G. Aranda Almansa⁴, C.M. van der Meijden⁵

¹ Energy research Centre of the Netherlands (ECN), P.O. Box 1, 1755 ZG Petten, The Netherlands, Phone: +31 88 515 4504, Fax: +31 224-568487, E-mail: grootjes@ecn.nl

² HyET, Leemansweg 15, 6827 BX Arnhem, The Netherlands, Phone: +31 26 36 23 944, Fax: +31 26 36 23 945, E-mail: peter.bouwman@hyet.nl

³ HyET, Phone: +31 26 36 23 944, E-mail: <u>leonard.raymakers@hyet.nl</u>

⁴ ECN, Phone: +31 88 515 4663, E-mail: <u>aranda@ecn.nl</u>

⁵ ECN, Phone: +31 224-564582, E-mail: <u>vandermeijden@ecn.nl</u>

ABSTRACT: Bio-SNG (Synthetic Natural Gas) is a gas containing mostly CH₄, with properties similar to natural gas, which can be produced from thermochemical gasification of biomass coupled to subsequent methanation. Due to its interchangeability with natural gas, SNG can be injected into the existing grid and easily distributed for transport, heat, and electricity applications. SNG can also be efficiently converted in a number of proven end-use technologies.

Recently, ECN has developed and patented a novel technology for the methanation of gas from biomass gasification. The ECN System for MEthanation (ESME) allows the efficient conversion of producer gas from biomass gasification to SNG because the hydrocarbons contained in the producer gas (e.g. benzene, toluene) are not removed but converted, and are thus potentially available to be converted to methane. In October 2014, the main parts of the system were successfully tested downstream atmospheric gasification in a 500-hour experiment. The results were an important step for the scale-up of bio-SNG production. Engineering of a 4 MWth bio-SNG pilot plant to be located in Alkmaar (the Netherlands) is currently underway.

One of the main challenges for bio-SNG is to reduce the production cost in order to come closer to the price level of natural gas. A smart approach to achieve this goal is the co-production of fuels and chemicals. In this work, co-production of bio-SNG and hydrogen has been evaluated due to the promising market prospects for renewable hydrogen in fuel-cell (automotive/stationary) applications as well as hydrogen for (glass, steel, petrochemical) industry. The recovery of hydrogen from biomass gasification producer gas is an attractive option for the production of renewable hydrogen from biomass. For this, novel technologies for the separation of hydrogen from producer gas are required. The Electrochemical Hydrogen Compression (EHC) technology developed by HyET can selectively extract high-purity (> 99.9%), high-pressure (> 20 MPa), fuel-cell grade hydrogen from producer gas. The integration of ECN and HyET technology leads to a reduction of the bio-SNG yield, but with similar or higher overall energy efficiency and higher total value of the products, thus lowering the overall cost of bio-SNG production.

Within the 'PurifHy' project, the concept of co-production of hydrogen from SNG combining the ESME bio-SNG process and the EHC technology has been proven at lab scale. This paper shows the main results obtained within the project and includes an economic outlook for a full scale application.

Keywords: Biomass gasification, HyET, Electrochemical Hydrogen Compression, Synthetic Natural Gas, H₂, hydrogen, MILENA, ESME, co-production.

1 INTRODUCTION

SNG (Substitute Natural Gas, or Synthetic Natural Gas) is defined as a gas containing mostly CH₄, with similar properties to natural gas, which can be produced from thermochemical gasification of fuels (e.g. coal, biomass) coupled to subsequent methanation. The overall efficiency of conversion from biomass to SNG can be up to 70% in energy basis [1].

Due to its interchangeability with natural gas, SNG can be efficiently converted in a number of proven enduse technologies. It can be cheaply produced at large scale, and is a storable energy carrier, thus enabling whole year operation independently of fluctuations in demand. Moreover, SNG can be injected into the existing grid and easily distributed for transport, heat, and electricity applications [2].

SNG is not only an attractive, versatile energy carrier for bioenergy, but can also be used for storage of surplus power from renewable sources (e.g. solar, wind). This is the so-called "power-to-gas" concept where excess

renewable power produces via electrolysis H_2 that is added to an existing plant to convert additional CO_2 into CH_4 [3].

Recently, ECN has developed and patented a novel technology for the methanation of gas from biomass gasification. The ECN System for MEthanation (ESME) [4] is designed especially for gas from fluidized bed gasifiers and allothermal gasifiers. The ESME concept is unique because of the smart sequence and the operating conditions of the different units. ESME allows the efficient conversion of producer gas from biomass gasification to SNG because the hydrocarbons contained in the producer gas (e.g. benzene, toluene) are not removed but converted, and are thus potentially available to be converted to methane.

The proposed process by ECN for efficient production of bio-SNG is shown in Figure 1. The process starts with the conversion of solid biomass into producer gas in the MILENA indirect gasifier, which can achieve total conversion of the fuel and the production of a nearly N_2 -free producer gas without the need for an air

separation unit in an integrated design [1][5]. The producer gas from the gasifier is then cleaned from tars in the OLGA tar removal system, which is based on the staged removal of tars above the water dew point in a series of oil scrubbing columns [6]. After tar removal, most of the water contained in producer gas is removed in a gas cooler and compressed to approximately 6 bar. This first compression step will not be needed if the gasifier operates at a similar pressure. A commercial hydrodesulphurisation (HDS) catalyst is applied to convert organic sulphur compounds into H₂S and COS. The HDS catalyst also hydrogenates alkenes and alkynes into alkanes. After complete sulphur removal in a ZnO bed, aromatic hydrocarbons (e.g. C₆H₆) are reformed and some of the CO and H2 is converted into CH4 in the prereformer. In this reactor, reforming and methanation take place at the same time in such a way that the heat from the methanation reactions can be used to reform the aromatic hydrocarbons. Then, CO, CO2 and H2 are converted into CH₄ and H₂O in the low-pressure methanation unit. After CO2 removal (e.g. by amine scrubbing), the last traces of CO and H₂ are converted in the high-pressure methanation unit. After polishing (drying, odorization), the produced bio-SNG can be injected in the grid.

In October 2014, the main parts of the system (HDS, prereformer and methanation reactors) were successfully tested downstream atmospheric gasification in a 500-hour experiment [1]. The results were an important step for the scale-up of bio-SNG production. The basic engineering of a 4 MWth bio-SNG pilot plant to be located in Alkmaar (the Netherlands) is currently underway [7]. The foreseen installation, based on MILENA/OLGA/ESME technology, is intended as a demonstration of the technology before going to commercial scale (50-100 MWth).

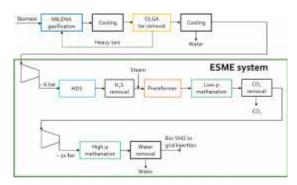


Figure 1: Schematic layout of ECN process for bio-SNG production from biomass gasification.

One of the main challenges for bio-SNG is to reduce the production cost in order to come closer to the price level of the equivalent fossil-based natural gas. A smart approach to achieve this goal is the implementation of coproduction schemes of fuels and chemicals. Low-medium temperature gasification (typical of biomass gasification processes) leads to a producer gas containing significant amounts of CH₄, C₂-C₄ hydrocarbons and BTX. ECN has been recently working in the recovery of BTX (benzene, toluene, xylenes) and ethylene from producer gas [8]. In this work, co-production of bio-SNG and hydrogen has been evaluated due to the promising market prospects for renewable high purity and high pressure hydrogen in fuel-cell (automotive/stationary) applications as well as

hydrogen for (glass, steel, petrochemical) industry. The recovery of hydrogen from biomass gasification producer gas is an attractive option for the production of "green" hydrogen from biomass. For this, novel technologies for the separation of hydrogen from producer gas are required. The Electrochemical Hydrogen Compression (EHC) technology developed by HyET can selectively extract high-purity (> 99.9%), high-pressure (> 20 MPa), fuel-cell grade hydrogen from producer gasFout! Verwijzingsbron niet gevonden.. The basis of the EHC technology is electrochemical H₂ compression. Hydrogen is simultaneously separated from the gas mixture and compressed through an impermeable catalyzed membrane by applying electricity, as depicted schematically in Figure 2. Electrochemical compression is an isothermal process, compared to mechanical compression which is an adiabatic process. The theoretical energy consumption of EHC is 3.2 times lower than mechanical compression when compressing from 1 bar to 1000 bar in a single stage.

The transport of protons through the membrane starts already at a potential of only 5 mV due to the platinum catalyst material on both sides of the membrane but in the electrochemical cell there are several phenomena causing losses. The two main factors are the Nernst potential and the proton conductivity of the membrane. The Nernst potential is caused by the partial pressure difference of hydrogen over the membrane. The proton conductivity mainly is influenced by the thickness of the membrane, the water concentration inside the membrane and the temperature.

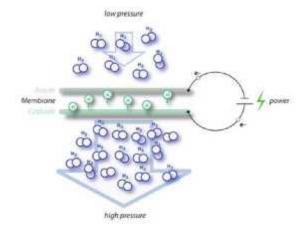




Figure 2: Schematic of HyET's EHC technology and EHC unit [9].

The integration of ECN and HyET technology leads to a reduction of the bio-SNG yield due to the fact that the $\rm H_2$ recovered via EHC technology is not available anymore to be converted in methane, but on the other hand the overall energy efficiency is not affected. Moreover, thanks to the higher value of high purity, high pressure hydrogen with respect to SNG (2 – 10 \oplus kg, i.e. 15-84 \oplus GJ LHV, compared to \sim 10 \oplus GJ LHV for bio-SNG [11]), the total value of the process products is increased by the implementation of co-production, thus lowering the overall cost of bio-SNG production.

Two options for the integration of H_2 + bio-SNG production (which will be studied in Section 3) can be considered. In all cases, the reference scenario, case a), is the ESME bio-SNG process shown in Figure 1. The first option, case b), plotted in Figure 3, considers the replacement of the high-pressure methanation section by an EHC unit for the separation of hydrogen from the gas. After CO_2 removal by conventional means (e.g. amine scrubbing), the remaining H_2 would be separated from the SNG. SNG after polishing (water removal, odorization) would be ready for grid injection.

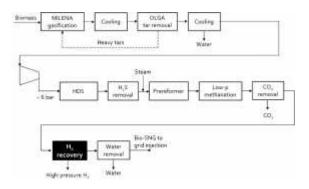


Figure 3: Case b), integration of bio-SNG + H₂ coproduction: replacement of high-pressure methanation section by EHC unit.

An alternative option, case c), plotted in Figure 4, consists of the optimization of the process to favor H_2 yield with respect to bio-SNG. This option would be implemented through the addition of a WGS unit to convert CO in additional H_2 followed by the harvesting of the hydrogen. This option not only produces more hydrogen, but also decreases the CO content, which is a poison for a platinum-based catalyst.

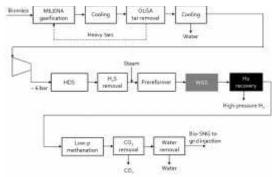


Figure 4: Case c), integration of bio-SNG + H_2 coproduction, option 2: increasing H_2 yield via WGS.

With this background, the co-production of hydrogen and bio-SNG has been assessed and tested at lab scale within the 'PurifHy' project [12]. This paper will present and discuss the most relevant results from experiments and a techno-economic evaluation.

2 MATERIALS AND METHODS

2.1 MILENA/OLGA/ESME lab-scale setup

The lab-scale MILENA/OLGA/ESME experimental facility for production of bio-SNG from biomass gasification was used for the production of gas. The experimental bio-SNG setup is composed of a 25 kWth MILENA indirect gasifier, from which a slipstream of ~ 1 Nm3/h dry gas is directed to the system downstream. The gas is cleaned from tars in the OLGA unit, cooled down for the removal of water, compressed to ~ 6 bar and directed to the ESME system at a target gas flow rate of 11-12 NL/min. The ESME test rig is composed of 6 reactors in series. The first reactor is hydrodesulphurisation (HDS), which consists of a fixedbed reactor filled with a commercial CoMoO catalyst. The HDS catalyst converts the organic sulphur compounds (e.g. thiophene, mercaptans) into mainly H₂S and COS, and hydrogenates alkanes and alkynes into alkanes (e.g. C₂H₄ and C₂H₂ into C₂H₆). The WGS reaction also takes place in this reactor. The produced H₂S and COS is removed from the gas downstream in a conventional adsorption ZnO bed and a guard bed. The next reactor is the prereformer unit, which consists of a fixed bed filled with a commercial Ni-based catalyst. Steam is added to the gas upstream the reactor. The prereformer converts aromatic hydrocarbons (benzene, toluene), and simultaneously WGS and methanation reactions take also place. Two methanation units consisting of fixed bed reactors filled with a commercial Ni-based catalyst further convert CO and H₂ into methane. The dry raw bio-SNG produced in the lab-scale setup contains ~ 52% CO₂, 39% CH₄, 2% H₂, traces of CO (~100 ppmv) and C₂H₆, some N₂, and Ar and Ne which were added for molar balances. Further details about the setup can be found elsewhere [2][4].

During tests performed in October –November 2014 [1], part of the raw bio-SNG produced at the lab-scale setup was stored in bottles which were afterwards used for testing the HyET EHC setup using real gas.

2.1 HyET EHC experimental setup

For the effective removal of the small percentage of H_2 out of the raw bio-SNG, HyET has developed the MoHyTO test system (Mobile Hydrogen Test Object). This system basically consists of all Balance-of-Plant equipment to support EHC stack operation, including a humidifier for the feed gas, piping and valves, thermal control, current supply, data logging and the stack itself. The EHC output provides pure, high pressure hydrogen (permeate) on one side and retentate off-gas on the other side. The system was completely traced to prevent liquid water condensation, which can block the gas supply to the membrane and the uptake of the hydrogen to the catalyst on the membrane.

The heart of the system is the EHC stack. First an EHC stack consisting of 30 electrochemical cells with an active area of 42.6 cm² each was tested. Inside this stack of cells, the gas flow was distributed equally over every

membrane to guarantee equal performance of each membrane. The EHC could deliver pure hydrogen at pressures up to 200 bar, but in this experiment 30 bar was chosen as a suitable output working point.

In the second experiment a different EHC setup was used consisting of only a single membrane with an area of 285 cm². This EHC was capable of a pressure difference up to a maximum of 5 bar, but designed to achieve superior purification results for low level hydrogen extraction.

The membrane and catalyst combination used here operated best at a temperature of 150 °C. This temperature avoided excessive adsorption of the CO-traces on the catalyst material reducing its activity.

The system is capable of autonomous operation and if needed it can be controlled remotely. The hydrogen concentration of both the feed gas stream and retentate has been measured with two gas analyzers (ABB Advance Optima Continuous Emission Monitoring).

3 RESULTS AND DISCUSSION

3.1 Experimental assessment of performance of HyET technology using real raw bio-SNG from the ESME system

Two experimental plans were carried out at different setups: in the first experiments, raw bio-SNG as feed gas was tested in the MoHyTO setup (42.6 cm 2 membrane area/cell). In the second experimental plan, a mixture of $N_2 + H_2$ was tested as feed gas in a 285 cm 2 single membrane setup. In all cases, the hydrogen concentration entering the EHC system was 3.5 vol.%.

During the tests the recovery rate of hydrogen is determined (Eq. 1). This is the ratio of concentration difference between feed gas and retentate to the concentration in feed gas (bio-SNG). A recovery rate of 100% means that all $\rm H_2$ in the bio-SNG gas stream is removed by the EHC.

Recovery (%) =
$$\frac{p_{H2_{period}}}{p_{H2_{period}}} \frac{p_{H2_{period}}}{p_{H2_{period}}} \cdot 100$$
 (Eq. 1)

During the first experiment with raw bio-SNG, the flow of bio-SNG to the EHC was set at three different values: 118, 308 and 630 NmL/min per cell. These flows are normalized to flow per cell, for comparing single and multiple cell test results.

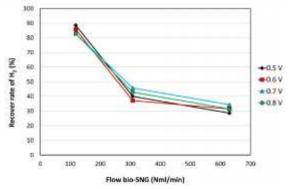


Figure 5: Recovery rate of H₂ in the feed gas (raw bio-SNG feed gas) plotted against total flow of bio-SNG per cell on the set-up with 42.6 cm² membrane area.

At the lowest flow rate of 118 Nml/min the recovery rate reaches between 83 and 89%. Lower flow rates will reach even higher recovery rates, but the flows become too small compared to the membrane area. At the higher flow rates of 308 and 630 Nml/min the recovery rate drops down to the rangs of 37-46% and 29-24% respectively.

During the experiment, the cell potential over each cell was varied between 0.5 V and 0.8 V. Higher cell potentials are not used to prevent degradation of the catalyst material. There is an improvement when the cell potential is increased, with 0.7 V as an optimum. However, the differences are fairly small.

In the second test set-up, a mixture of N_2 and H_2 was used. In this mixture the H_2 concentration was also 3.5%. The flow rates in this test with 285 cm² membrane area are chosen to meet the same normalised flow rates with the set up with 42.6 cm²: 285/42.6 = 6.7 times higher. These flow rate in this test are: 0.79, 2.06 and 4.20 Nl/min. Normalising the flow rates is done to compare both test set-ups.

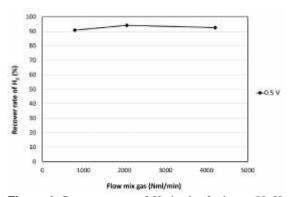


Figure 6: Recovery rate of H_2 in the feed gas (N_2 - H_2 mixture) plotted against total flow on the set-up with 285 cm² membrane area.

The recovery rate on this set-up does not drop significantly at higher flow rates and varies between 91 and 94%, as can be seen in Figure 6. The main difference between both set-ups is the flow pattern that distributes the gas (H₂) to the membrane.

The results of both tests show that it is possible to extract H₂ out of a raw bio-SNG stream at a small scale. It is expected with a further optimised flow pattern the recovery rate can reach above 95% at a lower potential. When scaling up this technology an assessment should be made on costs. For example, a comparison should be made of the electric power needed and the electricity price compared to recovery rate and the hydrogen price. The commercial price of hydrogen varies between 1 and 10€kg depending on the pressure and purity of the hydrogen delivered. The assessment should lead to the most cost effective working point of the EHC.

3.2 Assessment of options for co-production of high-pressure H₂ and bio-SNG

Once the technical feasibility of H₂ recovery was successfully tested at lab-scale, it is necessary to identify the most suitable configuration for the implementation of EHC technology in a bio-SNG process. For this, a 100 MWth bio-SNG plant based on MILENA/OLGA/ESME technologies, utilizing wood chips as feedstock, has been

considered. The three cases, as defined in the introduction, have been studied:

Case a) 100 MW_{th} bio-SNG production (Figure 1). Case b) 100 MW_{th} bio-SNG + H_2 production (Figure 3). Case c) 100 MW_{th} bio-SNG + H_2 production (Figure 4).

For this, simulations in Aspen Plus® have been performed for cases a) to c) in order to determine the overall energy efficiency. Case b) implies the addition to the system of a HyET EHC unit with respect to the base case, whereas case c) adds a WGS unit and 2 EHC units (one for the removal of the bulk of hydrogen before the methanation section, and an extra unit for final polishing of the bio-SNG before grid injection). However, cases b) and c) do not include anymore the high-pressure methanation unit considered in case a), but compression of the bio-SNG product at ~22 bar is performed before grid injection.

For the economic analysis (Table II), the plant revenues have been considered through the EBITDA (Earnings before interest, taxes, depreciation and amortization) and the profitability of the options has been analyzed using the return on investment (ROI). The ROI is defined as the net plant income (EBITDA) divided with the investment cost. For the business case study, the bio-SNG CAPEX data has been taken from a previous ECN report [13]. For cases b) and c), the OPEX is higher due to the electricity demand of the EHC unit. The assumptions for the revenues are 10 €GJ for SNG [11] and for the high purity, high pressure hydrogen a medium price level of 50€GJ (6.7 €kg).

The results of the analysis are summarized in Table I and Table II. As can be observed, case b) does not offer any significant efficiency improvement with respect to the base case a), although the return on investment, ROI, increases from 0.3% to 1.1%. This value remains, however, still unattractive. On the contrary, if case c) is implemented, the overall energy efficiency (chemical energy contained in the products/energy contained in the solid biomass) can be increased from 69% to 73%, and the ROI dramatically increases to 9.5%, a value that can be considered attractive for investment. On the other hand, the analysis reveals that that the bio-SNG base case would need subsidies in order to reach a reasonable ROI. If, for example, an additional subsidy is given of 8 €GJ, giving a total of 18 €GJ of revenues, the ROI would be in the order of 7%, which corresponds to about 14 years.

Table I: Summary of results of simulations of layouts for integration of hydrogen recovery in a 100 MW_{th} bio-SNG plant.

	Cases		
	a)	b)	c)
Bio-SNG (MW)	69.1	67.6	50.5
$H_2(MW)$	0	1.7	22.4
Overall energy efficiency (%, LHV based)	69.1	69.3	72.9

Table II: High level business case of layouts for integration of hydrogen recovery in a 100 MW_{th} bio-SNG plant.

	Cases		
	a)	b)	c)
OPEX (M€)	19.3	19.6	22.6
Revenues SNG (M€)	19.9	19.5	14.5
Revenues H_2 (M \mathfrak{E})	0	2.5	32.3
EBITDA (M€)	0.6	2.4	24.2
CAPEX Bio-SNG (M€)	231	216	221
CAPEX H_2 recovery (M€)	0	2	35
CAPEX total (M€)	231	218	256
ROI (%)	0.3	1.1	9.5

And for case a) it demonstrates

The sensitivity of the plant revenues, for the cases a) to c), to the sell price of bio-SNG and hydrogen has been assessed. Three price levels have been chosen for hydrogen, whereas the price of SNG was kept fixed at 10 €GJ. The results of the sensitivity analysis are shown in Figure 7. As can be observed, for case c), the plant that is designed to favor the yield of hydrogen, a medium price level for hydrogen > 50 €GJ is necessary for a ROI that is close to 10%. The recovery of hydrogen after methanation (thus, favoring the production of bio-SNG) hardly improves the base case.

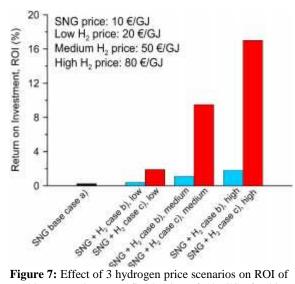


Figure 7: Effect of 3 hydrogen price scenarios on ROI of a 100 MW_{th} plant at a fixed price of 10 €GJ for bio-SNG.

4 CONCLUSIONS AND OUTLOOK

This paper has analyzed the technical and economic feasibility of implementing co-production schemes of bio-SNG and hydrogen from biomass gasification through the combination of ECN ESME and HyET EHC technologies. The recovery of high-pressure, high-purity hydrogen from producer gas (a promising biofuel with good market prospects) boosts the value of the plant

products, thus effectively contributing to decrease the production cost of bio-SNG.

H₂ extraction from raw bio-SNG stream has been proven at a small scale in this work. The experimental results have shown that hydrogen can be extracted from raw bio-SNG up to 89% in a setup with a non-optimal flow distribution at relatively low flow rates. With a slightly optimized flow distribution, the recovery rates can reach 94% at higher flow rates. On the other hand, a basic economic assessment of different options for the implementation of hydrogen and bio-SNG co-production has revealed that the promotion of hydrogen yield via WGS plus harvesting of hydrogen before methanation has a favorable effect on the overall energy efficiency and the plant profitability. On the contrary, the final recovery of hydrogen after methanation (i.e. promotion of bio-SNG yield) hardly improves the base bio-SNG case.

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