FEASIBILITY OF SNG PRODUCTION BY BIOMASS HYDROGASIFICATION

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Contribution to the "12th European Conference and Technology Exhibition on Biomass for Energy, Industry and Climate Protection"

17-21 June 2002, Amsterdam, the Netherlands

Revisions	
A	
B	

Made by:	Issued by:	
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ABSTRACT: Hydrogen is likely to play an important role in future renewable energy supply. Storage and transport of hydrogen can take place in its free form, or chemically bound e.g. as methane. An important advantage of the indirect use of hydrogen as energy carrier is, that in future renewable energy supply, parts of the existing large-scale energy infrastructure could still be used. Production of Substitute Natural Gas (SNG) by biomass hydrogasification has been assessed as a process for chemical storage of hydrogen. Thermodynamic analysis has shown the feasibility of this process. The product gas of the process has a quality quite comparable to the quality of the Dutch natural gas and might potentially be transported through the existing gas net without any adjustments. In the long term, hydrogen from water electrolysis will become available for the energy market, and can be used within this process. In the short term, the required hydrogen may be obtained from hydrogen-rich gases available as by-product from industrial processes. Results of experimental and modelling work, as well as the economic and ecological feasibility of the process, application potentials in the Netherlands, and plans for future development are presented.

Keywords: feasibility studies, SNG, hydrogasification

1 INTRODUCTION

Hydrogen, generated from renewable sources, is likely to play a major role as an energy carrier in the future energy supply. Due to the finiteness of fossil energy sources, and the global environmental damage caused by them, the world has to switch gradually to other primary energy sources. In the long term, only biomass and other renewable sources such as water, wind, and sun will be available. Most of these energy sources, however, have a fluctuating character, resulting in dissimilarities between energy availability and energy demand. Discrepancies between demand and supply of energy can be solved by temporary storage of the surplus of energy as hydrogen, through water electrolysis.

The storage and transport of hydrogen can take place in its free form (H_2), or chemically bound e.g. as methane or methanol. However, the storage and transport of hydrogen in its free form are more complex, and would probably require more energy than the storage and transport of hydrogen in chemical form. Several routes for chemical storage of hydrogen have been studied[1], from which SNG production by biomass hydrogasification has been identified as the option with the highest energetic efficiency.

Between the present fossil fuel-based energy supply system and the future hydrogen economy, there would be a long transition phase, during which both fossil and renewable sources of energy would be applied simultaneously. During this phase hydrogen, produced from renewable sources, might be introduced to the energy market by the biomass hydrogasification process. The use of the existing gas infrastructure for transportation of the produced SNG makes a gradual transition to a hydrogen economy possible.

The process of SNG production by biomass hydrogasification has to be developed practically.

1.1 Hydrogasification

Gasification of carbon-containing feedstocks in a hydrogen atmosphere is called hydrogasification. Hydrogasification of coal has been investigated since the 1930s in Germany, Great Britain and The United States[2]. Generally, the conversion increases with increasing pressure, temperature and residence time. Carbon conversions over 80%, with a selectivity of 90% for methane and ethane, have been obtained in hydrogasification of brown coal, in a 240 tons per day plant in Germany. In the early 1980s, Steinberg et al[2] carried out flash hydrogasification experiments with wood in an entrained-flow reactor. At pressures between 14-34 bar and temperatures between 800-1000°C, carbon conversions were over 90%.

Several processes have been developed for production of methane-rich gases from coal, biomass, or organic solid wastes[3],[4],[5],[6]. The required hydrogen in these processes is produced within the process[7], e.g. by gasification of residual char from the hydrogasifier. The use of an external hydrogen source is new[8], and gives the possibility to apply the hydrogasification process not only for upgrading of biomass and organic wastes to a methane-rich gas, but also as a process for chemical storage of hydrogen.

2 MODELLING WORK

A simplified flowsheet for SNG production by biomass hydrogasification is shown in figure 1. The complete process has been modelled in the ASPEN[PLUS]
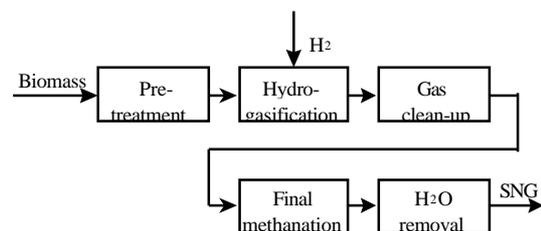

Figure 1: Simplified flowsheet for SNG production by biomass hydrogasification

simulation package. Hydrogen and pre-treated biomass (dried and reduced in size) are fed to the hydrogasifier.

The gasifier is operated autothermally at 30 bar and 850°C. It is modelled as a restricted equilibrium reactor with a temperature approach, in order to obtain a carbon conversion of 80%. The produced gas passes a gas clean-up section for removal of contaminants, followed by a final methanation step for the conversion of residual CO. Removal of H_2O from the product gas of the methanation step, results in SNG as the final product.

For mid-term implementation of the process, a low-temperature gas clean-up is proposed. The gas is cleaned at a temperature of 30°C before being preheated to the inlet temperature of the methanation section (about 250°C). The syngas is methanated in two serial adiabatic methanation reactors according to the reactor concept applied in the Lurgi coal-to-SNG process [9]. The heat released from the hydrogasifier product gas, and the heat generated in the methanation reactors, are used to generate superheated steam (40 bar and 540°C), which enters a steam turbine. A fraction of the partly expanded steam is used to dry the biomass, while the remaining part of the steam is used for power generation.

The simulations are based on willow wood as biomass, with the following specification (obtained from Phyllis[10]): moisture content 11.1 wt%$_{wet}$, volatile matter 81.51 wt%$_{dry}$, ash 1.91 wt%$_{dry}$, fixed carbon 16.58 wt%$_{dry}$, C 48.72 wt%$_{dry}$, H 5.94 wt%$_{dry}$, O 42.74 wt%$_{dry}$, N 0.62 wt%$_{dry}$, S 0.06 wt%$_{dry}$, Cl 0.01 wt%$_{dry}$, and LHV 18.5 MJ/kg$_{daf}$. The amount of biomass will correspond to a thermal input of 100 MW. The main mass and energy flows are presented in table 1, while the specifications of both the SNG product gas, as well as the standard Dutch natural gas are presented in table 2.

Table 1: Mass & energy flows

		Mass kg/s	Energy MW
In:	Biomass	6.182	100.00
	Hydrogen	0.790	94.75
	CO_2	1.182	-
	Power	-	6.73
Out:	SNG	3.894	154.08
	Char	0.533	17.49
	Power	-	10.93
Carbon conversion:		80.1%	
SNG efficiency:		79.1%	

Table 2: Specifications of SNG product gas and the standard Dutch natural gas

|		SNG	Natural Gas
CH_4	[mol.%]	82.97	81.3
H_2	[mol.%]	8.02	0.0
CO_2	[mol.%]	8.37	0.9
C_2^+	[mol.%]	0.00	3.5
N_2	[mol.%]	0.53	14.3
O_2	[mol.%]	0.00	-
H_2O	[mol.%]	0.01	Dew point < -10°C
CO	[mol.%]	0.09	-
LHV	[MJ/kg]	39.57	38.0
LHV	[MJ/Nm³]	30.67	31.7
Wobbe	[MJ/Nm³]	44.03	44.2

3 EXPERIMENTAL WORK

As part of the technical assessment of the biomass hydrogasification concept, an experimental programme

was performed at Deutsche Montan Technologie (DMT), Germany, with willow wood, or char produced from willow wood, as feedstock.

Several isothermal experiments have been carried out in the DMT pressurised thermobalance facility[11]. Typical amount of feedstock was about 500 mg willow wood or 350 mg char. The size of the feedstock was less than 0.7 mm. As gasifying agent hydrogen was used. The tests were run over a period of 3 hours or longer.

During the pyrolysis step, which took place in the first minutes of the thermobalance experiments, a high percentage of biomass was converted, from which a high fraction to methane and ethane, especially at high pressures. Figure 2 presents the biomass conversion, and the release rate of the main carbon-containing components of the product gas at 850°C and two different pressures (1.5 bar and 30 bar). In both cases, a biomass conversion of about 80 wt% was achieved within a minute. The figure shows a shift from carbon monoxide and carbon dioxide to methane, by increasing the operating pressure. This can be explained by a combination of methanation and reversed shift reactions:

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \qquad (1)$$
$$CO_2 + H_2 \leftrightarrow CO + H_2O \qquad (2)$$
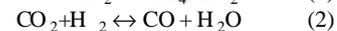
The same trend is observed for the release rate of ethylene and ethane, i.e. a shift from ethylene to ethane by increasing the operating pressure. This can be explained by hydrogenation of ethylene:

$$C_2H_4 + H_2 \leftrightarrow C_2H_6 \qquad (3)$$
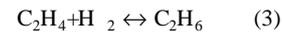
At 850°C and 30 bar, beside 100% formation of the pyrolysis products: C_2H_4, CO, and CO_2, more than 95% of ethane, and about 80% of methane were formed within the first minutes. After that only methane, and in much less extent ethane, continued to be formed, through the
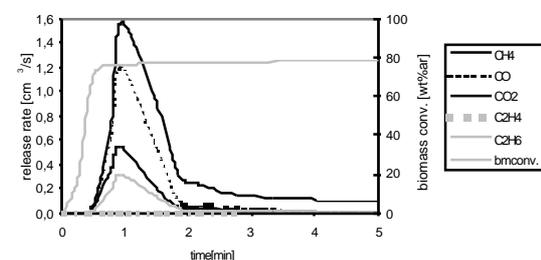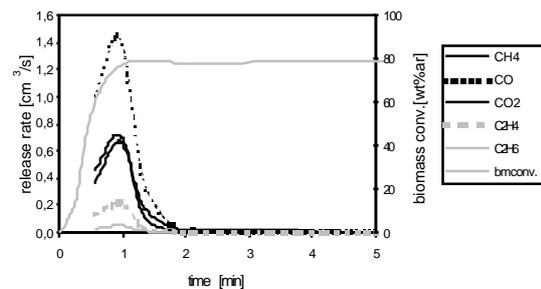

Figure 2: Biomass conversion and release rate of main carbon-containing components in the product gas at 1.5 bar and 30 bar(T=850°C)

hydrogasification of char. Figure 3 presents the mean concentration of the main carbon-containing components in the product gas as a function of pressure. Figure 4 presents the conversion of biomass and carbon, as a function of time, for different pressures. The test runs begin with rapid biomass conversions of 80 wt%, and

carbon conversions of 60 wt%, followed by hydrogasification of the remained char, with a slow reaction rate.

Char gasification with steam resulted in the highest conversion rate, followed by CO_2 and H_2 gasification. Based on this observation, a higher char conversion is expected during the hydrogasification of biomass in a pressurised fluidised bed reactor than in a thermobalance, because in addition to hydrogen, also the pyrolysis products, CO_2 and H_2O can take part in the conversion of char, according to the following reactions:

$$C + 2H_2 \leftrightarrow CH_4 \qquad (4)$$
$$C + CO_2 \leftrightarrow 2CO \qquad (5)$$
$$C + H_2O \leftrightarrow CO + H_2 \qquad (6)$$
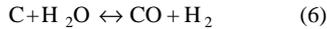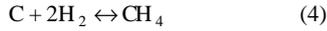
Beside the thermobalance experiments, also some tests were carried out at 5 bar and 800°C in the DMT pressurised fluidised bed (PFB) facility[12]. In summary, the experimental results have shown the feasibility of biomass hydrogasification as the most important step within the total process of SNG production by gasification of biomass in a hydrogen atmosphere, with respect to the following aspects:

- Production of a gas, very rich in methane at the same process conditions (P,T), applied within the previous modelling work.
- Conversion of a sufficient amount of biomass to gaseous product, within a reasonable residence time of the biomass feedstock. The remaining char might be used within the process or to generate steam.
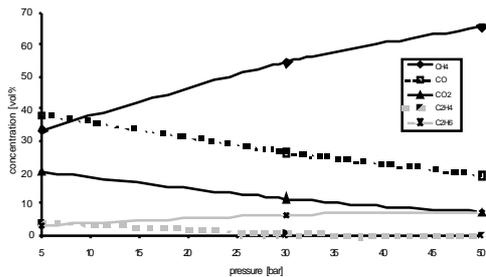

Figure 3: Mean concentration of main carbon-containing components in the product gas at 850 °C as a function of pressure
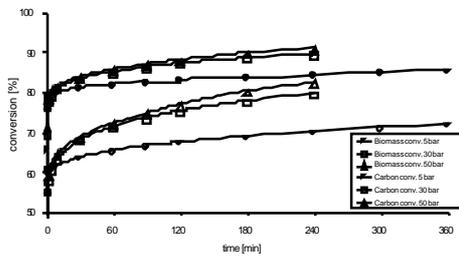

Figure 4: Biomass and carbon conversions at 850 °C as functions of time for different operating pressures

The product gas in the thermobalance tests, as well as in the PFB tests is highly diluted with hydrogen. This is not desired in practice, due to the following reasons:

- In case of a diluted gas, hydrogen should partly be recirculated, resulting in higher costs and process complexity.

- Presence of too much hydrogen in the hydrogasifier might result in a process, which cannot be operated autothermally any more.

These aspects are at the moment, as part of a detailed experimental work on biomass hydrogasification, being studied in a bench-scale pressurised fixed-bed gasifier, which is recently constructed at the University of Twente.

4 REACTOR MODELLING

A kinetic model has been taken from a study performed by Dong and Borgwardt concerning the biomass reactivity in gasificaton by the Hynol process[13],[14]. The model is based on the idea that hydrogasification of biomass involves a rapid devolatilisation and pyrolysis reaction of the volatile matter in biomass, and a very slow reaction of residual carbon with the process gas. The first reaction comprises the homogeneous gas phase reactions (1), (2), and (3), whereas the heterogeneous reaction (4) occurs during the whole experimental period.

The two reactions are considered to initiate simultaneously and are first order with respect to the remaining solid reactants, and have the following rates:

$$\frac{dX_1}{dt} = k_1(X_C - X_1) \qquad \text{and} \qquad \frac{dX_2}{dt} = k_2(1 - X_C - X_2)$$

with X_1 and X_2 the biomass conversions by the rapid and slow reactions at time t, X_C the maximum attainable conversion by the rapid reaction, and k_1 and k_2 the reaction rate constants, with temperature dependency implemented by the Arrhenius equation. The maximum attainable conversion by the rapid reaction can be correlated as a function of temperature in Kelvin by:

$$X_C = a + b\ T$$

where α and β are parameters that have to be determined empirically. The parameters involved in the model can be determined by fitting the redefined equation of the total biomass conversion to the experimental conversion data obtained from the DMT thermobalance experiments.

The kinetic model, combined with a hydrodynamic model[15],[16],[17], are used to determine the dimension of bubbling fluidised bed hydrogasifiers at different biomass feeding rates.

The amount of biomass present in the reactor depends on the time dependent biomass conversion and the feed rate of the biomass. The biomass conversion X_{ar} can be rewritten into an equation for the variation of the biomass with the time (with C_A the weight fraction of ash, obtained from ultimate analysis of the original sample):

$$F(t) = 1 - (1 - C_A) \cdot X_{ar}$$

Under the assumption that after residence time t the remaining biomass will be removed from the reactor the mass of the biomass in the reactor can be determined by:

$$M(t) = \Phi_{m,bm} \cdot \int_0^t F(t)\ dt$$

The surface of the reactor depends on the gas velocity in the fluidised bed, u, and the feed rate of the hydrogen gas. Based on the determined gas velocity, and the amount of gas flowing through the reactor, $F_{v,g}$, the surface of the reactor is determined by:

$$S_{reactor} = \frac{\Phi_{v,g}}{u}$$

The height of the reactor depends on the reactor surface and the amount of biomass in the reactor. However, the bed will also contain a certain amount of sand, α_{sp}, and will have a certain porosity, ε. The height of the reactor, therefore, is given by:

$$H_{reactor} = \frac{M_{bm}}{\rho_{bm} S_{reactor} (1 - \alpha_{sp})(1 - \varepsilon)}$$

The determination of the dimensions of the PFB gasifiers is based on a desired carbon conversion of 80%. For such conversions, based on the kinetic model, a residence time of approximately 1 hour will be required. As a bench-scale gasifier should not become too large, the bed diameter is set to 15 cm. A large-scale gasifier is designed to handle a 100 MW$_{th}$ biomass input. As good mixing will be required in order to maintain a uniform bed temperature, the amount of sand in the bed is set to 90%. In order to be economically feasible as well, the system is evaluated for biomass particles with diameters of 1.0 to 3.0 cm. The diameter of the sand particle is taken as 0.5mm. The results are presented in table 3.

Table 3: Results of bench-scale and large-scale reactor modelling

Biomass fraction [%]	Bed diameter [m]	Bed height [m]	$\Phi_{biomass}$ [g/s]	Superficial gas velocity [m/s]
Bench-scale hydrogasifier				
50	0.15	0.35	0.48	0.049
Large-scale hydrogasifier				
10	6.30	11.90	5790	0.335

5 ECONOMIC & ECOLOGICAL FEASIBILITY

Based on the availability of biomass and hydrogen in the Netherlands, the amount of produced SNG is determined. The SNG production costs are determined by taking into account the costs of biomass and hydrogen, the investment costs, and the operating and maintenance costs. Considering the costs of natural gas, the additional production costs of the SNG, as well as the costs per tonne CO_2 avoided can be determined.

The availability of biomass is related to the availability of residual, cultivated and imported biomass, taking into account the biomass consumption due to feeding to coal-fired power plants and waste incineration stations. The availability of residual biomass is supposed to linearly increase from 159 PJ in the year 2000 to 196 PJ in 2020 [18]. The availability of cultivated biomass is estimated to be between 0 and 50 PJ, whereas the availability of imported biomass will be between 0 and 200 PJ[18]. The application of willow wood is considered to be CO_2 neutral. The costs are expected to lie between 0 and 3.6 € per GJ (0 to 63.5 € per dry tonne), with an expected value of 2.5 € per GJ [19],[20].

Based on an inventory of hydrogen(-rich) residue gases[21], the total hydrogen(-rich) residue gases available in the Netherlands is estimated to amount to 2884 million Nm³ (19.66 PJ of hydrogen), present at approximately 7 locations. For the market share of the hydrogasification process with regards to the available hydrogen, an uncertainty factor of 50% is assumed. Large differences in the available amount of hydrogen at different locations, result in large variations in the implemented scale of a hydrogasification process and the related specific investment costs [19]. As a result of the application of hydrogen(-rich) residue gases, the hydrogen costs are equal to the costs of natural gas, required as a replacement within the current application of the residue gases. The natural gas costs are related to the mineral oil costs and, therefore, take into account the uncertainty of mineral oil costs and dollar exchange rate.

Based on a market share of the hydrogasification process of 50% [19], a plant capacity factor of 90%, as well as the minimum and maximum availability of hydrogen at the various locations, a hydrogasification process with thermal biomass input varying between 5 and 160 MW, with an expected value of 50 MW, might be constructed. The specific investment are based on the costs of atmospheric Circulating Fluidised Bed (CFB) power generating systems [22] with low-temperature gas clean-up. By taking the costs of the gas engine within these systems into account, the estimated costs of the hydrogasification process are reduced. Due to the absence of cyclones within a Bubbling Fluidised Bed system, the costs of the CFB-systems are reduced with 30%, to determine the costs of a BFB-system. By means of the Williams 0.6 scaling rule, the cost of the 5, 50 and 160 MW$_{th}$ BFB-systems are determined. In addition, a pressure factor[23] is applied, in order to estimate the cost of the BFB-systems at a pressure of 30 bar. The costs of the methanation reactors are derived from the cost of methanation of syngas, obtained from oxygen-blown gasification. This gas will have a relatively high amount of CO, and a relatively low heating value, compared to the syngas obtained from the hydrogasifier. By relating the investment costs to the thermal biomass input of the gasifier, an estimation of the methanation within the hydrogasification process can be made. The calculated specific investment costs are presented in table 4.

The expected SNG production costs, the potential of SNG production until 2020, and the SNG production in 2020 are presented in table 5. The SNG production within one year (in this case 2020) represents the potential SNG production per year from the moment that the hydrogasification process is completely developed and implemented. According to the "Third White Paper of the Netherlands Government on Energy Policy", the share of renewables in the primary energy supply should increase from 1% in 1990 to 10% in 2020 (288 PJ). 26% of this share of renewables in the primary energy supply should result from implementation of biomass (75 PJ). With a potential SNG production capacity of 5.7 PJ in 2020, the hydrogasification process has the potential to fulfil approximately 2% of this goal (or even ±7.5% of the biomass related goal).

By producing SNG a certain amount of CO_2 emission is avoided, due to the replacement of natural gas by SNG. The specific environmental costs involved can be determined by relating the additional production costs to the avoided CO_2 emission. However, as the applied hydrogen residue gas, that normally is used for e.g. firing purposes, has to be replaced by fossil-based fuel, the produced SNG will only be partly CO_2 neutral. The expected costs per tonne CO_2 avoided are equal to 157 € Based on the potential SNG production until 2020 an expected potential reduction of CO_2 emission of 124 ktonne a year has been determined.

Table 4: The specific investment costs of willow wood hydrogasification

Investment costs	Biomass input (MW)		
	5	50	160
Total investment costs [M €]	9.4	33.0	62.4
Specific investment costs [€/W]	1.880	0.661	0.391

Table 5: SNG production potential until 2020 and SNG production costs in case of hydrogasification of willow

	Mean
SNG production costs [€/GJ]	6.6
SNG production capacity until 2020 [PJ]	60
Maximum SNG production capacity [PJ/yr]	5.7

6 CASE STUDIES

An important aspect in the evaluation of the biomass hydrogasification concept is its market applicability. This aspect is investigated by carrying out the following three case studies:

- Case 1: hydrogasification of willow wood with an external hydrogen-rich residue stream from methanol synthesis;
- Case 2: hydrogasification of organic residues from the food industry, as well as hydrogasification of sewage sludge, with external hydrogen from a hydrogen grid;
- Case 3: hydrogasification of cacao shells and hulls with internal hydrogen, produced by gasification of residual char from the hydrogasifier (Feldmann concept [4],[5],[6]).

Case 1 is situated at Methanor (Delfzijl), a methanol producer, where large amounts of hydrogen(-rich) residue streams are available at high pressures of about 100 bar. Willow wood is considered as feedstock, as Methanor is easy to reach from the sea. Case 2 is situated at Bergen op Zoom, in the vicinity of a hydrogen grid operated by Air Liquide. As the costs of hydrogen from the grid will be higher than the costs of hydrogen(-rich) residue gases, it is decided to use waste streams (organic residues from the food industry, and sewage sludge) as biomass source. These waste streams are available in the direct vicinity of the selected location. The third case results from the Feldmann concept of hydrogasification. The hydrogasification of biomass is based on hydrogen, produced within the process by gasifying the char, remained from the hydrogasifier. Zaandam has been selected for this case, as cacao shells and hulls, that might be an interesting fuel for this concept of hydrogasification, are available in large amounts in the vicinity of this location. In all three selected locations, extensive natural gas infrastructure for transport of the produced SNG is available.

Thermodynamic analysis, carried out for each case with a biomass thermal input of 100 MW, has resulted in SNG production efficiencies of 76.8-82.6% (on LHV basis). The produced SNG has, in spite of a different composition, a comparable quality to the standard Dutch natural gas. The hydrogen content of the SNG is below 12 mol%. At such hydrogen concentrations, the produced SNG might potentially be transported through the existing gas infrastructure without any adjustment. According to table 6, the SNG production costs are in the range of 2.5 to 8.3 €/GJ. For comparison, the price of natural gas for large consumers is about 3.2 €/GJ, and for domestic consumers (excluding the taxes) is about 6.8 €/GJ. Hydrogasification of sludge, in spite of using hydrogen from the grid and assuming a 25% additional investment for the gasifier, compared with hydrogasification of willow, has resulted in the lowest SNG production cost, due to the assumption of negative value for the biomass costs. This production cost is even lower than the price of natural gas for large consumers, and therefore promising.

When determining the costs per tonne CO_2 avoided it is assumed, that hydrogasification of sewage sludge is not CO_2 neutral, hydrogasification of cacao shells and hulls is considered to be CO_2 neutral, while both hydrogasification of willow and organic residues are only partly CO_2 neutral and the amount of fossil-based hydrogen has been taken into account.

Table 6: SNG production costs and costs per tonne CO_2 avoided for 100 MW_{th} systems

Case	SNG prod. costs [€/GJ]	CO_2 reduction costs [€/tonne]
Willow	4.7	133
Organic residues	7.6	197
Sewage sludge	2.5	-
Cacao	8.3	91

7 FUTURE WORK

The first phase of the ECN RD&D programme on SNG production by biomass hydrogasification process comprises an assessment of technical and economic prospects of the concept. The final evaluation of this phase involves an additional experimental research in the bench-scale pressurised fixed-bed gasifier, at the University of Twente. Finally, within the framework of a new project, recently started at ECN, the potential of different SNG production routes, will be compared. This will lead to the decision, whether ECN will, on the short term, continue the research on SNG production from biomass, either by hydrogasification or by an alternative conversion process.

8 CONCLUSIONS

Based on thermodynamic and flowsheeting analysis, the feasibility of SNG production by biomass hydrogasification has been demonstrated. The autothermal operation of a hydrogasifier at a specific hydrogen-to-biomass ratio is possible and will result in a final product gas with a quality comparable to that of standard Dutch natural gas. If 10 to 12 mol% hydrogen would be acceptable, the produced SNG can be transported to end-users through the existing natural gas infrastructure. The energetic efficiency of SNG production from willow wood and hydrogen by hydrogasification is up to 80% on low heating value basis.

According to reactor modelling, using the same hydrogen-to-biomass ratio as in the flowsheeting analysis, it is possible to realise a bubbling fluidised bed hydrogasifier with acceptable dimensions. Both the

reactor modelling and the flowsheeting analysis are based on the results of the experimental programme performed at Deutsche Montan Technologie (DMT) in Germany.

The experimental programme has shown the feasibility of biomass hydrogasification with respect to the production of a methane-rich gas at the same process conditions (P,T) as used in the modelling work. It has also demonstrated the conversion of a sufficien t amount of biomass to a gaseous product within a reasonable residence time of biomass feedstock.

Based on the application of hydrogen(-rich) residue gases in the Netherlands, the market potential and the avoided CO_2 emissions of the hydrogasification con cept are relatively low, however, these quantities will increase, if in addition to hydrogen(-rich) residue streams also other hydrogen sources (e.g. hydrogen from existing hydrogen grids) can be used or when a more global market potential is evaluated.

The case studies have shown that hydrogasification of sewage sludge, in spite of using hydrogen from a hydrogen grid and assuming a 25% additional investment cost for the gasifier, results in an SNG production cost lower than the current market price of nat ural gas. Hydrogasification of cacao shells and hulls, with internal hydrogen source, will be promising at higher natural gas prices, or with current gas prices at thermal biomass inputs above 100 MW.

It seems therefore possible to increase the Dutch marke t potential of the hyd rogasification concept by using other hydrogen sources than hydrogen(-rich) streams in special applications. At the moment, however, SNG production by biomass hydrogasification in the Netherlands can only be economically feasible for specific applications (waste treatment). Production of "green gas" by th e hydrogasification process still requires additional financial measures, in order to compete with natural gas.

ACKNOWLEDGEMENTS

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