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FEASIBILITY OF BIOMASS / WASTE-RELATED SNG PRODUCTION TECHNOLOGIES

Final report

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This project is realised by financial support of the NECST- Programme, that Novem carries out in order of the Dutch Ministry of Economic Affairs. The Novem project number is: 249-01-03-12-0001

ERRATA

Within this project a potential market share of 1.5% to the total Dutch natural gas consumption of 1500 PJ has been assumed for the SNG production through the biomass gasification / methanation routes. Based on this assumption, the SNG production capacity until 2020, the avoided CO_2 emission until 2020, the maximum annual SNG production capacity, and the maximum annual avoided CO_2 emission for biomass gasification / methanation routes are higher than the values presented in the abstract, table 5, chapter 4, and table 7.1. The reported, as well as the correct values for each case are presented in the table below. These corrections have no effect on other results presented in this report.

		gasification / methanation	
		oxygen-blown	indirect
SNG production capacity until 2020	[PJ]	157 → 237	128 → 191
Maximum annual SNG production capacity	[PJ/yr]	14.9 → 22.5	15.1 → 22.5
Avoided CO_2 emission until 2020	[Mtonne]	8.8 → 13.3	7.2 → 10.7
Maximum annual avoided CO_2 emission	[Mtonne/yr]	0.84 → 1.26	0.84 → 1.26

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Account

The work described in this report has been performed by the Business Unit Biomass of the Energy Research Centre of the Netherlands, ECN, in cooperation with Gasunie Trade & Supply. This project has been carried out within the framework of the Novem-NECST Programme. The Novem project number is 249-01-03-12-0001, and the ECN project number is 7.2253.

Abstract

Based on comparable basic assumptions, a technical, economic, and ecological assessment has been performed for the biomass/waste-related SNG production technologies, with the objective to make a selection for future implementation of the most promising options. Anaerobic digestion, upstream pressurised oxygen-blown or indirect gasification with downstream methanation, hydrogasification followed by methanation, and, in less extent, the co-production of Fischer-Tropsch-SNG have been studied within this project. Although substitute natural gas (SNG) has not yet been incorporated as an alternative fuel in the Dutch and European renewable energy policies, it can play an important role in realisation of the Dutch and European climate and renewable energy targets. Based on the modelling results, the upstream pressurised oxygen-blown or indirect gasification with downstream methanation routes seemed to be the most promising options for SNG production from biomass. The RD&D trajectory for candidate SNG production processes, consisting of three development stages (Proof of Principle, Proof of Concept, demonstration of a complete integrated process on pilot scale), is expected to take about 10 years for indirect gasification and 8 years for pressurised oxygen-blown gasification. Also a period of four years is expected to be necessary for the market introduction. The indirect gasification technology, after development, will score better with respect to all aspects, compared to pressurised oxygen-blown CFB gasification, and therefore it has been selected for future development for SNG production. Through this gasification technology almost 100% carbon conversion can be achieved, resulting in a tar-free (by using the ECN OLGA process), low-nitrogen, and high-hydrocarbon content synthesis gas. The technology has also the potential of upscaling to a commercial scale on the long term. In combination with downstream methanation, SNG production efficiencies up to 70% can be achieved. The SNG production cost for this process is 7.8 €/GJ (compared to the current market price of 8.7 €/GJ). With a potential SNG production capacity of approximately 15 PJ a year, this process has the potential to fulfil approximately 5% of the Dutch renewable energy target in 2020, or even 20% of the biomass-related target. The SNG production capacity of this process until 2020 is 128 PJ. The costs per tonne CO_2 avoided and the total avoided CO_2 emission until 2020 are € 83, respectively, 7.2 Mtonne.

Keywords

Biomass, green gas, substitute natural gas, SNG, biofuels, cogeneration, digestion, gasification, hydrogasification, methanation, Fischer-Tropsch, feasibility, modelling, Aspen Plus

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EXECUTIVE SUMMARY

Introduction

Within the Dutch renewable energy policy an important role is foreseen for the application of biomass and waste. 42% (120 PJ_{th}) of the policy targets for 2020 (288 PJ_{th} = 10% renewables) has to be realised by substituting fossil-based energy carriers with biomass (75 PJ_{th}) and waste (45 PJ_{th}).

In the short term (< 5 years) biomass will mainly be used for direct and indirect cofiring in conventional power plants. In mid term (5-10 years) it is expected, that the technology for stand-alone decentralised CHP would be mature enough to become an important option. However, in order to satisfy the long-term (> 10 years) expectations of using biomass in the future energy infrastructure, this renewable energy source should also be used as a substitute for fossil fuels in other market sectors. To realise this, conversion technologies should be developed to produce secondary energy carriers from biomass (such as Fischer-Tropsch diesel, methanol, bio-ethanol, SNG, or H_2), substituting the present conventional energy carriers based on petroleum and natural gas. Such a substitution will be potentially capable of saving a significant amount of fossil-related energy, with the resulted ecological advantages.

Concerning the SNG production routes from biomass, the produced “green gas” can be transported through the existing natural gas infrastructure to places where it will be used for the desired energy functions. The energy distribution companies have in this way a strong alternative in addition to “green” electricity at their disposal, in order to achieve their renewable energy targets.

Compared to large-scale (centralised) applications of biomass, SNG production will prevent eventual market problems for products (mainly heat), while thanks to the existing gas infrastructure a large market and certainty in delivery of SNG are guaranteed. On the other hand, using biomass for decentralised CHP applications is not always possible, due to the lack of market for the produced heat. The facility aspect of SNG transport through the existing natural gas infrastructure can play an essential role in the successful market introduction of biomass and waste within the national energy infrastructure on mid and long term.

For transport of hydrogen either a new infrastructure would be necessary, or it might in long term be added to the natural gas infrastructure up to some extent. With respect to SNG, however, as the quality (calorific value, Wobbe-index) of the gas can be matched to the quality of natural gas, there will be no limitation in mixing ratio of these two gases.

For the Netherlands, the biomass-related SNG production route is an interesting option to import renewable energy. Another option is to trade in (green) certificates, by converting biomass (at its origin) to SNG. This is in accordance with the Kyoto agreements, concerning the recognition of the role of emission trading and other economic flexibility mechanisms.

Problem definition

In recent years research on SNG production from biomass / waste in the Netherlands has been focussed on two technologies, viz. combined gasification / methanation (Gastec)[2], and combined hydrogasification / methanation (ECN)[3][4][5][6][7]. However, a technical, economic, and ecological assessment of these processes, based on comparable basic assumptions, had not been performed yet, and is therefore a part of this project. Beside the above mentioned technologies, there are a number of other potential interesting SNG production technologies, which are also evaluated within this project.

Objective

The objective of this project is to make a selection for future implementation of the most promising technologies for the production of SNG from biomass and waste.

Modelling work

The following gasification-based SNG production routes have been considered within this study:

- Pressurised oxygen-blown CFB gasification followed by methanation.
- Atmospheric indirect gasification followed by methanation.
- Pressurised BFB hydrogasification followed by methanation.
- Pressurised oxygen-blown CFB gasification followed by Fischer-Tropsch synthesis and methanation.
- Atmospheric indirect gasification followed by Fischer-Tropsch synthesis and methanation.

The first three SNG production routes have been modelled within the Aspen Plus simulation package, in accordance to the flow sheets presented in figures 1 to 3, while the last two (cogeneration of Fischer-Tropsch liquids and SNG) have been modelled within the software package MS-Excel, in accordance to the general flowsheet presented in figure 4.

Pressurised O_2-blown biomass gasification / methanation process

A CO_2 stream is used to pressurise the biomass to the gasification operating pressure. The produced syngas from the oxygen/steam gasifier, after a low-temperature clean-up, and passing through a methanation reactor, is used for the production of SNG as main product. The heat generated at various points in the process is used for steam and electricity generation in a steam cycle, in order to satisfy the demand within the system. The system pressure is 15 bar, from gasification through methanation. Pressure drop has not been modelled.

Indirect biomass gasification (Battelle) / methanation process

The indirect gasification process (Battelle) is operated at atmospheric pressure; hence, the product gas from the gasifier has to be compressed to 15 bar. Again, pressure drop has not been modelled. The heat required within the gasifier is provided by the combustion of char in a separate reactor.

Biomass hydrogasification / methanation process

Hydrogen and pre-treated biomass (dried and reduced in size) are fed to the hydrogasifier. A CO_2 stream is used to pressurise the biomass to the hydrogasification operating pressure. The produced methane-rich syngas, after gas clean-up, is used for the production of SNG as main product. The heat generated at various points in the process is used for steam and electricity generation in a steam cycle, in order to satisfy the demand within the system. The system pressure is 30 bar, from hydrogasification through methanation.

Cogeneration of FT-liquids / SNG from biomass gasification processes

As during Fischer-Tropsch synthesis one mole of carbon monoxide reacts with approximately two moles of hydrogen, the bio-syngas from the gasifier is shifted to a hydrogen-to-carbon-monoxide ratio of two, before entering the Fischer-Tropsch reactor. The gaseous stream leaving the Fischer-Tropsch reactor is then used for SNG production. In contrast to Fischer-Tropsch synthesis, methanation will require a hydrogen-to-carbon-monoxide ratio of approximately three. During methanation all CO, remaining after the FT-synthesis, is converted to methane. Hydrocarbons, already present in the syngas, are assumed to behave as inert during methanation.

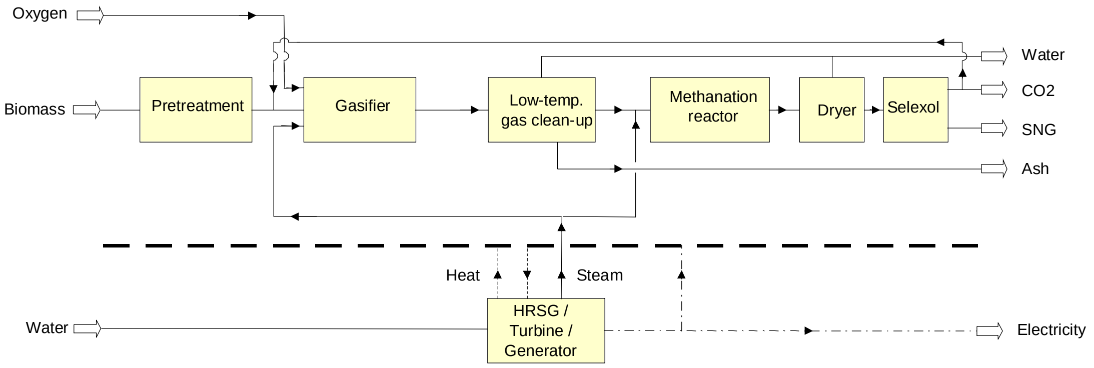

Figure 1 *Aspen Plus model for SNG production by a combined pressurised O_2-blown biomass gasification / methanation process*

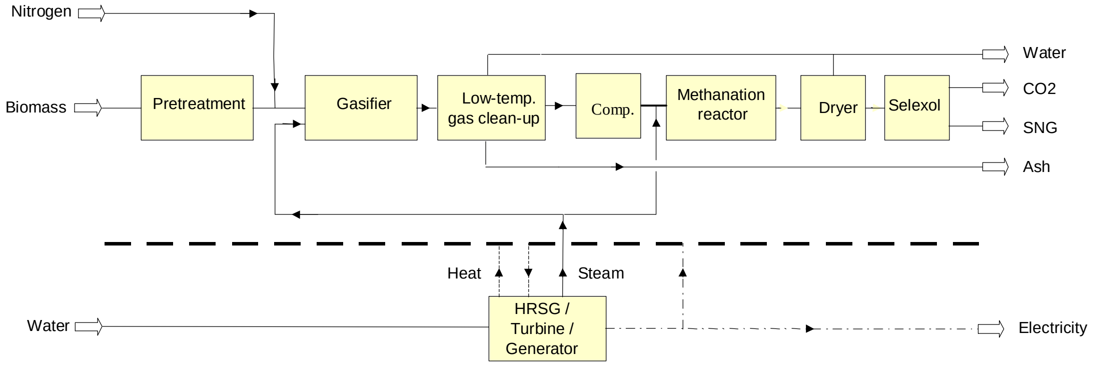

Figure 2 *Aspen Plus model for SNG production by a combined indirect biomass gasification (Battelle) / methanation process*

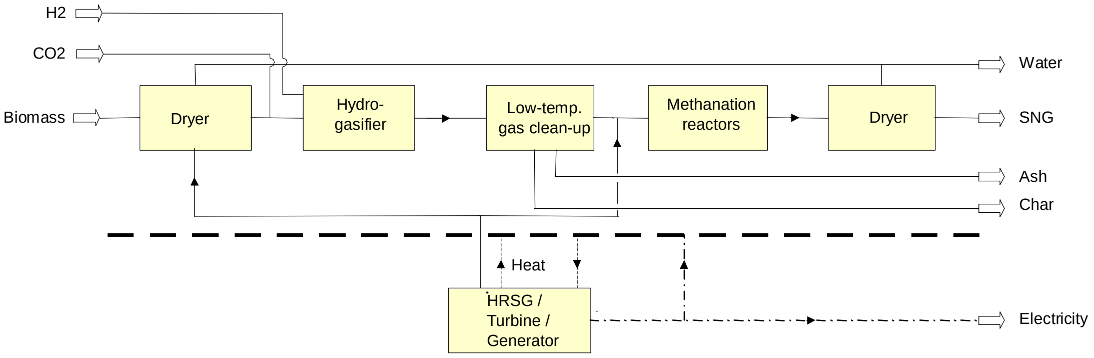

Figure 3 *Aspen Plus model for SNG production by a combined biomass hydrogasification / methanation process*

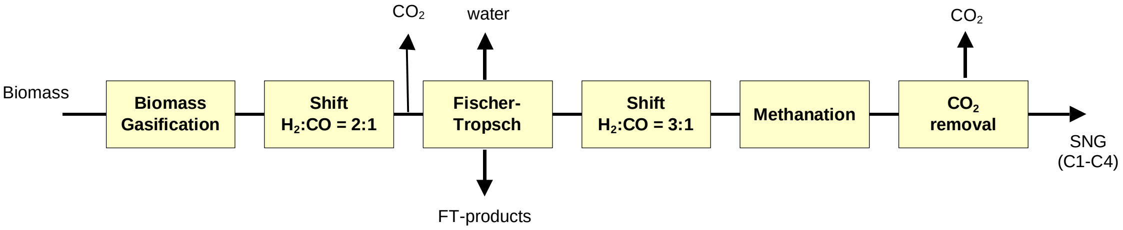

Figure 4 *General flowsheet of the cogeneration of FT-liquids and SNG from biomass*

Gas clean-up

The applied low-temperature gas clean-up consists of a dust filter, a quench, also serving as acidic wash for removing NH_3, a neutral wash for removal of sulphur components and halogens, a special wash (the ECN OLGA process) for reducing the tar, heavy metals and ammonia content, and guard beds in order to protect the Fischer-Tropsch or methanation catalysts against halogen and sulphur compounds (especially HCl and H_2S).

Methanation

The methanation process is based on the intercooled methanation process used within the Lurgi coal-to-SNG process.

Gas conditioning

The product gas is then cooled in a number of steps to condense water, in order to achieve a dew-point lower than -10ºC (at the gas-delivery pressure). This prevents condensation problems within the gas net, and therefore, corrosion problems[2].

In case of the gasification processes the Wobbe-index of the gas has been brought within the G-gas specification, by removal of the excess CO_2 from the gas in a Selexol unit. In case of hydrogasification process, the required Wobbe-index has been achieved by adjustments within the methanation section.

General and specific assumptions

For economic analysis of different cases several general assumptions have been made, as presented in table 1. Beside the general input parameters, each (hydro)gasification process has some specific input parameters, which are presented in tables 2 to 4.

Table 1 *General assumptions for the economic evaluation of the SNG production from different biomass (hydro)gasification processes*

Name	@Risk*	Minimum	Expected value	Maximum	Unit
Availability residual biomass	Linear	159 (in 2000)	-	196 (in 2020)	[PJ]
Uncertainty factor residual biomass	Normal(1)	normal distribution (μ=1, σ=0.1)			[-]
Availability cultivated biomass	Uniform(25)	0	-	50	[PJ]
Availability imported biomass	Uniform(1000)	0	-	2000	[PJ]
Biomass fed to coal-fired stations	Pert(25.8)	5	25	50	[PJ]
Biomass fed to waste incineration stations	40	12	-	40	[PJ]
Plant capacity factor	0.9	-	-	-	[-]
Specific operating and maintenance costs	Pert(0.05)	0.03	0.05	0.07	[-]
Biomass costs	Pert(2.3)	0	2.5	3.6	[€/GJ]
Mineral oil costs	Pert(20.2)	15	20	26	[$/Barrel]
Ash/carbon processing	Pert(68)	61	68	75	[€/tonne]
Dollar exchange rate	Pert(1.17)	0.91	1.18	1.36	[€/$]
Natural gas costs	related to the costs of mineral oil				[€/GJ]
Interest rate	Pert(0.06)	0.04	0.06	0.08	[-]
Depreciation period	Uniform(13)	10	-	15	[yr]
Specific CO_2-emission of natural gas	56	-	-	-	[kg CO_2/GJ]

* *Either the (fixed) value of the parameter or the distribution of the parameter based on minimum, maximum and expected value is presented (with the calculation value mentioned in parenthesis). The depreciation period, as well as the required time for RD&D and market introduction should have a round value. The Normal, Pert, Cumul, and Uniform distribution are described in the appendix II.*

Table 2 *Specific assumptions for the economic evaluation of the willow wood gasification for SNG production in case of a pressurised O_2-blown gasifier*

Name	@Risk	Minimum	Expected value	Maximum	Unit
Natural gas consumption	Pert(1500)	1400	1500	1600	[PJ]
Potential market share SNG	Pert(1.5)	0	1	5	[%]
Specific investment costs	Pert(485)	386	482	593	[€/kW_{th}]
Required RD&D time*	Pert(8)	7	8	9	[yr]
Required time for market introduction	Pert(4)	3	4	5	[yr]
Biomass efficiency:	0.663	-	-	-	[-]
Electricity price (grey)	Pert(3.6)	2.5	3.8	4.0	[€ct/kWh]
Power requirement	2.7	-	-	-	[MW_e]

* Starting-point within the feasibility study has been the year 2000

Table 3 *Specific assumptions for the economic evaluation of the willow wood gasification for SNG production in case of an indirect gasifier*

Name	@Risk	Minimum	Expected value	Maximum	Unit
Natural gas consumption	Pert(1500)	1400	1500	1600	[PJ]
Potential market share SNG	Pert(1.5)	0	1	5	[%]
Specific investment costs	Pert(440)	340	449	506	[€/kW_{th}]
Required RD&D time	Pert(10)	9	10	11	[yr]
Required time for market introduction	Pert(4)	3	4	5	[yr]
Biomass efficiency:	0.670	-	-	-	[-]
Electricity price (grey)	Pert(3.6)	2.5	3.8	4.0	[€ct/kWh]
Power requirement	2.8	-	-	-	[MW_e]

Table 4 *Specific assumptions for the economic evaluation of the willow wood hydrogasification for SNG production*

Name	@Risk	Minimum	Expected value	Maximum	Unit
Availability hydrogen gas	Cumul(7.1)	0	-	20	[PJ]
Heating value hydrogen gas	120	-	-	-	[MJ/kg]
Costs hydrogen gas		related to the natural gas costs			[€/GJ]
Biomass-efficiency*	1.5408	-	-	-	[-]
Hydrogen-efficiency*	1.6262	-	-	-	[-]
Required RD&D time	Pert(8)	7	8	9	[yr]
Required time for market introduction	Pert(4)	3	4	5	[yr]
Market share with regards to available H_2	Pert(0.5)	0	0.5	1	[-]
Specific investment costs**	Pert(819)	413	616	1,541	[€/kW_{th}]
Electricity benefits (green)	Pert (10.3)	9.3	10.6	10.8	[€ct/kWh]

* The biomass and hydrogen efficiencies are defined as the thermal output of SNG, divided by either the thermal input of biomass, or the thermal input of hydrogen

** The specific investment costs are directly related to the availability of hydrogen gas, as in case of low availability of hydrogen gas the scale of the hydrogasifier might be restricted by the availability, hence the specific investment costs becoming relatively high

The main results of the technical, economic, and ecological analyses of the different gasification-based SNG production routes are summarised in table 5. Table 6 presents the production cost, as well as the market price of SNG for different cases.

Table 5 *Evaluation data for gasification-based SNG production routes*

		gasification/methanation		hydrogasification	FT-SNG	
		oxygen-blown	indirect		oxygen-blown	indirect
Assumed thermal input	[MW]					
biomass		100	100	50	10,000	10,000
hydrogen				47		
Required RD&D time	[yr]	8	10	8	8	10
Required time for market introduction	[yr]	4	4	4	4	4
Efficiency SNG production	[%]	66.3	67.0	79.1	72.8 / 53.2**	71.2 / 45.5**
Carbon conversion	[%]	93.3	100	80.1	93.3	100
Specific investment costs***	[€/kW$_{th}$]	482	449	616	*	*
SNG production costs	[€/GJ]	8.5	7.8	5.6	*	*
SNG production capacity until 2020	[PJ]	157	128	60	*	*
Maximum annual SNG production capacity	[PJ/yr]	14.9	15.1	5.7	*	*
Costs per tonne CO_2 avoided	[€/tonne]	95	83	115	*	*
Avoided CO_2 emission until 2020	[Mtonne]	8.8	7.2	1.3	*	*
Maximum annual avoided CO_2 emission	[Mtonne/yr]	0.84	0.84	0.12	*	*

* The final evaluation of the technical as well as economic and ecological feasibility of cogeneration of SNG and FT-products will be carried out within the Novem project “High efficiency cogeneration of "green" Substitute Natural Gas (SNG) and "green" transport fuels by the Fischer-Tropsch synthesis”[72].

** Efficiencies SNG production for stand-alone SNG, respectively for FT-SNG co-production options

*** Including oxygen plant

In order to determine the possibility of the candidate processes for SNG production becoming a technical success, a diagram containing development stages, as well as possible problems has been constructed, and presented in figure 5. Each development stage is subjected to uncertainties of certain parameters, for example of the problem of biomass feeding being solved, or of agglomeration becoming problematic, or of a successful upscaling. After the development stages, a period of four years is expected to be necessary for the market introduction.

Table 6 *Production cost and market price of SNG for different cases*

		gasification/methanation		hydrogasification
		oxygen-blown	Indirect	
Assumed thermal input biomass hydrogen	[MW]	 100	 100	 50 47
Production cost	[€/GJ]	8.5	7.8	5.6
Market price	[€/GJ]	8.5 – 8.9	8.5 – 8.9	4.9 – 5.4

Conclusions

1. Although substitute natural gas (SNG) has not yet been incorporated as an alternative fuel in the Dutch and European renewable energy policies, it can play an important role in realisation of the Dutch and European climate and renewable energy targets. The produced SNG can most suitably be used for heat (and power) production in the domestic (households) sector, followed by decentralised CHP application in the industrial (and services) sectors. Besides, when the transport sector (at EU-level) would be so far to apply natural gas as an alternative transportation fuel, it should also be possible to use SNG as a substitute for natural gas within the transportation sector.

2. Anaerobic digestion is a proven technology being applied for small-scale decentralised conversion of “wet” organic residues at their origin. SNG production in this sector should always compete with the well-known combined heat and power application. The produced heat in a prime mover (at the moment mainly gas engines, in the future the more efficient fuel cell technologies) can be used optimally within the digestion process, and for other purposes such as space heating. Within this study, therefore, the production of SNG through anaerobic digestion has been considered as a reference case, and not as a competing route to SNG production by (hydro)gasification processes.

3. The overall energetic efficiency of the FT-SNG co-production options is practically equal to the energetic efficiency of the stand-alone SNG options (see table 6.2). Advantages of a co-production option over a stand-alone SNG option will therefore be completely determined by the economy of the considered options. Higher expected specific investment costs of a combined FT-SNG, compared to a stand-alone SNG, will probably result in higher SNG production costs. This would make a combined FT-SNG option less interesting than a stand-alone SNG option. The final evaluation of the technical as well as economic and ecological feasibility of cogeneration of SNG and FT-products will be carried out within the Novem project “High efficiency cogeneration of "green" Substitute Natural Gas (SNG) and "green" transport fuels by the Fischer-Tropsch synthesis”.

4. The energetic efficiency of SNG production by biomass hydrogasification is higher, and the production costs are lower than in the case of SNG production by biomass gasification/ methanation routes. However, the potential of the SNG production, as well as the avoided fossil-based CO_2 emissions until 2020 are much higher for biomass gasification / methanation routes, and the costs per tonne avoided CO_2 emissions are lower than in the case of SNG production by biomass hydrogasification. This is due to both the limited availability (until 2020), and the origin (fossil-based) of the applied hydrogen. Fossil-based

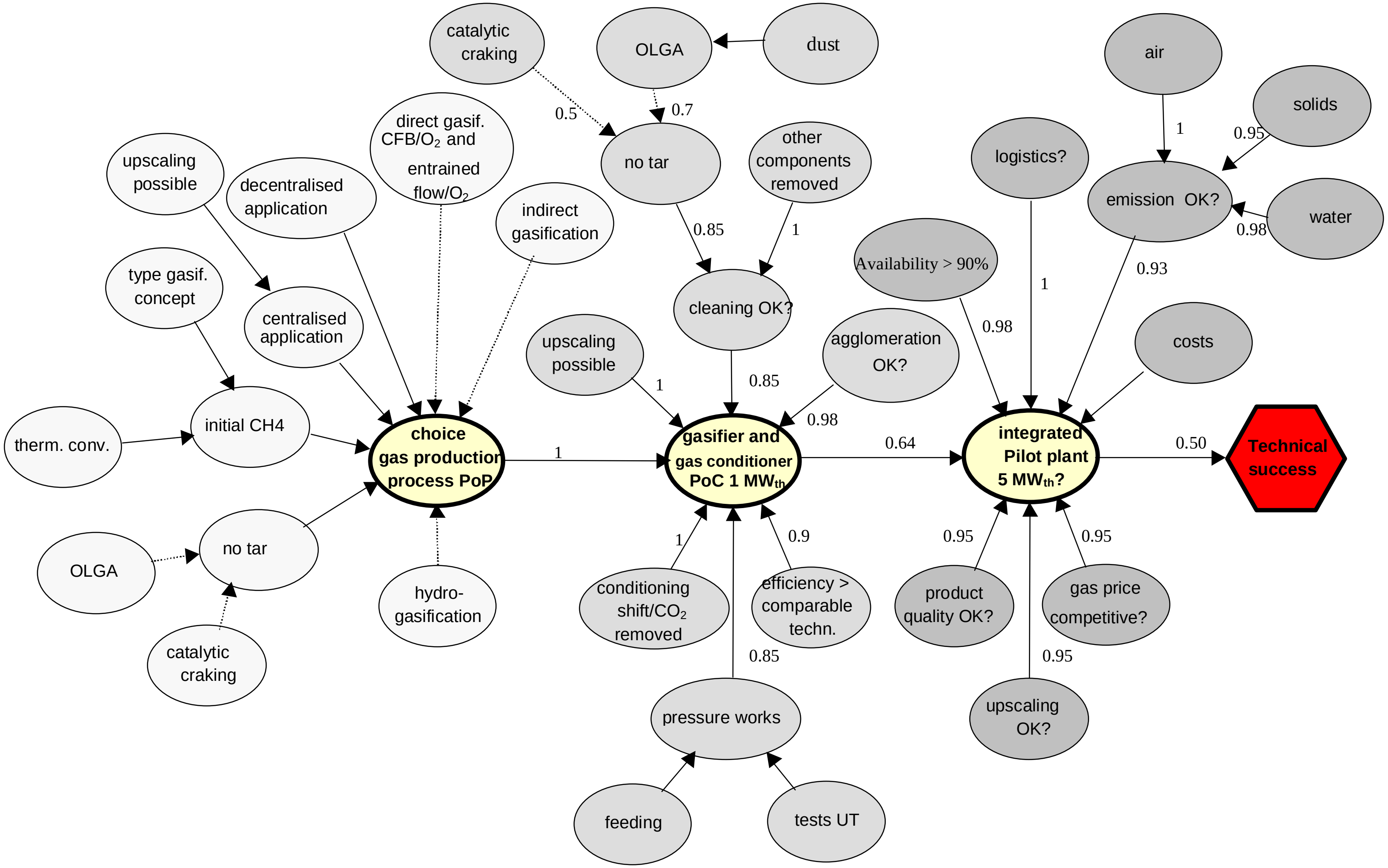

Figure 5 *Technical success diagram of processes for green gas (SNG) production*

hydrogen lowers the market price for SNG from the hydrogasification process (see table 6), as only a part of the produced SNG can be considered green. Based on these results it can be concluded, that production of SNG from hydrogasification of biomass is less interesting than upstream pressurised oxygen-blown or indirect gasification with downstream methanation.

5. The energetic efficiency of SNG production by an indirect gasification process is higher, and the SNG production costs, and the costs per tonne avoided CO_2 emissions are lower than in case of pressurised oxygen-blown CFB gasification. This is, among others, due to lower investment costs and/or higher carbon conversion of an indirect gasification process. On the other hand, the potential of the SNG production as well as the total avoided fossil-based CO_2 emissions until 2020 are higher for pressurised oxygen-blown CFB gasification than in the case of SNG production through an indirect gasification process. The reason for this is the assumption, that indirect gasification would require a somewhat longer development time than the pressurised oxygen-blown CFB gasification. In other words after development, indirect gasification will score better with respect to all aspects, compared to pressurised oxygen-blown CFB gasification, and therefore it is more promising for SNG production.

6. The RD&D trajectory for candidate SNG production processes is expected to take about 10 years for indirect gasification and 8 years for pressurised oxygen-blown gasification, and can be divided into the following three stages (see also figure 5):
    - determination of the Proof of Principle (PoP);
    - determination of the Proof of Concept (PoC);
    - demonstration of a complete integrated process on pilot scale.

    In the PoP-stage (this project) the indirect gasification technology has been selected for further development for green gas production. The preconditions which have been taken into account in this stage are, among others, production of a tar-free (by using the ECN OLGA process), low-nitrogen, and high hydrocarbon content synthesis gas, and the possibility of upscaling of the technology to a commercial scale on the long term.

    In the PoC-stage a bench-scale gasification / gas clean-up installation should be realised at ECN, with which a synthesis gas can be produced that satisfies the specifications for downstream methanation. An extensive R&D programme will be coupled to the realisation of this installation, mainly in the field of analysis of the “optimal” gasification conditions (among others agglomeration behaviour) and gas clean-up (removal of tar and other components). As the first step in realisation of a bench-scale gasification / gas clean-up installation, a lab-scale gasification facility is being constructed at ECN. This new test facility, the Milena (**M**ultipurpose **I**ntegrated **L**ab-unit for **E**xplorative and i**N**ovative **A**chievements in biomass gasification) has a thermal input of about 17 kW, corresponding to a biomass input of about 3.5 kg/h. The installation will be suitable for indirect gasification experiments. Moreover, the facility will also be suitable for agglomeration studies under oxygen-blown gasification conditions.

    In the demonstration stage a complete integrated concept should be realised, that finally can be upscaled to commercial scale on the long term. The R&D aspects that should be analysed in this phase are, among others, the availability of the integrated concept, logistics aspects, ecological aspects (emissions to air and water, quality of solid residues), the quality and price of the product.

Recommendations

1. According to a first estimation[87], the realisation of the PoC-stage of the RD&D trajectory will require a budget of about M€5.5. Main part of this amount should be financed by the industrial partners and different RD&D programmes (DEN/NEO/EU). This requires an intensive acquisition activity. For further technology development also joining international consortiums through EU-projects is recommended.

2. Contacts with developers / producers of indirect gasification technologies (Battelle and Güssing), as well as industries providing downstream methanation (among others Lurgi) and gas conditioning are recommended.

3. Within this study no attention has been paid to developments regarding the super critical biomass gasification (at about 600°C and 300 bar) for SNG production. It seems, however, that high concentrations of methane and other hydrocarbons could be achieved in the product gas resulting from this process. Therefore, it is recommended to study the potential of super critical gasification for production of SNG from biomass; especially from “wet” raw organic materials.

1. INTRODUCTION

1.1 Background

Within the Dutch renewable energy policy an important role is foreseen for the application of biomass and waste. 42% (120 PJ_{th}) of the policy targets for 2020 (288 PJ_{th} = 10% renewables) has to be realised by substituting fossil-based energy carriers with biomass (75 PJ_{th}) and waste (45 PJ_{th}).

In the short term (< 5 years) biomass will mainly be used for direct and indirect cofiring in conventional power plants. In the mid term (5-10 years) it is expected, that the technology for stand-alone decentralised CHP would be mature enough to become an important option. However, in order to satisfy the long-term (> 10 years) expectations of using biomass in the future energy infrastructure, this renewable energy source should also be used as a substitute for fossil fuels in other market sectors. To realise this, conversion technologies should be developed to produce secondary energy carriers from biomass (such as Fischer-Tropsch diesel, methanol, bio-ethanol, SNG, or H_2), substituting the present conventional energy carriers based on petroleum and natural gas. Such a substitution will be potentially capable of saving a significant amount of fossil-related energy, with the resulted ecological advantages.

Concerning the SNG production routes from biomass, the produced “green gas” can be transported through the existing natural gas infrastructure to places where it will be used for the desired energy functions. The energy distribution companies have in this way a strong alternative in addition to “green” electricity at their disposal, in order to achieve their renewable energy targets.

Compared to large-scale (centralised) applications of biomass, SNG production will prevent eventual market problems for products (mainly heat), while thanks to the existing gas infrastructure a large market and certainty in delivery of SNG are guaranteed. On the other hand, using biomass for decentralised CHP application is not always possible, due to the lack of market for the produced heat. The facility aspect of SNG transport through the existing natural gas infrastructure can play an essential role in the successful market introduction of biomass and waste within the national energy infrastructure on mid and long term.

For transport of hydrogen either a new infrastructure would be necessary, or it might in long term be added to the natural gas infrastructure up to some extent. According to Okken[1], for instance, mixing of 10% hydrogen is simply possible within the limits for the Wobbe-index[1] of natural gas. With respect to SNG, however, as the quality (calorific value, Wobbe-index) of the gas can be matched to the quality of natural gas, there will be no limitation in mixing ratio of these two gases.

For the Netherlands, the biomass-related SNG production route is an interesting option to import renewable energy. Another option is to trade in (green) certificates, by converting biomass (at its origin) to SNG. This is in accordance with the Kyoto agreements, concerning the recognition of the role of emission trading and other economic flexibility mechanisms.

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[1] The Wobbe-index W is defined as the ratio of the gross calorific value to the square root of the relative density, with HHV the High Heating Value (MJ/Nm³), and ρ_g and ρ_{air} the gas and air density (kg/Nm³)[70].

$$W = \frac{HHV}{\sqrt{\rho_g - \rho_{air}}}$$

1.2 Problem definition

In recent years research on SNG production from biomass / waste in the Netherlands has been focussed on two technologies, viz. combined gasification / methanation (Gastec)[2], and hydrogasification (ECN)[3][4][5][6][7]. However, a technical, economic, and ecological assessment of these processes, based on comparable basic assumptions, had not been performed yet, and is therefore a part of this project. Beside the above mentioned technologies, there are a number of other potential interesting SNG production technologies, which will also be evaluated within this project.

1.3 Objective

The objective of this project is to make a selection for future implementation of the most promising technologies for the production of SNG from biomass and waste.

The following main activities have been defined within this project:

- *Analysis of the role of biomass/waste and the related SNG production routes (on short term, as well as on mid and long term) in the European and the Dutch climate and renewable energy policies.*
- *Market analysis for technologies with which SNG can be produced from biomass and waste.* The analysis will include both commercial available technologies as well as technologies under development. It will also include technologies which are applied exclusively for SNG production as well as technologies with which more secondary energy carriers, including SNG, can be produced (polygeneration). Based on a literature survey, contacts with the suppliers, and indicative system analysis, a summary will be made concerning the status, technical, economic, and ecological aspects of potential applicable technologies.
- *Classification of the SNG production technologies on the basis of the results of market analysis.* For this purpose an evaluation matrix will be made, in which for each production route important data such as conversion efficiency, specific investment costs, operating and maintenance costs, SNG production costs, emissions, etc. will be presented. Based on these data a competition analysis will be carried out, resulting in selection of the most promising technologies for the production of SNG from biomass and waste.
- *Determination of the required RD&D trajectory (total costs and the possibility of becoming a technical success) for the selected SNG production technologies, in order to come to actual implementation.* This activity will be based on the ‘Decision Analysis’ methodology, applied within ECN. The achieved results will then be used to select one or more technologies for further financial support, with the final objective of implementation on the mid term.

1.4 Report outline

In chapter 2 the role of biomass/waste and the related SNG production routes in the European and the Dutch climate and renewable energy policies are described. Different SNG production routes and their technical, economic, and ecological aspects are studied in chapter 3 to chapter 6. Chapter 7 presents the results of the competition analysis of different SNG production routes, and the RD&D trajectories for candidate SNG production technologies. Finally, the conclusions and recommendations are given in chapter 8.

2. THE ROLE OF BIOMASS/WASTE AND THE RELATED SNG PRODUCTION ROUTES IN THE EUROPEAN AND THE DUTCH CLIMATE AND RENEWABLE ENERGY POLICIES

2.1 European climate policy

According to the Kyoto Protocol the European Union, causing 14% of the world CO_2 emissions, has to cut its greenhouse gas emissions to 8% below 1990 levels by 2008-2012, equivalent to a 346 million tonne CO_2 reduction.

In the interest of cost effectiveness it was decided in Kyoto that countries may meet part of their emission reduction target through measures taken outside of their own borders, by means of the so-called flexible instruments (Joint Implementation[2], Clean Development Mechanism[3], and Emission Trading). The EU point of view is that countries should make no more than 50% of their total effort outside their own borders.

The European Commission launched the European Climate Change Programme (ECCP) in June 2000, whose goal is to identify and develop all the necessary elements of an EU strategy to implement the Kyoto Protocol. The ECCP is preparing a range of additional EU-level policies and measures to cut greenhouse gas emissions as well as to manage an emission trading scheme that could start operating within the EU by 2005.

The internal trading system would have the benefits of both limiting the cost of meeting the Kyoto targets as well as giving the EU early experience in emission trading before a global trading scheme gets off the ground in 2008. The EC proposes that the EU trading system should initially focus on CO_2 emissions and involve only a relatively small number of economic sectors and sources that contribute significantly to the emissions.

In October 2001 the EC confirmed a proposal to claim from the large industries and power plants a penalty per tonne surplus CO_2 emission (from 2005: 50 €/tonne, from 2008: 100 €/tonne). The penalties would then be used to pay for emission rights (quota).

2.2 European renewable energy policy

According to the EC's Green paper[9] any consideration of the future of Europe's energy supply, especially options for diversification, has to include two new factors: 1) climate change, and 2) establishment of a progressively integrated energy market. Taking the second factor into consideration, the measures have to be adopted to offset the challenge of climate change at European level.

Table 2.1 shows the contribution of the different energy sources to the EU's current energy demand, and the expected energy demand in 2030 in absence of an active energy policy[9].

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[2] According to the Joint Implementation mechanism the countries with specific commitments within the Kyoto Protocol can carry out joint investment projects in order to reduce their greenhouse gas emissions. The achieved Emission Reduction Units (ERUs) will be subtracted from the emission budget of the country which has received the investment and will be added to the emission budget of the country which has invested in the project. It is also possible to share the emission reductions, resulted from the project, between two countries (credit sharing)[8].

[3] The Clean Development Mechanism (CDM) has different objectives. It is aimed to promote sustainable developments in developing countries and to contribute to the final objective of the Kyoto Protocol. Beside that, the CDM is an instrument for countries with specific commitments within the Kyoto Protocol to realise part of their Kyoto objectives by joint investment projects in developing countries[8].

Table 2.1 *Contribution of different energy sources (%) to the EU's current energy demand, and the expected energy demand in 2030 in absence of an active energy policy*[9]

Energy source	2000	2030
Oil	41	38
Gas	22	29
Coal	16	19
Nuclear	15	6
Renewable	6	8

In terms of energy source, oil consumption on its own accounts for 50% of CO_2 emissions in the EU, natural gas for 22%, and coal for 28%[4]. Electricity generation and steam production are responsible for 37% of CO_2 emissions, transport for 28%, households for 14%, industry for 16%, and the services sector for 5%.

The European Commission's 'White Paper on Renewable Energy'[10] foresees a doubling of current renewable energy utilisation rates, from under 6% in 1995 to 12% by 2010 (based on the total primary energy supply). The current share of 6% of renewable energy sources to Europe's energy consumption includes 2% for hydropower. Expanding hydropower, (which accounts for one-third of renewables at the present time), is almost impossible, as the development of new useable sites meet strong local resistance. Small-scale hydropower is the only sector with any prospect. Therefore, the EC's target to double the share of renewables to 12% in 2010 should almost all be provided by other forms of renewables like bioenergy, wind energy, solar power, and geothermal energy.

According to the EC's strategic targets, biomass should account for about 5650 PJ in 2010 (with 3140 PJ of bioheat). The present contribution of bioenergy to the energy balances of EU countries is not very different from the 1995 data presented in table 2.2. According to the table the present energy contribution in the EU is mainly in the form of bioheat, whereas bioelectricity is usually limited to biomass cofiring in coal plants and waste utilisation schemes. Transportation biofuels (mainly biodiesel and bioethanol) are used only in a few, local situations. The EC's 2010 targets for biomass and bioheat, according to data presented in table 2.2, mean a three times total increase in the contribution of bioenergy, and a doubling of bioheat contribution.

Table 2.2 *Biomass penetration in EU energy markets (1995)*[11]

Bioenergy vector	Contribution	Market penetration
Bioelectricity	64.8 TWh(e)/yr	2.8% of total electricity generation
Bioheat	38 Mtoe[5]/yr	about 6% of total heat utilisation
Liquid biofuels	< 1 Mtoe/yr	< 0.1% of total transportation fuel use
Total bioenergy	44.3 Mtoe/yr	3.3% of total primary energy demand

The European Commission's 'White Paper on Renewable Energy'[10] expects a great contribution for combined heat and power using biomass for realisation of the EC's 2010 renewable energy targets. Such decentralised biopower installations could range in scale from a few hundred kW to multi-MWs.

Although transport accounts for only 28% of total current CO_2 emissions, 90% of the expected increase in CO_2 emissions between 1990 and 2010 will be attributable to the transport sector, unless radical changes are made rapidly. According to the last estimates, if nothing is undertaken to reverse the growth trend, CO_2 emissions due to transport would increase by approximately

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[4] The specific CO_2 emissions for coal, oil, and natural gas are respectively 0.094, 0.073, and 0.056 Mtonne/PJ.

[5] Million tonne oil equivalent (41.852 PJ)

50% between 1990 and 2010, reaching 1113 Mtonne of emissions, compared to 739 Mtonne in 1990. Road transport is the principal cause as it alone accounts for 84% of CO_2 emissions ascribed to transport.

Taking these facts into account the Commission's Green Paper[9] introduces the objective of 20% substitution of diesel or gasoline by alternative fuels in the road transport sector by the year 2020 with the dual purpose of improving security of supply and reducing greenhouse gas emissions. Only on a sufficiently large level (hence implementation on an EU-wide level), it is realistic to imagine the introduction of alternative fuels with significant market shares.

Out of numerous possible alternative fuels and engine technologies the following three options would appear to have high volume potential (each more than 5% of the total transportation fuel consumption) over the next 20 years[12]:
- biofuels;
- natural gas;
- hydrogen/fuel cells.

Biofuels
Ever since the first oil crisis in 1973, biomass has been considered, and in some cases promoted, as an alternative to fossil fuel as energy source, particularly considering alternatives for motor vehicle fuels (diesel or gasoline). Biological materials can be used as fuel for road transport in several ways:
- plant oils (colza, soybean, sunflower, etc.) by conversion into a diesel substitute;
- sugar beets, cereals and other crops can be fermented to produce alcohol (conventional bio-ethanol);
- organic waste material: oil (cooking oil), vegetable, and animal waste into biodiesel, animal manure and organic household waste into biogas, and plant waste products into bio-ethanol;
- biomass to other liquid and gaseous biofuels by thermochemical (Fischer Tropsch diesel, biomethanol, biodimethylether, bio-oils, SNG and hydrogen) or biochemical (cellulosic bioethanol, ETBE) conversion.

In principle biofuels offer an ideal alternative, when based on EU grown crops, as they are (almost) CO_2 neutral since their carbon content is captured from the atmosphere.

Application of biofuels as motor fuels in the short to mid term has the advantage, compared to natural gas and hydrogen, that they can be used in the existing vehicles and distribution systems, and, thus, do not require expensive infrastructure investment. Present consumption of biofuels (mainly biodiesel, conventional bioethanol) is still below 0.5% of overall diesel and gasoline consumption.

Natural gas
Natural gas consisting primarily of methane (CH_4) can be used as a motor fuel in a conventional gasoline engine. However, it requires special storage and injection equipment. Natural gas as a motor vehicle fuel will have to be kept either under high pressure of 200 bar (compressed natural gas or CNG) or in liquefied form (LNG) at -162°C, in order to allow vehicles to carry fuel for a sufficient range ($\geq$ 400 km) between refuelling. The energy used for compressing the natural gas to 200 bar represents a loss of about 4% of the natural gas energy input. Natural gas offers potential for a 20-25% lower CO_2-emission than the energy equivalent amount of gasoline. However, there is no significant CO_2 advantage over the more efficient diesel engine. Methane is a powerful greenhouse gas, therefore extended use of natural gas must include measures to minimise losses.

Concerning the alternative fuels, an "optimistic development scenario" at this stage might look like the one, presented in table 2.3. As regards the data for biofuel, it should be pointed out that the 2% in 2005 results from the assumption that the current situation in the Member States that are most advanced in this field can be extrapolated to the other Member States. The 6% in 2010 presumes an active policy in promoting biofuels and is based on the available potential in agriculture and waste treatment. The Commission proposes that an increasing proportion of all diesel and gasoline sold in the Member States has to be biofuel, announcing for a second phase an obligation of a certain percentage of biofuels to be blended into all gasoline and diesel. The second proposal creates a European-wide framework allowing Member States to apply differentiated tax rates in favour of biofuels.

Table 2.3 Concept EC's active policy (optimistic development scenario) targets concerning future use of alternative fuels (based on % of total fuel consumption for transportation)[12]

Year	Biofuels	Natural gas	Hydrogen	Total
2005	2	-	-	2
2006	2.8	-	-	2.8
2007	3.5	-	-	3.5
2008	4.3	-	-	4.3
2009	5	-	-	5
2010	6	2	-	8
2015	7	5	2	14
2020	8	10	5	23

For the application of natural gas a new distribution infrastructure must be established and a change of vehicles will be necessary. As it is unlikely that existing vehicles will be adapted at a large scale, this means that the gradual introduction of this alternative fuel depends on the sale of new adapted vehicles. Therefore 2% in 2010 and 5% in 2015 seems an optimistic scenario based on active policy[6].

Whereas other alternative fuels can be applied on the basis of one or several of either existing vehicles (biofuels), available fuels (natural gas), or available distribution infrastructure (biofuels and partly natural gas), hydrogen/fuel cell technology requires everything to be developed. An additional issue is the production capacity, which makes it unlikely that a substantial market penetration will take place before 2015. Fuel cells fuelled by hydrogen are the most complicated alternative, requiring alternative engine technology, as well as large investment in plants to produce the hydrogen and a totally new distribution system. Shifting to a hydrogen-based transport system is a major decision, which will only make sense as part of a large-scale, long-term strategy, in principle extending even beyond the EU.

2.3 Role of biomass/waste-related SNG production routes in the European climate and renewable energy policies

As mentioned in the previous paragraph, the present contribution of bioenergy to the energy balances of the EU countries is mainly in the form of bioheat. Bioelectricity is usually limited to biomass cofiring in coal plants and waste utilisation schemes. Transportation biofuels are used only in a few, local situations.

Within the strategic renewable energy targets of the EC for 2010 an important contribution has been considered for biomass (135 Mtoe/year). The commission has also introduced the objective of 20% substitution of diesel or gasoline by alternative fuels in the road transport sector by the year 2020. From the latter target 8% of substitution is expected to be realised in 2010 (6% by (liquid) biofuels and 2% by natural gas). When this sector would be so far to apply natural gas as

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[6] It should be mentioned that even now compressed or liquefied natural gas is used as fuel in a large number of cars, busses, trucks, boats, and trains in different European countries (such as Italy, Portugal, France, Sweden, Germany, etc.)[20].

an alternative transportation fuel, it should also be possible to use SNG as a substitute for natural gas within this sector.

According to the EC's active policy, the introduction of SNG in the transport sector can be realised in 2010 as a substitute to natural gas. Up to 2% of total fuel consumption in the transport sector can be substituted by SNG, resulting in a much higher reduction in CO_2 emissions, compared to natural gas that replaces gasoline. After 2010 there would be a much higher potential for application of SNG as a fuel in the transport sector.

Other sectors in which biomass/waste-related SNG production routes can play an important role are the households for heat (and power) production, followed by decentralised CHP application in the industrial (and services) sectors. Introduction of substitute natural gas within these sectors has the major advantage, compared to the transport sector, that there is no need for a new distribution infrastructure.

2.4 Dutch climate policy

The agreements reached in Kyoto and subsequently in the European Union, resulted in a target of 6% emission reduction of greenhouse gases, relative to the 1990 emission level, for the Netherlands. This reduction amounting to 50 Mtonne CO_2-equivalent must be achieved in the period 2008-2012. The way the Netherlands will realise this target has been described in two Dutch government's memorials[8][13].

Based on the EU point of view, the Netherlands should achieve 50% of its emission reduction, i.e. 25 Mtonne, through domestic measures. The measures selected have been divided into three packages. The basic package contains measures which can be taken now and which offer a reasonable degree of certainty. These measures should be good for a total reduction of 25 Mtonne CO_2-equivalent per year.

A reserve package was also defined in addition to the basic package. The reserve package contains measures that will be prepared and that can be taken, if necessary, during the run-up to the 2008-2012 period. Policies may be less successful than is currently assumed, or external circumstances may turn out less favourably than expected. The reserve package gives an added edge of certainty that the commitment to reduce emissions will actually be met.

Finally, a third package of measures has been adopted, containing initiatives intended to lead to innovation. It is expected that further reductions (60-80%) of greenhouse gas emissions will be needed after 2008-2012, in order to reduce the negative consequences of a climate change. The innovation package contains steps that the Netherlands will have to prepare for that situation. This package is aimed chiefly at developing new technology and new policy instruments that the government can use to achieve the necessary emission reductions. Technological innovation involves the development of new, climate neutral energy carriers[7], alongside the more traditional renewable energy sources.

2.5 Dutch renewable energy policy

The Dutch government aims to increase the current contribution of renewable energy sources to the total primary energy supply from a share of 1.2% of total energy use in 1999 to 10% by the year 2020. Table 2.4 presents the potential contribution of renewable energy sources to the total primary energy use in the Netherlands. Biomass and waste, with a desired contribution of 120/288 = 42% are the most important renewable energy sources for realisation of the long-term (2020) renewable energy targets.

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[7] Climate neutral energy carriers are ones which emit little or no greenhouse gases during their entire life cycle.

At the time of formulating the renewable energy targets it was expected, that the total contribution of fossil fuels to energy supply in 2020 would amount to 2880 PJ_{th}[14][15]. However, the energy intensity of the Dutch society increases more than it was assumed at that time[17]. In order to achieve the renewable energy targets for 2020, it is therefore necessary to use 170 PJ_{th} of biomass and waste in the energy supply instead of 120 PJ_{th}. Assuming that this additional contribution would completely be managed by using biomass, the required biomass in 2020 would amount to 125 PJ_{th}. On the other hand due to a new definition for the concept "renewable", a "gap" of 55 PJ_{th} has been formed in the renewable energy targets[8]. Assuming that also this gap would be covered by using additional biomass, the total required biomass in 2020 would amount to about 180 PJ_{th}. Including waste the total contribution would therefore amount to about 200 PJ_{th}.

Table 2.4 *Potential contribution of renewable energy sources to total energy use in the Netherlands [PJ_{th} avoided fossil fuel use]*[14][15]

Source	status 1999	target 2000	target 2007	target 2020
Wind	5.3	16	33	45
Sun (PV)	0.1	1	2	10
Sun (thermal)	0.4	2	5	10
Waste	12.1	30	40	45
Biomass	16.0	24	45	75
Heat pumps	0.2	7	50	65
Water (incl. import)	0.7	0	21 (18)	21 (18)
Thermal storage	0.5	3	8	15
Geothermal	0.0	0	0	2
Total	35.3	83	204	288
Rel. contribution	1.2% [16]			10%

The potential contribution of conversion technologies for biomass and waste to meet the renewable energy targets is presented in table 2.5. The table shows, that for the time being the Dutch renewable energy policy is exclusively directed towards using biomass in the power production sector. Policy concerning the application of biomass in other sectors of the Dutch energy infrastructure is missing as yet. It is however expected, that also for these sectors policy targets for application of biomass will be presented in short term.

Table 2.5 *Potential contribution of conversion technologies for biomass and waste to renewable energy targets [PJ_{th} avoided fossil fuel use]*[18]

Technology	status 1999	target 2000	target 2007	target 2020
Waste combustion	12.1	30	40	45
Biomass				
Domestic wood combustion	10.6	8	8	8
Industrial combustion		5	5	5
Power plant cofiring		3	18	20
Decentralised CHP		2	6	30
Landfilling/digestion	5.4	6	8	8
Others	0	0	0	4
Total	16.0	24	45	75
Total biomass and waste	28.1	54	85	120
Contribution renewable sources		65%	42%	42%
Contribution energy supply		1.8%	3.0%	4.2%

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[8] Only 50% of the applied waste (22,5 PJ_{th}) is considered as renewable. Also the contribution of industrial heat pumps (33 PJ_{th}) is defined as not renewable.

2.6 Role of biomass/waste-related SNG production routes in the Dutch climate and renewable energy policies

Figure 2.1 gives a general view of the expected short-term, mid-term, and long-term biomass/waste-related technology implementations within the Dutch energy infrastructure[19].

In the short term (< 5 years) the gasification technology will mainly be used for indirect cofiring of biomass and waste in coal-based power plants as well as in natural gas-based boilers. At the same time the integrated gasification, gas clean-up, gas engine technology for decentralised heat and power production will be demonstrated in this period.

In the mid term (5-10 years) the gasification technology will be developed (technological bottlenecks solved, efficiency improved, costs reduced), allowing stand-alone CHP units (BIG/CCs) to be introduced to the energy market. At the same time application of pressurised gasification technology for cofiring of biomass and waste in natural gas combined cycles will be realised, while (hydro)gasification/methanation technology for the production of “green” gas (SNG) from biomass will be introduced to the energy market.

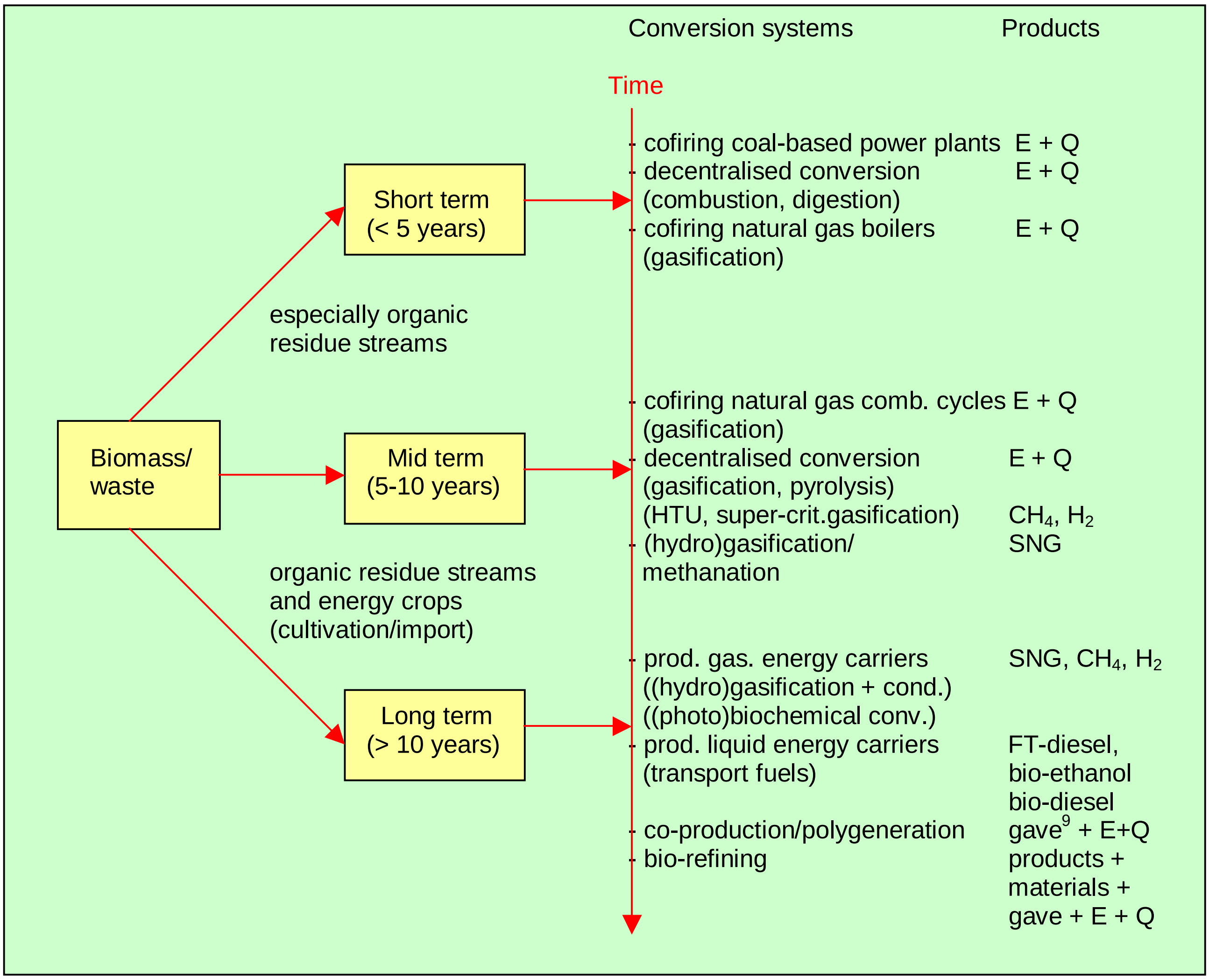

Figure 2.1 *General view of the expected short-, mid-, and long-term biomass/waste-related technology implementations within the Dutch energy infrastructure*

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[9] Dutch abbreviation for gaseous and liquid energy carriers

In the long term (> 10 years) the gasification technology will play a pronounced role for the production of gaseous (H_2, CH_4, SNG, syngas) and liquid (FT-diesel, methanol, etc.) energy carriers from energy crops. The technology will also play an important role in bio-refinery processes in which biomass will be used optimally for the production of a variety of materials, products and secondary energy carriers.

2.7 Discussion

As an important element in achieving the Kyoto targets, the EU respectively the Netherlands aim to increase their contribution of renewable energy sources to 12% (in 2010), respectively to 10% (in 2020) of their total primary energy supply. In both cases biomass and waste are the most important renewable energy sources for realisation of the European and the Dutch renewable energy targets.

In the short term biomass will mainly be used for direct/indirect cofiring in conventional power plants, due to technical and economic reasons. In the mid term it is expected, that the technology for CHP would be mature enough to become an important option, while biomass/waste-related SNG production routes will also be introduced to the energy market.

In order to satisfy the long-term expectations of using biomass in the future energy infrastructure, this renewable energy source should, in addition to application in the electricity production sector, also be used as a substitute for fossil fuels in other market sectors, such as the domestic (households) sector, the industrial (and services) sectors, and the transport sector. To realise this, conversion technologies should be developed to produce secondary energy carriers from biomass (such as FT-diesel, methanol, bio-ethanol, SNG, H_2), substituting the present conventional energy carriers based on petroleum and natural gas.

Although SNG as an alternative fuel has not yet been incorporated in the Dutch and the European renewable energy policies, it can play an important role in realisation of their climate and renewable energy targets.

Due to technical and economic reasons the market introduction of biomass/waste-related SNG production routes can at best take place in mid term. Using the existing natural gas infrastructure is the major advantage for the market introduction of biomass and waste, as SNG, in the national and the European energy infrastructure, compared to other biomass conversion routes.

In this way, the produced SNG can most suitably be used for heat (and power) production in the domestic (households) sector, followed by decentralised CHP application in the industrial (and services) sectors. Besides, when the transport sector (at EU-level) would be so far to apply natural gas as an alternative transportation fuel, it should also be possible to use SNG as a substitute for natural gas within the transport sector.

Substitution of natural gas by SNG, produced from biomass and waste, will lead to a significant fossil-related energy saving and the resulted ecological advantages.

3. SNG PRODUCTION BY UPGRADING BIOGAS FROM ANAEROBIC DIGESTION

Anaerobic digestion is a biological process by which organic wastes, in absence of air (anaerobic), are converted to biogas, i.e. a mixture of methane (40-75 mol.%) and carbon dioxide. A simplified stoichiometry for anaerobic digestion of biomass is:

$$C_6H_{10}O_5 + H_2O \rightarrow 3\ CH_4 + 3\ CO_2 \qquad (3.1)$$

The process is based on the breakdown of the organic macro-molecules of biomass by naturally occurring micro-organisms, through the following four steps:

- *hydrolysis*: polymers such as cellulose, starch, proteins and lipids are hydrolysed to soluble compounds (monomers) such as sugars;
- *fermentation*: soluble compounds are converted to different compounds such as amino acids, fatty acids, alcohols, CO_2, H_2, NH_3, and H_2S;
- *acetogenesis*: fermentation products are then converted to a mixture of hydrogen, low molecular weight acids (primarily acetic acid) and carbon dioxide;
- *methanogenesis*: products of the acetogenesis step are reacted together to produce methane.

The first three steps of anaerobic digestion are relatively rapid, while the methanogenesis step is a slow and more sensitive process. Compared to thermal processes, the residence time of biomass/waste in a digester is relatively long (seconds versus weeks). Optimal condition for methanogene bacteria is essential for a good proceeding of anaerobic conversion. The activity of methanogene bacteria is the highest within two temperature ranges: 30-40°C (mesophillic condition) and 50-70°C (thermophillic condition). Other important process conditions are, among others, a neutral pH (6.5-8), and low concentrations of ammonia and heavy metals.

During anaerobic digestion, typically 30-60% of the input solids is converted to biogas, The co-products consist of an undigested residue (sludge) and various water-soluble substances[11]. The ratio of CH_4/CO_2 is determined by the composition of the feedstock, especially the degree by which the feedstock is reduced or oxidised. Conversion of ethanol (CH_3CH_2OH), for instance, gives a CH_4/CO_2 ratio of 3, while conversion of oxalic acid (COOHCOOH) results in a CH_4/CO_2 ratio of 1/7. In some conversion systems, therefore, a mixture of biomass/waste streams will be applied in order to achieve a higher biogas yield as well as a higher methane fraction.

3.1 Development status

Anaerobic digestion is a well-established technology for waste treatment, and generally available on a commercial basis. Millions of anaerobic digesters (commonly known as biogas plants), have been built around the world, most of which are very small, built in developing countries, while several hundred relatively small digesters have been built on farms in the EU. These have shown varying level of success with both technical faults and poor economics reflecting limited rates of gas production. Greater commercial success has been achieved by high-rate systems installed in sugar, starch or cheese factories, vegetable canneries, and meat processing plant, as well as breweries and distilleries. These provide both primary waste treatment and contribute to process energy needs. Further commercial success has been the development of large ‘joint’ digestion systems in Denmark. Similar plants have been built in Italy, where the technology has also been applied on a large scale. Elsewhere large digesters treating Municipal Solid Waste (MSW) and other residues have been built in France, Germany, and Belgium, with numerous industrial and farm-based digesters dotted around Europe. In the UK use of landfill gas is significant. The technology is fully commercial. Landfill gas is also recovered in Germany, and to a lesser extent elsewhere in Europe[21].

3.2 Digestion systems

Digestion systems can be divided in ‘wet’ and ‘dry’ systems, as presented in figure 3.1. In wet anaerobic conversion systems the process takes place in two separate, and independently controlled reactors. The average residence time is between 10 and 20 days. Acid formation (hydrolyse, fermentation and acetogenesis) occurs in the first reactor, after which the soluble products will be led to the second reactor. This reactor is a water purification reactor, in which the products of the first step will be converted to biogas. The purified water will then be recycled to the first reactor, or it will be discharged to the sewer.

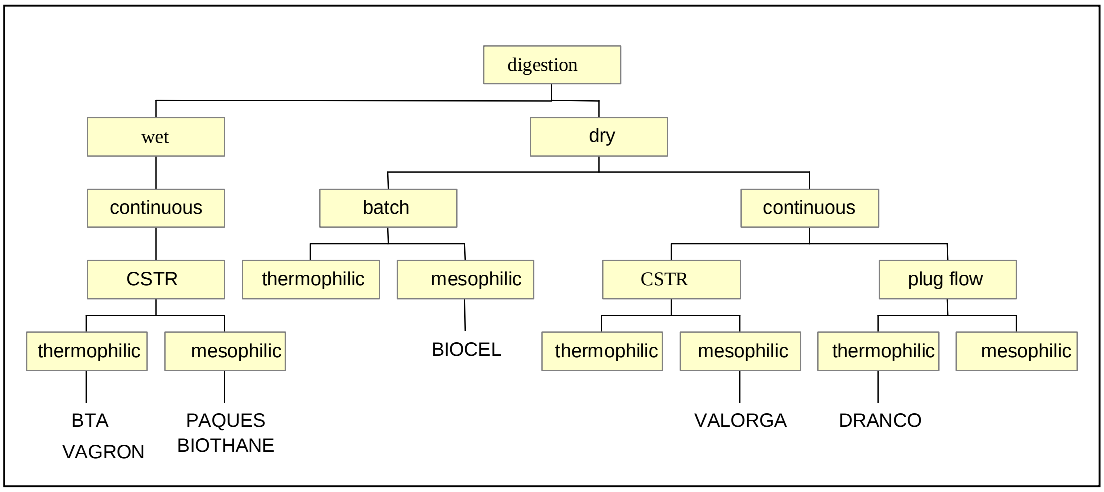

Figure 3.1 *Digestion systems*

In dry anaerobic conversion systems the digestion process takes place in one reactor. These systems can be operated batch-wise or in continuous mode. The average residence time is between 2 and 4 weeks. The dry matter (dm) content in the available systems is between 20 to 25%. In case of a higher dm% of the feedstock, water should be added to the feedstock. After conversion a part of the added water should be removed, in order to achieve the desired dm% for the digestion residue. The removed water can be recycled, or it can be discharged to the sewer, after passing a purification step.

Some examples of the wet digestion systems (BTA, Paques, Biothane), as well as examples of the dry digestion systems (Biocel, Vagron, Valogra, Dranco) are presented in figure 3.1[23][24].

3.3 Biogas applications

Beside the conventional application of the produced biogas for heat production, it can also be used for combined heat and power (CHP) application, or upgraded to natural gas quality.

3.3.1 Combined heat and power application

Biogas, either raw or usually after some enrichment in CH_4, can be used to generate heat and power through prime movers, at capacities up to 10 MW (electric output)[11]. In a gas engine, electric efficiencies of 30-35% (on LHV basis), and total efficiencies of 85% can be achieved (table 3.1). Higher electric efficiencies of up to 50% can be reached by using fuel cell systems instead of gas engines. The amount of heat produced in a fuel cell system, however, may in some cases not be sufficient to cover the total required heat for the digestion process and other purposes, such as space heating. In these cases, therefore, additional fuel should be used to cover the heat requirement.

An important disadvantage of converting biogas in gas engines, compared to application of fuel cell systems, is the high NO_x emission of the gas engines. Beside the high thermal NO_x emissions, also most part of the ammonia present in biogas will be combusted, resulting in extra NO_x emissions. This can in some cases lead to doubling of the NO_x emission[25]. In order to satisfy the emission requirements, additional costly flue gas clean-up (de-NO_x systems) will be necessary. Table 3.2 gives the NO_x emissions of gas engine and SOFC on biogas from manure digestion, compared to average national emissions (on natural gas). It can be seen that SOFC systems have significant lower NO_x emissions, compared to gas engines (even with an additional clean-up step).

Beside the low NO_x emissions of the fuel cells, also the SO_2 emissions of these systems are very low, as due to low fuel cell tolerance to sulphur compounds, these compounds are removed deeply from the biogas, before entering the fuel cell. Other advantages of fuel cells, compared to gas engines, are that they are free of vibration and produce much less noise.

Table 3.1 *Efficiencies gas engines and SOFC systems on biogas*[25]

	Electric efficiency %LHV	Thermal efficiency %LHV
Small gas engines (± 15kW$_e$)	30	55
large gas engines (± 45 kW$_e$)	35	50
SOFC	50	35

Table 3.2 *NO_x emissions of gas engine and SOFC on biogas (from manure digestion), compared with average national emissions (on natural gas) [g NO_x/GJ_{in}]*[25]

	0% NH_3	1% NH_3	Average national emission
Gas engine (without clean-up)	730	1600	280
Gas engine (with clean-up)	70	160	280
SOFC	3	45	
Combined cycles			45

3.3.2 Pipeline quality gas from biogas[22]

In order to obtain pipeline quality gas, the biogas must pass through two major processes:

- A cleaning process, in which trace components harmful to the natural gas grid, appliances or end-users are removed.
- An upgrading process, in which the calorific value, Wobbe-index and other parameters are adjusted in order to meet the pipeline specifications.

Furthermore, the gas must be odourised before it is added to the natural gas grid. If biogas is distributed in a closed biogas network or in a town gas network, only the cleaning of biogas is necessary. This option has been demonstrated in Sweden and Denmark.

3.3.2.1 Cleaning

Gas cleaning includes removal of hydrogen sulfide, water (drying methods: refrigeration; adsorption on the surface of a drying agent such as silica gel, aluminium oxide, or magnesium oxide; absorption with hygroscopic salts e.g. glycol, or triethylene glycol), particles, halogenated hydrocarbons (particularly chloro- and fluoro-compounds; can cause corrosion; can be removed with impregnated activated carbon; regeneration by heating the activated carbon to 200°C), ammonia (removal with activated charcoal units; also in some upgrading processes like adsorption processes and water scrubbing), oxygen (removal during upgrading processes like membrane separation or PSA), and organic silicon compounds (can cause severe damage to gas driven engines; can be removed by absorption in a liquid medium).

Hagen et al.[22] have described the following processes with respect to hydrogen sulphide removal:

- *In-situ reduction (directly in the digester vessel):* either with a metal ion to form an insoluble metal sulphide or oxidised to elementary sulphur, achieving sulphide levels of typically 100 to 150 ppm in the gas stream.
- *Removal with metal oxides / hydroxides:* examples: iron oxide/hydroxide and zinc oxide.
- *Removal by oxidation with air:* Addition of 5 - 10% air to the biogas in a cleaning unit after the biogas production results in biological conversion of H_2S to sulphur (widely utilised in Danish biogas plants); H_2S reductions from 2000 - 3000 ppm to 50 – 100 ppm possible.
- *Removal by adsorption on activated carbon:* H_2S input 50-2000 ppm, H_2S output 10-100 ppm; adsorption at ambient temperature; carbon often impregnated with KI or H_2SO_4 to increase the reaction rate to sulphur.

3.3.2.2 Upgrading

The upgrading process is basically a separation of the methane and carbon dioxide of the biogas, in order to obtain pipeline gas quality with regard to calorific value, Wobbe-index, relative density etc. Possible upgrading processes are[22]:

- *Membrane separation*.
- *Pressure Swing Adsorption (PSA)*.
- *Absorption without chemical reaction:* examples: water wash with regeneration; water wash without regeneration; removal of carbon dioxide using Selexol.
- *Absorption with chemical reaction*.
- *Cryogenic removal of carbon dioxide*.

3.4 European perspective[22][24][86]

Biogas is typically utilised in combined heat and power installations, with excess power fed into the grid. In Denmark biogas is produced at 20 large-scale plants and approximately 20 minor farm plants, with primarily manure (82%) and industrial waste (16%) as feedstock. The large-scale plants produced approximately 50 million m^3 of biogas in 1998, corresponding to an energy production of approximately 1.2 PJ. The produced gas has been utilised in district heating or CHP. The largest[10] CHP plant in Europe, operating on landfill gas, has an electric power output of 8 MW. This plant using biogas from the largest Vienna's landfill at Rautenweg is in operation since 1991.

In Germany biogas is produced at about 30 large-scale plants and approximately 770 small- to medium-scale farm plants. The electricity from the large-scale plants fed into the public grid amounts to over 300 million kWh (1.1 PJ), corresponding to about 100 million m^3 biogas per year. The average installed power in all biogas plants is 60 kW_e and the average digester volume 600 m^3 per biogas plant, providing a total installed power of about 48 MW_e. All digesters produce about 250 million m^3 biogas per year.

The history of biogas technology in Germany started with production of biogas in sewage treatment plants. From the middle of the 1930's to the middle of the 1960's biogas in many large sewage treatment plants was upgraded and used as 'bio-natural gas' as fuel in car engines. Peak production of upgraded biogas for fuel and injection into the natural gas pipeline grid was reached during the Second World War and after 1950. After a break of about 20 years, two sewage treatment plants constructed upgrading plants for feeding the upgraded biogas into the natural gas grid. In the central sewage plant at Stuttgart-Muhlhausen an EU demonstration project was conducted. It was the first upgrading plant in the world using MEA (Monoethanolamine) for absorption of CO_2 and H_2S. The capacity was 500 m^3/h crude biogas. This plant successfully operated from 1984 to the end of 1999. The second plant operated from 1982 to the end of 1998 and was located in the sewage treatment plant of the Niersverband in Viersen. This pilot plant had

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[10] in 1996

a capacity of 450 m^3/h crude biogas. CO_2 was absorbed in water at a pressure of 10 bar. There are currently no examples of introduction of upgraded biogas to the natural gas grid.

Utilisation of the natural gas grid, as a transporting system for biogas, plays an important role in promoting the use of biogas as a vehicle fuel, as it will be possible to produce biogas in any place along the gas grid, with the possibility to trade 100% of the gas as vehicle fuel.

In Switzerland gas as a fuel for vehicles is becoming more and more popular. The gas-fuelled vehicles are mainly cars and not so many trucks and buses. Both natural gas and biogas is utilised as vehicle fuel. Biogas is fed into the natural gas grid at two sites. One is a low-pressure branch of the grid and the biogas is therefore upgraded with PSA technology to meet the natural gas specifications. The other site is at the high-pressure transmission grid and here the biogas is added to the grid without upgrading. This is due to the very small percentage of the biogas added, so that there is no need for upgrading.

Also in Sweden biogas has become very popular as a fuel for vehicles. The reason for this is mainly that the value of biogas is greatly increased if it is upgraded and utilised as a replacement for petrol or diesel oil. During the last 10 years 12 upgrading plants have been started, making vehicle fuel mainly from sewage sludge. The capacity of the plants is between 10-700 m^3/h, and as upgrading process water scrubbing, Selexol scrubbing, or PSA process has been used. Biogas as a vehicle fuel is free from fuel tax and thus competitive to the traditional fuels. The Laholm co-digestion plant in Sweden is producing natural gas quality from biogas since 2000, by upgrading biogas and adding propane to correct the heat value and Wobbe-index. The plant capacity is 250 m^3/h and upgrading of biogas takes place by Selexol scrubbing process.

In France two upgrading plants, based on water scrubbing, with a capacity of 100 respectively 200 m^3/h are in operation since 1994 respectively 1995, making vehicle fuel from sewage sludge, or landfill gas. Another plant with a capacity of 500 m^3/h upgrades landfill gas for injection in natural gas grid. However, the French authorities have not yet authorised injection because of possible contamination.

The Netherlands

In 1998, more than 225 installations, based on anaerobic digestion, were in operation. Also in this year 44 landfill locations produced 1.3 PJ of energy.

There are six commercial waste treatment systems in the Netherlands, as presented in table 3.3, where biogas is produced from digestion of either Vegetable, Fruit, and Garden (VFG) wastes, or the Organic Wet Fraction (OWF) of municipal solid waste. The produced biogas is then, in most cases, used for CHP application. In Tilburg, the produced biogas (41,000,000 Nm^3/year wet biogas with an average methane content of 55%) is delivered to the landfill gas treatment plant, which is in the vicinity of the VFG treatment plant. From there, the landlfill gas and biogas, after gas cleaning / conditioning, are injected into the gasnet at a price of 22 €ct/Nm^3[86].

Table 3.3 *Survey of the commercial waste treatment systems in the Netherlands*

Location	Feedstock	Digestion technology
Drachten	VFG	Biocel
Lelystad	VFG	Biocel
Lelystad	VFG	Biocel
Tilburg	VFG	Valogra
Breda	VFG	Biothane
Groningen	OWF	Vagron

The Vagron plant[24] is the central waste facility of the Province of Groningen, and is the only Dutch facility with a combination of mechanical and biological treatment of municipal solid waste. The digestion section consists of four fermentation tanks, each of which with a volume of 2750 m^3. The temperature in the fermentation tanks is approximately 55ºC, resulting in thermophilic fermentation. The washed OWF remains in the tank for about 18 days, during which about 60-70% of the organic material is converted into biomethane. The produced biogas (about 162000 GJ/a) undergoes combustion in a gas engine used for combined heat and power. In total 2.5 MW of electric energy is produced - enough for 6400 households. Of this, about one-third is used within the facility. The remainder is supplied to the power grid. Also 3.6 MWh of thermal energy is produced, resulting in a total conversion of biomethane into energy (electricity + heat) of 85%.

There are five locations where gas from biomass is added to the gas grid (table 3.4). This addition was quite successful at natural gas prices of about 11 €ct/Nm^3 in the period 1986 - 1990. As a consequence of lowering gas prices, new projects became less attractive. With the introduction of the green electricity concept in 1998, it became more profitable to convert the biomass to green electricity.

Table 3.4 *Survey of the locations in the Netherlands where biogas is added to the grid*

Location	Feedstock	Carbon dioxide removal technology	Output capacity Nm^3/h	Status
Tilburg	landfill gas	water wash	1200	operating since 1986
Nuenen	landfill gas	PSA	3000	operating since 1986
Wijster	landfill gas	PSA	4000	operating since 1986
Vasse	landfill gas	membrane separation	300	operating 1992-1998
Wolvega	landfill gas	membrane separation	1800	operating since 1993

3.5 Conclusion

Anaerobic digestion is a proven technology being applied for small-scale decentralised conversion of “wet” organic residues at their origin. SNG production in this sector should always compete with the well-known combined heat and power application. The produced heat in a prime mover (at the moment mainly gas engines, in the future the more efficient fuel cell technologies) can be used optimally within the digestion process, and for other purposes such as space heating. Within this study, therefore, the production of SNG through anaerobic digestion has been considered as a reference case, and not as a competing route to SNG production by (hydro)gasification processes.

4. SNG PRODUCTION BY COMBINED BIOMASS GASIFICATION /METHANATION PROCESS

Figure 4.1 shows a simplified flowsheet of an SNG production route by a combined biomass gasification / methanation process. Biomass is fed to an O_2-blown or an indirect gasifier. The produced gas after passing a gas clean-up section for removal of contaminants, is led to a methanation step. Upgrading of the product gas from the methanation step results in SNG as the final product.

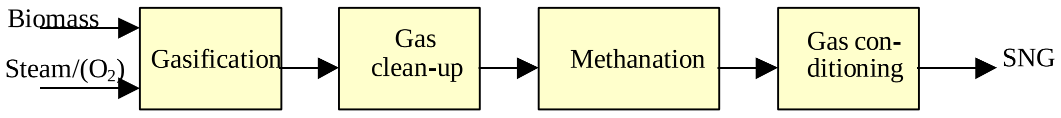

Figure 4.1 *SNG production by a combined biomass gasification / methanation process*

4.1 Gasification

4.1.1 Background

Gasification is a thermochemical process that converts solid fuel (such as biomass) into a gaseous fuel consisting mainly of hydrogen and carbon monoxide. The produced gas can be used as fuel for heat and / or power generation, or as syngas in the chemical industry. It can also be used to produce liquid fuels or hydrogen (after additional reforming and shifting process steps), or be upgraded to substitute natural gas (after additional methanation / gas conditioning process steps) and distributed through the existing gas infrastructure.

While the traditional combustion routes for power generation are suffering from increased investments (due to increasingly stringent emission regulations) and low conversion efficiencies, gasification routes can lead to relatively higher conversion efficiencies at reduced investment costs. Typical efficiencies of power generation in steam turbines, for instance, are about 25%, while power generation via integrated gasification combined cycles (IGCC) for both fossil fuels and biomass offers first-generation efficiencies of greater than 35%, with 45-50% as a reasonable near-term goal. Advanced concepts involving combined cycles with fuel cells as the primary converter could attain efficiencies greater than 60% at relatively modest 5-10 MW_e scales[26].

The suitability of different gasification technologies is, among others, dependent on the supplied fuel, scale of the installation, and the fuel gas application. The most important characteristics of different technologies are the way by which heat is supplied to the gasifier, the operating pressure of the gasifier, and the reactor type.

Heat supply to the gasifier: direct versus indirect
In autothermal or direct gasification the required heat can be delivered by combustion of a part of the feedstock within the gasifier. As gasifying agent air, enriched air, or oxygen can be used. In case of an external heat source, allothermal or indirect gasification, normally steam will be used as gasifying agent.

Operating pressure of the gasifier: atmospheric versus pressurised
Most gasifiers are operated at atmospheric pressure. In some cases also pressurised gasification is used for large systems with fuel gas application in a gas turbine.

Reactor type: fixed-bed reactor versus fluidised bed and entrained flow reactors

In fixed-bed reactors the feedstock, mostly fed at the top of the reactor, moves down slowly as a result of the feedstock conversion in the lower layers. In fluidised bed reactors the crushed feedstock particles are fluidised by a gasifying agent, while in an entrained flow reactor the pulverised feedstock is fed to the reactor pneumatically. Thanks to the high heat and mass transport in both fluidised bed and entrained flow reactors, these reactors have almost no limitation with respect to the scale of the gasifier. The capacity of the gasifier is determined by economic and logistic considerations, the lower limit by the “economy of scale” and the upper limit by availability of the feedstock. This is in contrast with a fixed-bed reactor, which is only suitable for scales up to about 10 MW$_{th}$. As a result of this limitation, fixed-bed reactors are not suitable for large-scale SNG production by biomass gasification, and therefore, will not be discussed any more within this study.

Fluidised bed gasifiers

In a bubbling fluidised bed gasifier the feedstock, together with an inert bed material (mostly sand), is fluidised by a gasifying agent that is fed at the bottom of the gasifier. The velocity of the gasifying agent in a circulated fluidised bed gasifier is so high that the feedstock particles and bed material are circulated in a system consisting of reactor vessel, cyclone, and feedback pipe. Important characteristics of fluidised bed gasifiers are:

- The size of the feedstock should be reduced, mostly to a few centimetres.
- Fluidised bed gasifiers are mostly operated at atmospheric pressure with air as gasifying agent. Pressurised operation will be interesting at large scale in combination with a gas turbine, or for production of liquid fuels, hydrogen, or SNG.
- The minerals from the biofuels can be responsible for major problems. Generally, these problems are associated with the existence and development of low-melting compounds or eutectics, which form sticky layers. In a fluidised bed, this can result in bed agglomeration and defluidisation. This causes local high temperatures, which often accelerates the process. It ultimately can lead to a completely sintered bed content with a glassy phase gluing the bed particles together and shut down of the plant[82]. The operating temperature plays an important role. A higher temperature means, in general, a higher chance for agglomeration.
- The product gas has high methane and C$_{2+}$ fractions.

Entrained flow gasifiers

Important characteristics of entrained flow gasifiers are:

- The feedstock has to be pulverised for pneumatic supply to the gasifier.
- Oxygen is used as gasifying agent instead of air in order to achieve high conversions to gaseous products, in spite of a short residence time of the feedstock in the reactor.
- The gasification takes place at temperatures above the melting point of the ash. The ash, therefore, leaves the gasifier at molten state (slag).
- Due to high operating temperature (> 1300°C) no tar will be formed.
- The product gas has high concentrations of CO and H$_2$, and no methane or C$_{2+}$ fractions. This makes the entrained flow gasification less interesting for SNG production, compared to fluidised bed options, and therefore this technology will not be discussed in more detail within this study.

4.1.2 State of the art[26]

Biomass gasification is at a relatively early stage of development compared to developments using coal and petroleum residues, which are now passing from the demonstration phase to full deployment. Almost all of the gasifiers for biomass power generation in the size range 1 kW$_e$-5 MW$_e$ are fixed-bed units; bubbling or circulating fluidised beds are the gasification reactor systems of choice for the large-scale multi-MW developments.

There are many examples of biomass projects in the RD&D stage, The only units that are commercially deployed are CFB atmospheric, air-blown units being demonstrated as a co-firing technology with coal-fired utility boilers and CHP plants. Examples are the 60 MW$_{th}$ Lahti

Kymijärvi project in Finland (feedstock: paper and textiles, wood and peat), the 10 MW_{th} Zeltweg BioCoComb demonstration project in Austria, and the 83 MW_{th} Amergas BV project in the Netherlands (feedstock: demolition wood) based on the Lurgi low-pressure CFB gasifier.

There are three BIG/CC demonstration plants in Europe:
- The Värnamo project in Sweden;
- The ARBRE project in the United Kingdom;
- The Bioelecttrica SpA Energy Farm project in Italy.

The Värnamo plant is the world's first biomass-fuelled IGCC. The plant is based on Foster Wheeler pressurised CFB gasification technology, Alstom Power Typhoon gas turbine, and the Foster Wheeler heat recovery steam generator, producing 9 MW_{th} (used for district heating) and 6 MW_e. Between 1996 and 1999 the gasifier has been operated for 8500 hours (with different fuels such as wood, RDF, and straw), from which 3600 hours as a fully integrated biomass fuelled BIG/CC. The air-blown pressurised (18 bar) gasifier produced a fuel gas with a heating value (LHV) of 5 MJ/Nm^3. The net efficiency of the gasifier on a cold gas basis was 83%. The extensive operating data have been used to estimate the electricity costs for commercial plants: at scales of 60 MW_e the IGCC is more profitable than CFB combustion, with electricity selling prices of €33/MWh, and biomass fuel costs of €6/MWh, or slightly less than $2/GJ.

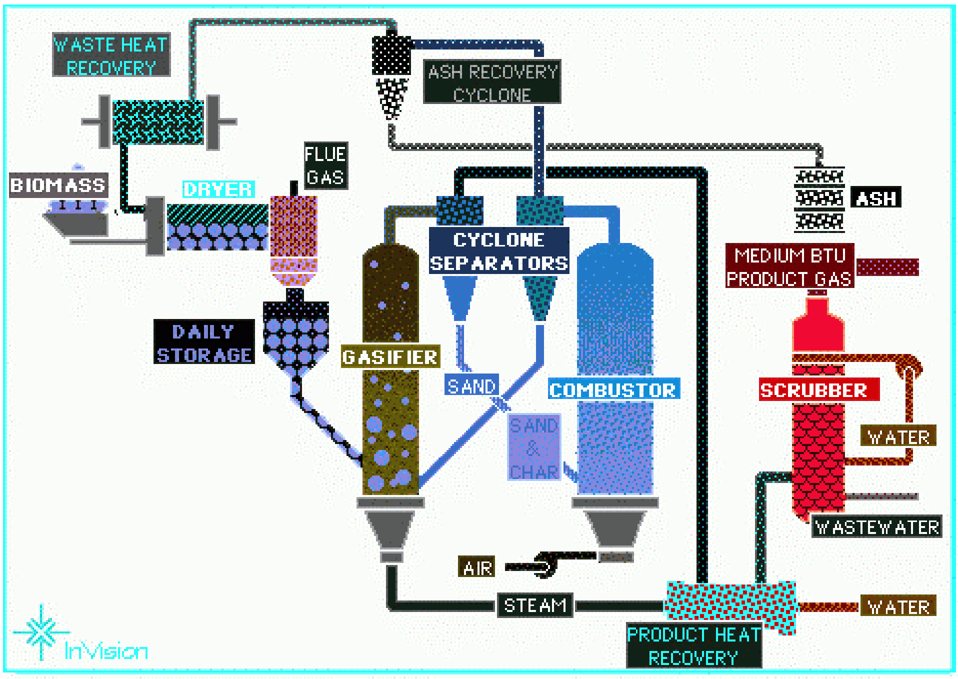

Figure 4.2 *Battelle indirect gasification process*[78]

The overall concept of the ARBRE project is the use of a dedicated willow crop as fuel, and the BIG/CC is based on the TPS atmospheric CFB gasification technology, coupled to an Alstom Power Typhoon gas turbine and steam turbine for a total output of 8 MW_e.

The Thermie Energy Farm project will produce 14 MW_e, from a mix of dedicated energy crops (robinia and poplar) as well as local biomass residues including olive, grape and sawdust from local industries. The design uses a Lurgi CFB gasifier, a Nuovo Pignone (11 MW_e) gas turbine and a conventional HRSG coupled to a steam turbine.

All of the existing BIG/CCs use air as the gasifying agent and require modified turbine combustors to handle the low heating value gas (4-7 MJ/Nm^3). There are now several research, development and demonstration units that use indirect gasification to produce a medium heating value gas in the range of 15-20 MJ/Nm^3. The largest development / demonstration unit is the FERCO unit located at the McNeil 50 MW$_e$ wood fired power station in Burlington, Vermont (figure 4.2). This 40 MW$_{th}$ unit (8.3 tonne/h) has been in operation since mid 1998. The indirect processes take advantage of a unique thermochemical characteristic of biomass in that it will volatise to about 70-85% fuel gases, leaving a char of around 15-20% of the input energy, that is separated and burnt with air to produce heat for the pyrolysis, volatilisation gasification process.

4.2 Methanation

4.2.1 Background

In the methanation step, carbon monoxide and carbon dioxide are converted to methane according to the following reactions[27]:

$$CO + 3\,H_2 \leftrightarrow CH_4 + H_2O \qquad (4.1)$$

$$CO_2 + 4\,H_2 \leftrightarrow CH_4 + 2\,H_2O \qquad (4.2)$$

Also depending on the process condition, either the forward or reverse of the water-gas shift reaction takes place.

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad (4.3)$$

Based on the methanation reactions, as the number of equilibrium stages increases, methane content increases, hydrogen concentration decreases, and the heating value of the gas increases. Also increasing the operating pressure, and decreasing the operating temperature, would lead to higher methane concentrations and higher heating values of the product gas, while the hydrogen concentration would decrease.

Beside the above mentioned reactions, the conditions mainly at the inlet of a methanator are such that thermodynamically carbon could be formed via reactions:

$$2\,CO \leftrightarrow CO_2 + C \qquad (4.4)$$

$$CO + H_2 \leftrightarrow H_2O + C \qquad (4.5)$$

Also cracking reactions of alkenes and aromatic compounds can lead to carbon formation[2].

Since carbon formation and carbon laydown would plug the catalyst bed, for practical operation of methanation it is essential to avoid the undesired carbon forming reactions. These reactions may be avoided by operating the methanation reactors under conditions where carbon is not a stable phase. Equilibrium calculations indicate that carbon (solid) would be a stable phase at low H_2/CO levels[28]. Hydrogen can possibly react with the carbon formed, to produce methane through the carbon hydrogenation reaction:

$$C + 2\,H_2 \leftrightarrow CH_4 \qquad (4.6)$$

Also presence of steam can avoid the undesired carbon formation to take place.

The catalytic methanation of carbon monoxide over nickel is established to proceed via an active surface carbon intermediate formed by the dissociation of carbon monoxide as shown in the following equations[29]:

$$CO\ (g) \rightarrow CO\ (ads) \qquad (4.7)$$

$$CO\ (ads) \rightarrow C\ (ads) + O\ (ads) \qquad (4.8)$$

For the methanation of carbon dioxide, a number of mechanisms have been proposed. These can be summarised in two categories[27]:

- The first category involves the conversion of carbon dioxide to carbon monoxide via the reverse water-gas shift reaction, followed by carbon monoxide methanation.
- The second category involves the direct hydrogenation of carbon dioxide to methane.

A number of investigations have led to the conclusion, that CO_2 is not hydrogenated in the presence of carbon monoxide. Such a conclusion might be erroneous because it does not take into account the formation of water which reacts with CO to make CO_2 by the water-gas shift reaction, a process evolving simultaneous with methane formation. This leads to the observation that the CO_2 concentration instead of decreasing may actually increase or at best maintain steady until the carbon monoxide is substantially consumed. Thus, only at low concentrations of CO, CO_2 reduction to methane might be observed[28].

The methanation of CO and CO_2 is catalysed by metals of Group VIII, by molybdenum (Group VI), and by silver (Group I). Methanation activity varies with the metal as follows: ruthenium > iridium > rhodium > nickel > cobalt > osmium > platinum > iron > molybdenum > palladium > silver. Nickel is a commonly used methanation catalyst. It is relatively cheap, very active, and the most selective to methane of all the metals. Its main drawback, however, is that it is easily poisoned by sulphur, a fault common to all the known active methanation catalysts[28], and hence thorough gas clean-up is required. The basic requirements for a satisfactory nickel methanation catalyst are ease of reducibility, activity, and stability[30].

Methanation catalysts are almost always manufactured and transported in the oxidised form, and therefore they must be reduced in the reactor to nickel metal in order to make them active. The reduction is usually carried out by the process gas and occurs by the two reactions:

$$NiO + H_2 \rightarrow Ni + H_2O \qquad (4.9)$$

$$NiO + CO \rightarrow Ni + CO_2 \qquad (4.10)$$

Since the reactions are not exothermic, the reduction process itself does not cause a large temperature rise in the catalyst bed. However, once some metallic nickel has been formed by reduction with process gas, methanation will start. The exothermic heat of reaction will increase the temperature and accelerate reduction of the catalyst[27].

Methanation activity is related to the surface area of the nickel metal obtained when the catalyst is reduced. The highest surface area of metal and highest activity are obtained when nickel is produced as very small crystallite, usually < 100Å in diameter[30]. Commercial methanation catalysts are mainly in the form of nickel metal dispersed on a support consisting of various oxide mixtures, such as alumina, silica, lime and magnesia, together with compounds such as calcium aluminate cements. The catalysts are made either by a precipitation method or by impregnation of nickel solution onto a preformed support[27]. One of the functions of the other oxides in the catalyst is to support the fine dispersion of nickel crystallites, so that they are available for reaction. The oxides mixed with nickel also slow down the growth of sintering of the metal to form large crystallites with a lower surface area and lower activity[30]. Under normal operating conditions the activity of a methanation catalyst may be diffusion controlled. Consequently, to some extent activity is dependent on total geometric surface area of the catalyst particle, and hence on particle size and shape. Under these conditions smaller particles can display higher apparent activity. However, if the particle size is too small the pressure drop across the bed is too high and the process power requirement excessive. In practice, catalyst particles such as pellets with a diameter of about 5 mm are suitable[27].

The mechanical properties of methanation catalyst (particularly strength) are also important. If strength is insufficient, breakdown of the catalyst will occur and the pressure drop across the reactor will increase. This could give rise to maldistribution of the gas through the bed, giving inefficient use of the catalyst charge, and this is particularly important with methanation where very high conversions are required. Packing densities are normally around 1.0 kg/l. Because of their usual long life in operation, catalyst stability is usually more important than initial activity, which makes the physical strength particularly important.

Methanators are normally vertically mounted with downward gas flow, and in order to ensure a good gas distribution in the catalyst bed a bed height / diameter ratio higher than 1.0 is recommended[27].

4.2.2 State of the art

Recognition during the early 1970s of a future shortfall of natural gas supplies led to the development and introduction, principally in the USA, of oil-based processes for the manufacture of synthetic natural gas. The first of these processes to be commercialised was the British Gas "Catalytic Rich Gas" (CRG) process[31], followed by two alternative processes, one from Lurgi and the other from a Japanese gasoline company. Plants began starting up in 1972-1973 and many more were planned, but the increasing oil price in late 1973 had major adverse effects on process economics, which caused shelving or cancellation of projects that were far from completion[27].

In the British Gas CRG process, which was developed and used in the UK for production of town gas before exploitation of North Sea gas, catalytic gasification of a naphtha or liquefied petroleum gas (LPG) feedstock with steam is used to produce a medium-energy gas which is rich in methane. This is upgraded by methanation to the required heating value of 38 MJ/m^3. In 1977 there were 68 CRG plants in operation, 53 with a total output of 28 million Nm^3/day for production of town gas (heating value: 24.6 MJ/m^3), and 15 with a total output of 35 million Nm^3/day for SNG production. In 1984 the process was still widely used for town gas production in Brazil, France, Greece, Italy, Japan, South Korea and Spain[27].

The oil-based processes for SNG production were never likely to provide a long-term solution to the forecast shortfall of natural gas supplies. It was recognised that this must ultimately be provided by coal and during 1974 and 1975 plans were made for the construction of huge (7 million $Nm^3 day^{-1}$) coal-based SNG plants. Because of the economic changes, only one of the projects reached mechanical completion. This was the project, based on the Lurgi coal gasification process, at Beulah, North Dakota, which first produced SNG commercially in 1984[32][33]. However, the role of this plant was that of a demonstration unit rather than of a major gas supply venture.

4.2.3 Methanation processes

The methanation reactions of both carbon monoxide and carbon dioxide are highly exothermic. Such high heat releases strongly affect the process design of the methanation plant since it is necessary to prevent excessively high temperatures in order to avoid catalyst deactivation and carbon laydown. Several approaches have been proposed[28], from which the following three will be described:

- the equilibrium-limited reactor;
- the throughwall-cooled reactor;
- the steam-moderated reactor.

4.2.3.1 The equilibrium-limited reactor

This concept recycles sufficient product gas to provide added mass to help absorb the heat liberated by the reaction, thereby limiting the temperature rise in the reactor to a safe level. These reactors operate adiabatically, and the temperature rise is controlled by the gas recycle. Most practical, commercial methanation plants have been on the basis of this system. Fresh make-up gas is mixed with the total recycle and fed to the first bed. The effluent from the first bed is partially cooled so that when it is combined with another portion of cold make-up, the mixture is at or above the initiating reaction temperature. The mixture is fed to the second bed. This procedure is continued to the third and subsequent beds[28].

Such a reactor concept has been applied in the Lurgi coal-to-SNG process, as shown in figure 4.3. This process has the attraction of using only moderate temperatures for which commercially available catalysts are satisfactory. The maximum temperature reached is 450°C, and the duty is suitable for “CRG type” catalysts, which is a precipitated nickel catalyst in the form of pellets, containing about 75% nickel oxide before reduction[27].

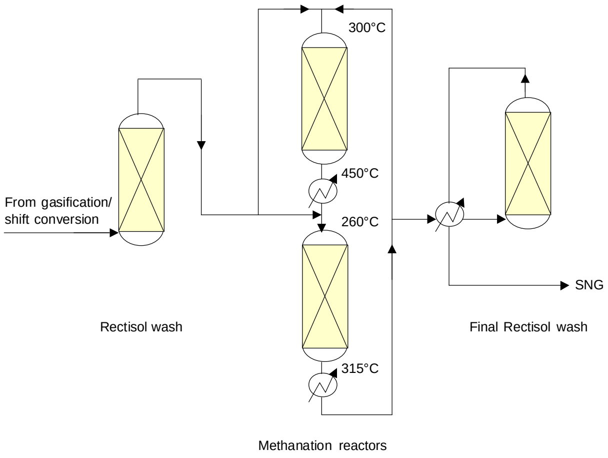

Figure 4.3 *Simplified Lurgi coal-to-SNG process, featuring recycle of product gas to moderate the temperature rise in the first methanator[27]*

Another process applying this methanation concept is the IGT-HYGAS process for converting coals of any rank or sulphur content to SNG[34], which has been under development since the mid-1950s. Coal is first pulverised, dried to a preferred particle size and pre-treated if necessary. It is then passed into the main reactor vessel where it is contacted in a fluidised bed with synthesis gases of high hydrogen content. This hydrogasification reactor operates at pressures of 70-100 bar and at temperatures of 750-1000ºC in order to obtain the proper reaction rates and yields of methane required for process optimisation. The volatile matter and much of the fixed carbon content of the coal (together about 50% of the total feed carbon) is converted to gases in the hydrogasifier. The residual char from the HYGAS process is used to produce hydrogen-rich synthesis gas by the steam-oxygen process.

Raw product gas from the hydrogasifier contains carbon oxides, unreacted hydrogen, methane, and sulphur compounds (principally hydrogen sulphide) produced by direct hydrogasification of the sulphur in the feed coals. The product gas must be purified before it can be upgraded to pipeline quality. Gas purification includes removal of carbon dioxide, liquid aromatics, sulphur, and ammonia. Purified product gas from the hydrogasifier typically contains 17% carbon monoxide, 53% hydrogen, 30% methane, and traces of ethane.

A diagram of the IGT cold-gas recycle methanation process is presented in figure 4.4. The system includes four reactor stages in series. Fresh feed with the proper hydrogen / carbon monoxide ratio (i.e., slightly above the 3:1 minimum stoichiometric ratio) is mixed with recycle gas and sent through a heat exchanger and into the first catalyst bed (stage I). The space velocity is controlled so that all of the carbon monoxide is completely converted. The inlet gases are preheated to about 290°C. The carbon monoxide concentration in the total feed is controlled so that the outlet temperature is limited to a maximum of 480°C. Since the maximum reactor temperature will have been reached, when all of the carbon monoxide is reacted, the temperature can be regulated, by controlling the carbon monoxide concentration in the fresh feed gases, to any stage of this multistage reactor system.

All of the reaction products from stage I are mixed at 482°C with fresh feed and recycle gas in the proper proportions so that the temperature of the total gas mixture is reduced to about 288°C, and the carbon monoxide concentration in the feed gas to the second-stage reactor is limited to about 4-4.5 mol.%. The same ratio of fresh feed to recycle gas is used in all subsequent stages of the system.

The HYGAS process produces approximately 65% of the total methane by hydrogenation and about 35% by clean-up methanation.

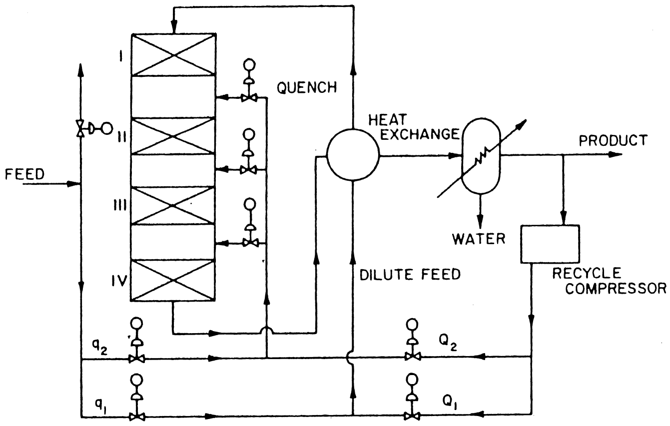

Figure 4.4 *IGT cold-gas recycle methanation system*[34]

4.2.3.2 The throughwall-cooled reactor[28]

Removing the heat of reaction through the walls of a bundle of catalyst-filled tubes controls the temperature rise within the bed. Boiling water in the shell absorbs the heat, and controlling the amount of recycle gas blended with the make-up feed controls the reaction temperature. Scale-up of this type of design is relatively simple since it is necessary to pilot only one of the tubes and to ensure uniform gas flow through the tubes by proper mechanical design. Hence, one can be reasonably sure of performance on the basis of piloting just one tube. The main advantage of this design over the adiabatic systems (e.g. product gas recycle) is that potentially it can achieve complete methanation in one reactor rather than in a series of reactors.

However, there is a greater possibility of excessively high, local temperatures of the catalyst surface with this design than with any other. This would lead to more rapid catalyst deterioration and higher catalyst consumption. Local hot spots can develop at the point of reaction. Prediction of these local hot spots is not easy. The cooling of the hot spots after completion of the reaction is so slow, that the high temperature lasts sufficiently long for sintering of catalyst crystallites to occur, with resulting decrease in catalyst activity.

4.2.3.3 The steam-moderated reactor

This concept involves the combination of gas shift and methanation with steam used in a multistage reactor system to control the equilibrium composition of the exit gas. The temperature rise is thereby controlled, and the need for a large recycle gas compressor, such as that in the equilibrium-limited reactor, is eliminated[28]. This concept makes it possible to achieve a maximum recovery of energy in the form of high-pressure steam at a minimum cost, while using the maximum temperature differential for the generation of steam[35]. In summary, the basic advantages of this process are[28]:

- elimination of a mechanical device (recycle gas compressor) for controlling the adiabatic temperature rise;
- combination of CO shift with methanation;
- significant increase in by-product steam recovery;
- significant capital advantages.

An example of the steam-moderated reactor concept is the ICI high-temperature once-through methanation process. This process is developed in co-operation between ICI and Krupp-Koppers, for the production of SNG based on the Koppers-Totzek coal gasification[11], and it allows direct handling of gases high in CO content. Desulphurised syngas mixed with steam is directed over methanation catalysts contained in a series of adiabatic fixed-bed reactors, operating at successively lower exit temperature[36]. Shift conversion occurs simultaneously with methanation, thereby eliminating the need for any pre-treatment to achieve a stoichiometric feedstock mix. Carbon formation is avoided by using steam and operating at high temperatures. Between the reactors, heat is removed from the system by the generation of high-pressure steam in conventional heat-exchange equipment. As the flow progresses through the series of reactors and exchangers and the bulk of the syngas is methanated, the temperature of the process gas is progressively lowered until it finally reaches an adequately reduced level that is favourable for high-efficiency conversion of hydrogen and carbon oxides to methane.

A flowsheet of this process is shown in figure 4.5, with the corresponding gas composition and temperature at each stage given in table 4.1. Conditions for nickel carbonyl[12] formation defined the minimum inlet temperatures to the first two methanators, while the amount of steam added is such that the maximum temperature in the first methanator was below 750°C. The high operating temperatures provide a large temperature difference for the production of high-pressure steam. Because of this, more steam can be produced at a higher pressure with less heat transfer surface than other processes.

ICI developed a new high-nickel catalyst (nickel oxide ≈ 60%) for this process, which appeared to have the necessary activity, stability and physical strength to methanate a variety of process gases at temperatures up to 750°C. However, the large-scale plants have not been built yet[27].

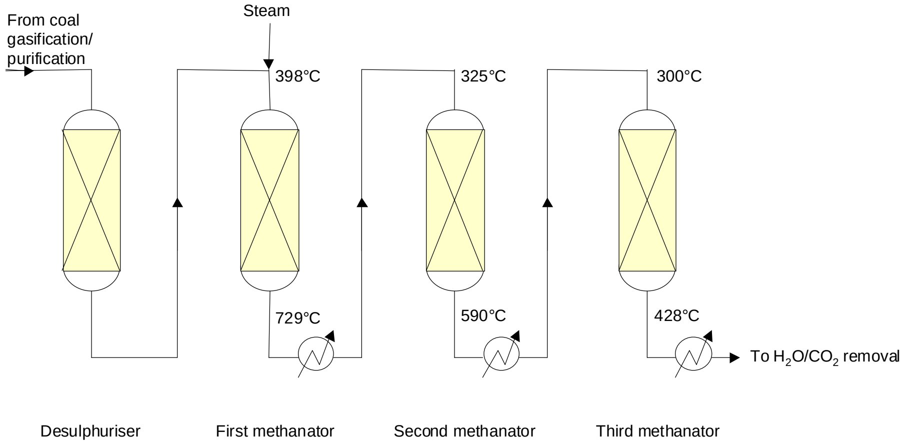

Figure 4.5 *ICI high-temperature once-through methanation process*[27]

Table 4.1 *Gas compositions (vol.%) and temperatures in ICI methanation process*

Composition[13]		1st Methanator		2nd Methanator		3rd Methanator	
		Inlet	Exit	Inlet	Exit	Inlet	Exit
CO	[%]	31.14	14.47	14.47	4.29	4.29	0.34
CO_2	[%]	24.66	40.15	40.15	53.93	53.93	62.70
H_2	[%]	42.91	35.50	35.50	20.26	20.26	5.83
CH_4	[%]	0.08	8.52	8.52	19.84	19.84	29.13
N_2+Ar	[%]	1.21	1.36	1.36	1.68	1.68	2.00
H_2O[14]	[%]	67.3	72.3	72.3	94.4	94.4	118.2
T	[°C]	398	729	325	590	300	428

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[11] The composition of the gasification product depends on the process and the operating conditions used, but in general the dry gas compositions are in the range: CO_2, 0-30%; CO, 10-60%; H_2, 25-75%; CH_4, 0-20%.

[12] Nickel tetracarbonyl, $Ni(CO)_4$, is an extremely toxic, almost odourless gas, stable at low temperatures.

[13] dry gas basis

[14] steam relative to 100 volume of dry gas

4.3 Gas clean-up

The product gas from the gasifier has to be cleaned before entering the methanation section and injection as SNG in the natural gas infrastructure. The gas clean-up requirements are, therefore, imposed by the methanation section as well as by the natural gas infrastructure.

4.3.1 Requirements methanation section

4.3.1.1 Particles

Particles can deposit on the surface of methanation catalysts and make them inactive. According to an IEA report[37] the amount of particles in the synthesis gas should be limited to 0.02 mg/Nm^3.

4.3.1.2 Hydrocarbons/tars

The light hydrocarbons do not seem to affect catalyst activity, and they do reform into methane[35]. Hausberger et al.[38] report, that ethylene, ethane, propylene, and propane undergo reforming reactions over the catalyst but do not poison it. However, the hydrocarbon content can be increased to levels where they do depress the methanation activity. If hydrocarbons are unsaturated enough, they will form carbon when they get to a certain level[35].

Benzene, although not a poison such as H_2S and HCl, does depress activity by reforming and adsorption onto the catalyst. At high levels it can produce carbon. Hausberger et al.[38] have investigated the effect of different benzene levels (0-5%) on the catalyst activity. At low levels benzene had no noticeable effect; it was reformed to methane, carbon oxides, and hydrogen. However, at levels above 5%, catalyst activity declined. At very high benzene levels, carbon formation was observed over the catalyst.

According to Polman et al.[2] the higher hydrocarbons, such as ethane, ethylene, and BTX (including naphthalene), which are still present in the product gas after the gas clean-up step, will not cause any problems to the methanation reactor. They will all be converted to methane and carbon dioxide. However, they may cause problems to guard beds before the methanation step, by polymerisation reactions, shortening the lifetime of the guard beds. This aspect should therefore be taken seriously into consideration, when selecting the guard beds[2].

4.3.1.3 Nitrogen compounds

Polman et al.[2] mention no further limitation for NH_3 en HCN after the conventional clean-up to about 1 ppm.

4.3.1.4 Halogen compounds

Hydrogen chloride is a permanent irreversible poison to the methanation activity of nickel catalyst, even though most of it is not picked up by the catalyst, but is observed in the effluent gas[38]. According to Polman et al.[2] an HCl/HF concentration of less than 25 ppb would be admissible for nickel catalysts. This means, that an additional clean-up step of the product gas from the gasifier, in a guard bed, will be necessary. As the adsorbent has to be stored after saturation, for continuous operation two parallel fixed beds will be applied.

4.3.1.5 Sulphur compounds

Sulphur compounds affect the nickel catalyst through the reaction of hydrogen sulphide with nickel, according to the reaction[28]:

$$\mathrm{Ni} + \mathrm{H_2S} \leftrightarrow \mathrm{NiS} + \mathrm{H_2} \qquad (4.11)$$

Hydrogen sulphide is present in the feed gas, or it can be formed by hydrogenation or hydrolysis of any sulphur-bearing compound (such as R-SH, COS) over the nickel catalyst. Hausberger et al.[38] report that sulphide poisoning occurs at levels as low as 0.3 ppm H_2S. According to Polman et al.[2] an H_2S/COS concentration of less than 0.1 ppm would be admissible for nickel catalysts. In order to achieve such low sulphur concentrations, an additional clean-up step of the product gas from the gasifier in a guard bed (such as activated carbon or zinc oxide) will be necessary. As the adsorbent has to be stored after saturation, for continuous operation two parallel fixed beds will be applied.

4.3.1.6 Metals

Based on the sharp requirements regarding other impurities (than metals), which are considered as catalyst poisons, it can be assumed that the methanation catalysts would tolerate no metals. Besides, it should be mentioned that the guard beds that are used for halogen and sulphur removal will remove any particles or metals still present in the cleaned gas.

4.3.1.7 Carbon formation

Carbon formation refers to the potential production of carbon from different sources through the reactions mentioned in §4.2. The conditions favourable for carbon formation from these sources can be predicted by straightforward thermodynamic calculations. However, because a number of other chemical reactions can occur simultaneously and the relative reaction rates are not well known, it is useful to know whether a specified mixture of syngas and steam would have the thermodynamic potential for carbon formation when it is at chemical equilibrium[39]. To assist in proper visualisation of the multiple chemical reactions that occur simultaneously, a ternary diagram can be used.

Figure 4.6 presents such a ternary diagram with a family of carbon isotherms at 28 bar total pressure. Interpretation of the isotherms reveals that mixtures of the elements, which fall above the curves, are in the carbon-forming region when at chemical equilibrium. Mixtures of elements, which fall below the curves, are outside the carbon-forming region at equilibrium. Gas mixtures falling within the family of curves should be at an operating temperature that will carry the specified concentration of carbon in the vapour phase.

In certain areas of the diagram, elevated temperatures support higher concentrations of carbon in the vapour phase, whereas in other areas lower temperatures favour higher carbon concentrations. Therefore, depending on the element mix of gases feeding a methanation reactor, an increase in temperature could cause the mixture to approach a condition under which carbon could theoretically be formed.

4.3.2 Requirements natural gas infrastructure

As the produced SNG would be transported to end users through the natural gas infrastructure, it should satisfy the requirements that are imposed to natural gas with Groningen quality (G-gas). The specification of the G-gas delivered by Gasunie to energy distribution companies is presented in table 4.2. Also the requirements concerning the SNG, according to a Gastec study[2], are

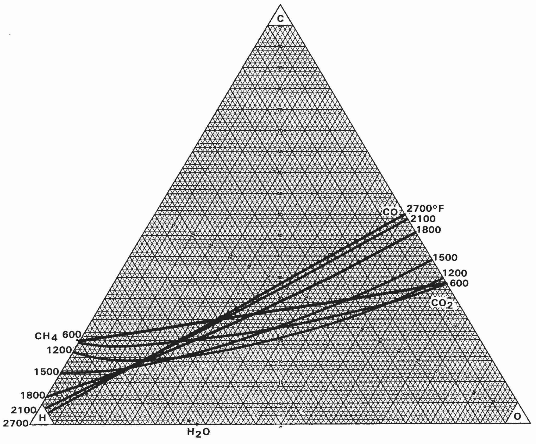

Figure 4.6 *Carbon formation equilibrium isotherms (°F) at 28 bar total pressure*

given in this table[15]. Natural gas with Groningen quality has a Wobbe-index of about 44 MJ/Nm^3. The SNG should also have a composition resulting in a comparable Wobbe-index. There are a number of compounds in SNG that are not present in Groningen natural gas. The assumed requirements for these compounds are, among others, based on the MAC-values and the sensitivity to corrosion. The water content of SNG should be reduced, in order to achieve a dew-point lower than -10ºC (at the gas-delivery pressure). This prevents condensation problems within the gas net, and therefore, corrosion problems[2].

Table 4.2 *Specification of the G-gas delivered by Gasunie to energy distribution companies and the requirements concerning SNG, according to Gastec*[2]

		G-gas	SNG
Wobbe-index	[MJ/Nm^3]	43.46< W <44.41	43.46< W <44.41
H_2	[mol.%]	-	<12
CO	[mol.%]	-	<0.8
O_2	[mol.%]	<0.5 (wet net)	<2.5 (dry net)
C_2H_4	[mol.%]	-	conform Wobbe-index
BTX (incl. naphthalene)	[ppm]	<500	<500
HCl	[ppm]	-	<1
NH_3	[ppm]	-	<1
HCN	[ppm]	-	<10
H_2O		Dew-point < -10ºC	Dew-point < -10ºC
H_2S	[mg/Nm^3]	<5	<2
Mercaptanes	[mg/Nm^3]	<10.0	<5.0
S total	[mg/Nm^3]	<45	<25
Hg	[mg/Nm^3]	0	<0.01
Dust	[mg/Nm^3]	0	<0.5
THT	[mg/Nm^3]	>10 and <30	>10 and <30

Table 4.3 *Typical impurities in product gas from a biomass gasifier and the requirements concerning the methanation step and the produced SNG*[2][7][37][40]

Component		Concentration in product gas from gasifier	Requirement methanation step	Requirement SNG
H_2S	ppm	100	0.1	1
COS	ppm	10	0.1	
HCl	ppb	25000	< 25	< 1000
HF	ppb		< 25	
NH_3	ppm	2830	100	< 1
HCN	ppm	280		< 10
Hg	mg/Nm^3	0.02	0.5	< 0.01
Cd	mg/Nm^3	0.94	0.05	
Na+K	mg/Nm^3	1630	1	
Dust	mg/Nm^3	10,000	10	< 0.5
BTX[16]	ppm			< 500
Tars	mg/Nm^3	10,000-15,000	5	0.1
Heavy metals	mg/Nm^3	< 300	< 1	< 0.01

4.3.3 Candidate gas clean-up processes

Typical main impurities in product gas from a biomass gasifier and the requirements concerning the methanation step and the produced SNG are summarised in table 4.3[2][7][37][40]. Generally, it can be concluded that a high degree of clean-up is required, before the product gas from the

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[15] Gasunie does not support the SNG requirements as proposed by Gastec. With regard to hydrogen a few percent of hydrogen may be allowed. Hydrogen might especially be problematic when gas is combusted in partial premixed burners, e.g. residential gas stoves. The hydrogen might cause flame backstroke, causing a blow-out of the stove, or an internal incomplete combustion of the gas. While the former might result in gas accumulation and possible explosion, the latter might result in CO production. Some lab experiments with such burners have been performed and the results indicate that a small fraction of hydrogen might be acceptable, however a hydrogen concentration of 12%, as mentioned by Gastec, would be too high. Furthermore, the effect of several gas components on the corrosion of the natural gas grid should be investigated in more detail, in order to come to a strict specification of the requirements concerning SNG[83].

[16] In the Gastec study [2] BTX (benzene, toluene and xylene) includes naphthalene as well.

gasifier can be supplied to the methanation reactors. It can also be seen that the methanation step is more sensitive to sulphur and halogen compounds, compared to the requirements with respect to the produced SNG.

In this paragraph different gas clean-up techniques that can be applied within a low-temperature gas clean-up process for removal of different impurities are presented.

4.3.3.1 Particles

In biomass gasification, cyclones are generally applied for the primary removal of particles. The d_{99} (the diameter of the particles, from which 99% is separated) lies at about 10μm for a conventional cyclone. However, the fuel gas from a biomass gasifier has usually a considerable fraction of particles with diameters less than 1μm. Therefore, to achieve high degrees of particle removal, additional measures, like application of filters, scrubbers, or guard beds would be necessary.

Different types of filters are available. Ceramic candle filters are suitable for particle removal from fuel gas at high temperatures and pressures. Filtration at up to 1000°C and 50 bar is possible. Removal efficiencies of 99.9 wt.% (>1μm) might be achieved for this type of filters. As candle filters are available as modules of a certain dimension, for higher gas streams a combination of a number of the modules in parallel is necessary to achieve the required filtration unit. Typical mean pressure drop across a filter is about 200 mbar. Using nitrogen or clean fuel gas can clean the filter.

Other filters for particle removal are bag-house filters and electrostatic precipitators (ESP). A bag-house filter works at temperatures of about 250°C and is among others applied by Lurgi. An electrostatic precipitator is an efficient way to remove particles. Particle removal is realised by giving them a charge, and removing them in an electric field. A low pressure drop and high removal efficiency (90-99%> 0.1μm) are advantages of this technique. Clean-up of the electrodes takes place by knocking at higher temperatures, and by washing at lower temperatures (40-60°C). The latter leads, however, to a very polluted wastewater stream. Operating costs and investments are high.

Also scrubbers can be used for removal of, among others, particles from the fuel gas. Other compounds that can be removed by scrubbers are tar, (alkali) metals, nitrogen, halogen, and sulphur compounds. The fuel gas will be cooled down to 40-60°C either indirectly, by using a heat exchanger, or directly by quenching with the applied liquid. In the first case, the gas after cooling will be led through a scrubber, while in the second case, the gas will not only be cooled, but at the same time will partly be cleaned from the impurities.

4.3.3.2 Tars

Cracking the tars and higher hydrocarbons to CO, CO_2, H_2, and small hydrocarbons can reduce the tar content in the product gas from the gasifier. Thermal cracking of the tars and higher hydrocarbons takes place at temperatures above 1000°C, and during the process a lot of soot will be formed. More interesting is catalytic cracking, using a suitable catalyst such as dolomite or nickel, resulting in a considerable reduction of tars at lower temperatures of about 750-900°C. According to TPS, using dolomite in a tar cracker after the gasifier can lead to tar reductions down to 100 mg/Nm^3.

Filters, scrubbers, activated carbon, and zeolitic filters can also partly remove tars. With respect to filters, condensation of large amounts of tar will lead to filter blinding, which makes the filters less suitable for direct removal of large amounts of tar.

Currently ECN is working on a tar removal process that can lead to total removal of tar compounds. A novel scrubbing system called “OLGA” has been developed for scrubbing at increased temperature levels (above the dew-point of water). The “OLGA” system makes use of a special solvent[84].

4.3.3.3 Nitrogen compounds

In a scrubber, depending on the impurities in the fuel gas, a low PH-value (for removal of NH_3), or a high PH-value (for removal of H_2S and HCl) may be applied in order to improve the removal efficiency. Often combinations of scrubbers with different PH-values are used to achieve the required clean-up degree.

4.3.3.4 Halogen compounds

Most part of the HCl/HF content of the fuel gas will be removed by scrubbing. A guard bed at the end of the gas clean-up section will remove the residual HCl/HF to the desired concentration.

4.3.3.5 Sulphur compounds

Comparable to HCl/HF removal, most part of the H_2S content of the fuel gas will be removed by scrubbing. A guard bed at the end of the gas clean-up section will remove the residual H_2S to the desired concentration.

When biomass residues with a high sulphur content are applied, the resulted fuel gas will contain high concentrations of H_2S. In such cases desulphurisation processes which are applied in oil refineries and coal gasification processes can be used. These processes are either based on physical absorption (e.g. Selexol process), or chemical absorption (e.g. methyl-diethyl-amine, MDEA, process), or a combination of physical and chemical absorption (e.g. Sulfinol process). Also in this case a guard bed at the end of the gas clean-up section will remove the residual H_2S to the desired concentration.

4.4 System modelling

For the SNG production by combined biomass gasification / methanation process the pressurised O_2-blown CFB gasification as well as the indirect gasification concept of Battelle process have been taken into consideration. In order to determine the mass and energy balances of these processes two Aspen Plus models have been developed, as shown in figures 4.7 and 4.8.

In case of the pressurised O_2-blown gasification a CO_2 stream is used to pressurise the biomass to the gasification operating pressure. The produced syngas from the oxygen/steam gasifier, after a low-temperature clean-up and passing through a methanation reactor, is used for the production of SNG as main product. The heat generated at various points in the process is used for steam and electricity generation in a steam cycle, in order to satisfy the demand within the system. The system pressure is 15 bar, from gasification through methanation. Pressure drop has not been modelled.

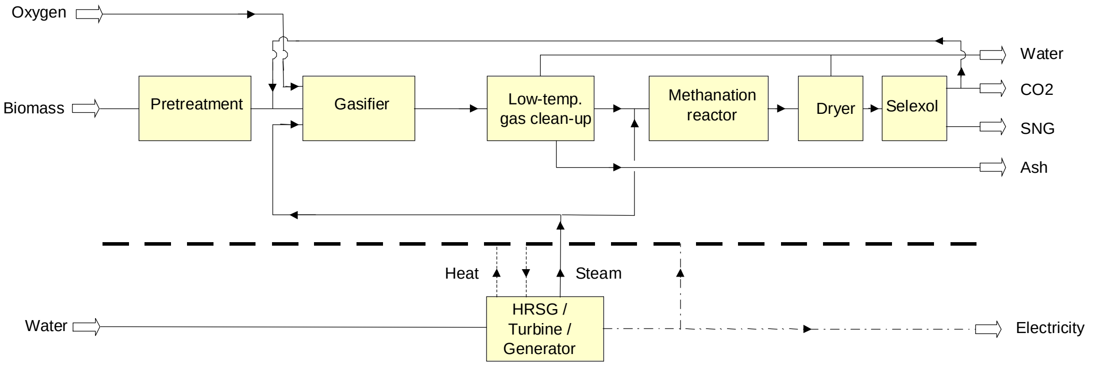

Figure 4.7 *Aspen Plus model for SNG production by a combined pressurised O_2-blown biomass gasification / methanation process*

The indirect gasification process (Battelle) is operated at atmospheric pressure; hence, the product gas from the gasifier has to be compressed to 15 bar. Again, pressure drop has not been modelled. The heat required within the gasifier is provided by the combustion of char in a separate reactor. Below follows a brief description of the different parts of the models.

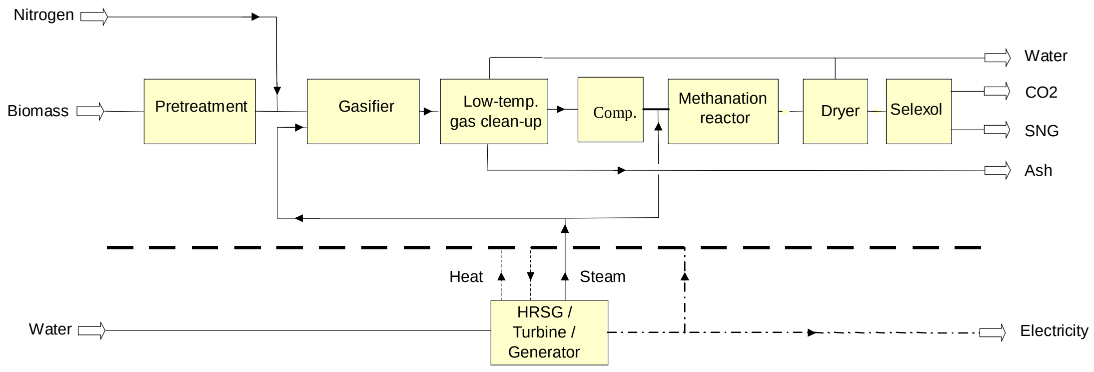

Figure 4.8 *Aspen Plus model for SNG production by a combined indirect biomass gasification (Battelle) / methanation process*

Gasifier

Pre-treated wood (reduced in size) is fed to the gasifier using carbon dioxide or nitrogen as a transport gas. In contrast to the hydrogasification process (chapter 5) the wood entering the gasifier has a moisture content of 15 wt.% instead of 5 wt.%, and therefore no drier is considered within the process. The gasifier model is based on an earlier Aspen Plus model of an air-blown CFB gasifier, which is based on the Excel model "vergasser-12"[74], and includes the within "vergasser-12" mentioned correlations. The composition of the syngas with regards to CO, CO_2, H_2O and H_2 is based on chemical equilibrium. In case of the Battelle gasifier the water-gas shift reaction is modelled with a temperature approach of 1000°C, in order to achieve a comparable composition with the ones mentioned in literature.

Ash

Ash is separated as a single stream from the reactor. The separation method depends on the design of the gasifier.

Syngas cooler

In the radiant syngas cooler the fuel gas is cooled to the inlet temperature of the tar removal process (OLGA). The heat that is released during cooling is used for generation and superheating of 40-bar steam.

Gas clean-up section

The clean-up will consist of a dust filter, a quench, also serving as acidic wash for removing NH_3, a neutral wash for removal of sulphur components and halogens, a special wash for reducing the tar, heavy metals and ammonia content (OLGA), and guard beds in order to protect the methanation catalysts against halogen and sulphur compounds. The gas clean-up section, however, is not modelled in detail.

In case of the Battelle gasifier the cleaned syngas is compressed to 15 bar, before entering the methanation section. In case of pressurised O_2-blown gasification the feed streams of the gasifier are already compressed to 15 bar.

Methanation

The methanation process is based on the intercooled methanation process used within the Lurgi coal-to-SNG process (§ 4.2.3.1). By regulating the amount of syngas fed directly to the second methanator and recycling a part of the product gas to the first methanator the inlet and outlet temperatures are regulated (temperatures remain between 260°C and 450°C) and almost all CO is converted. The reaction is modelled not to be restricted by a temperature approach. The reaction heat is used for producing 40 bar steam and for preheating boiler feed water.

Cooling and drying SNG

The product gas is cooled in two steps to 10°C. The first cooler preheats the boiler feed water. In the second cooler, cooling water is used to bring the gas to 10°C. Finally, an electric cooler is used for further cooling in order to condense water in the product stream, and to obtain SNG product gas with a dew-temperature of –10°C. The condensed water is destined for water-treatment.

CO_2-removal: Selexol unit

In order to bring the Wobbe-index of the gas within the G-gas specification, a Selexol unit removes the excess CO_2 from the gas. The unit separates 94.7% of the CO_2 from the SNG in case of the pressurised O_2-blown gasifier and 89.2% in case of the Battelle gasifier. The CO_2 can partly be recycled to serve as transport gas to the gasifier (in case of the pressurised O_2-blown CFB). The SNG product gas is destined for the natural gas grid. The conditions are 15 bar and 20°C. The Wobbe-index is 43.7 MJ/Nm^3 for both the pressurised O_2-blown and the Battelle gasifier, which is between the quality limits > 43.46 and < 44.41 MJ/Nm^3 for G-gas. It is assumed that the Selexol unit consumes 1.07 kJ of electric power for each mole of CO_2 that is removed.

Steam system

The heat released during several stages of the process is used for the generation and superheating of 40 bar steam. This steam is used for the generation of electricity by expanding in a condensing steam turbine to 0.05 bar.

Simulation results

The simulation is based on willow wood as biomass. The specification of willow wood is obtained from Phyllis[41], and is presented in table 4.4 (for detailed specification see appendix I). The amount of biomass will correspond to a thermal input of 100 MW. As the biomass is fed to the gasifiers with a moisture content of 15% additional drying of the biomass is not required. The main mass and energy flows, as well as the specifications of the SNG product gases are presented in table 4.5.

Table 4.4 *Specification of the used biomass*

		Willow
Moisture Content	[wt.%$_{wet}$]	15.00
Volatile Matter	[wt.%$_{wet}$]	81.51
Ash	[wt.%$_{dry}$]	1.91
Fixed Carbon	[wt.%$_{dry}$]	16.58
Carbon	[wt.%$_{dry}$]	48.72
Hydrogen	[wt.%$_{dry}$]	5.94
Oxygen	[wt.%$_{dry}$]	42.74
Nitrogen	[wt.%$_{dry}$]	0.62
Sulphur	[wt.%$_{dry}$]	0.06
Chlorine	[wt.%$_{dry}$]	0.01
Fluor	[wt.%$_{dry}$]	0.00
LHV$_{Aspen\ Plus}$	[MJ/kg$_{dry}$]	18.2

4.5 Economic analysis

In order to determine the economic feasibility of the process, a model has been constructed based on SNG production costs, as well as specific environmental costs (costs per tonne CO_2 avoided). The diagram for the determination of the economic feasibility is presented in figure 4.9.

Table 4.5 *Mass & energy flows and SNG product gas specification*

MASS & ENERGY FLOW		Pressurised O_2-blown gasifier		Indirect gasifier (Battelle)	
		Mass kg/s	Energy MW	Mass kg/s	Energy MW
In:	Biomass	6.5	100.0	6.5	100.0
	Steam (water)[*]	1.3	-	0.6	-
	CO_2[*]	1.4	-	-	-
	N_2	-	-	0.003	-
	O_2	1.5	-	-	-
	Power[*]	-	7.7	-	7.7
Out:	SNG	1.7	66.3	1.7	67.0
	Char / Ash	0.3	6.0	0.8[**]	22.3[**]
	Tar	0.1	4.2	0.1	5.0
	CO_2	6.4	-	3.1	-
	Wastewater	2.2	-	1.4	-
	Power	-	5.0		4.8
SNG-to-biomass ratio:		0.26	0.66	0.264	0.67
Carbon conversion:		93.3%		100%[**]	
SNG efficiency:		66.3%		67.0%	
CO_2 separation degree:		94.7%		89.2%	
SNG SPECIFICATION					
CH_4	[mol.%]	87.67		87.62	
H_2	[mol.%]	1.77		1.95	
CO_2	[mol.%]	8.65		8.90	
C_{2+}	[mol.%]	-		-	
N_2	[mol.%]	1.84		1.44	
O_2	[mol.%]	-		-	
H_2O	[mol.%]	0.01		0.01	
CO	[mol.%]	0.05		0.06	
LHV	[MJ/kg]	38.41		38.41	
LHV	[MJ/Nm^3]	31.26		31.26	
HHV	[MJ/kg]	42.64		42.64	
Wobbe [MJ/Nm^3]		43.74		43.74	

[*] *Steam and CO_2 are produced within the process. Electricity is partly produced within the process.*

[**] *The amount of char mentioned has to be combusted completely within the second reactor of the Battelle gasifier in order to generate the heat required within the primary reactor. Hence, the carbon conversion will, in case of the Battelle gasifier, be equal to 100%.*

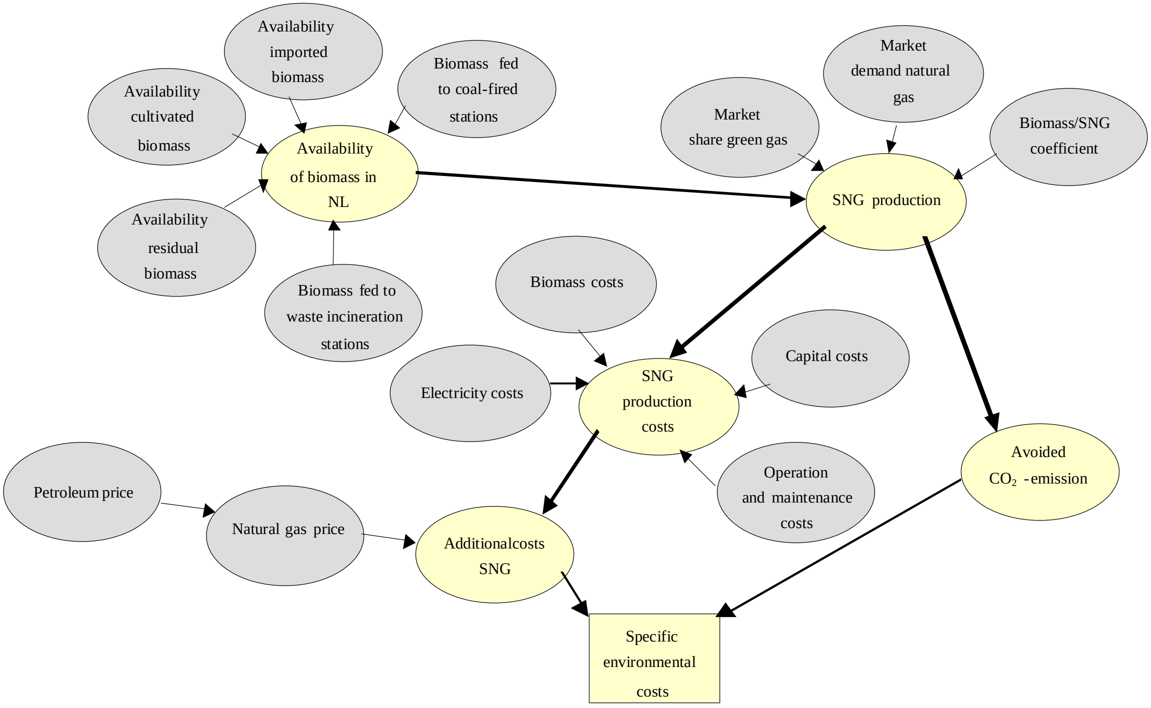

Figure 4.9 *Economic feasibility diagram*

Based on the availability of biomass in the Netherlands, as well as the specific process parameters (Aspen Plus simulations), the amount of produced SNG is determined.

The SNG production costs are determined by taking into consideration the capital investment costs, the operating and maintenance costs, the electricity price (taking the required additional external electricity into account), as well as the costs of biomass. Considering the costs of natural gas, the "additional production costs" of the SNG can be determined.

4.5.1 Availability and costs of biomass

The availability of biomass is related to the availability of residual, cultivated and imported biomass, as well as to the estimation of biomass consumption due to feeding of the biomass to coal-fired power plants and waste incineration stations (figure 4.9). The availability of residual biomass is supposed to linearly increase from 159 PJ in the year 2000 to 196 PJ in 2020, taking into consideration an uncertainty factor of ±10%[42]. 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 2000 PJ[87]. Both are considered to have a uniform probability distribution.

The amount of biomass fed to waste incineration stations at the moment is approximately 40 PJ a year and is expected to remain the same until 2015. Between 2015 and 2020 the amount of biomass is estimated to linearly decrease to 12 PJ in 2020[42], due to closure of a number of waste incineration stations. Similar to the amount of biomass fed to waste incineration, the amount of biomass fed to coal-fired stations will decrease, as a result of the closure of a number of coal-fired stations. Estimations are, that the amount of biomass fed to coal-fired stations until 2010 will be between 5 and 50 PJ a year with an expected value of 25 PJ. Between 2010 and 2020 this amount is expected to decrease with 6.7% a year[42].

As a result of market value, biomass has to be associated with positive costs. The costs, as given in table 4.6, are presented in a certain range with a mean value in order to enable sensitivity and risk analyses of the SNG production costs towards the uncertainty in biomass costs. The probability distribution of the cost of willow wood is given in figure 4.10.

Table 4.6 *The costs of biomass (willow wood)*[46][75]

		Minimum	Mean	Maximum
Costs	[€/dry tonne]	0	45.4	63.5
Costs	[€/GJ]	0	2.5	3.6

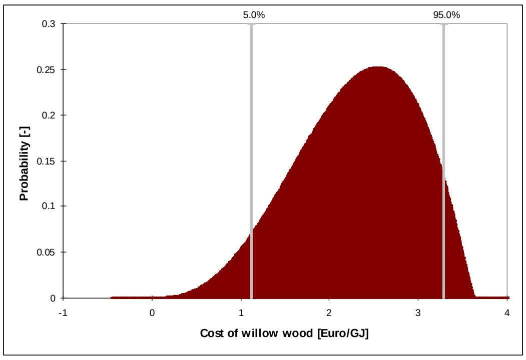

Figure 4.10 *Probability distribution of the cost of willow wood*

Within the new policy on emissions by power production from biomass and waste, willow wood is considered to be a clean biomass, as long as it has not been subjected to protective coating or impregnating. As a result, the application of willow wood is considered to be CO_2 neutral.

4.5.2 SNG production cost

In order to determine the production costs of SNG, several assumptions have been made as presented in table 4.7. Beside the general input parameters of the sensitivity and risk analysis on willow wood gasification for SNG production mentioned in table 4.7, the pressurised O_2-blown and the Battelle gasifiers have some specific input parameters. These specific parameters are presented in tables 4.8 and 4.9.

Table 4.7 *Input parameters of the sensitivity and risk analysis on willow wood gasification for SNG production*

Name	@Risk*	Minimum	Expected value	Maximum	Unit
Availability residual biomass	Linear	159 $^{\text{(in 2000)}}$	-	196 $^{\text{(in 2020)}}$	[PJ]
Uncertainty factor residual biomass	Normal(1)	normal distribution (μ=1, σ=0.1)			[-]
Availability cultivated biomass	Uniform(25)	0	-	50	[PJ]
Availability imported biomass	Uniform(1000)	0	-	2000	[PJ]
Biomass fed to coal-fired stations	Pert(25.8)	5	25	50	[PJ]
Biomass fed to waste incineration stations	40	12	-	40	[PJ]
Natural gas consumption[43]	Pert(1500)	1400	1500	1600	[PJ]
Potential market share SNG	Pert(1.5)	0	1	5	[%]
Plant capacity factor	0.9	-	-	-	[-]
Specific operating and maintenance costs	Pert(0.05)	0.03	0.05	0.07	[-]
Biomass costs	Pert(2.3)	0	2.5	3.6	[€/GJ]
Mineral oil costs	Pert(20.2)	15	20	26	[$/Barrel]
Ash/carbon processing[44]	Pert(68)	61	68	75	[€/tonne]
Electricity price (grey) [44]	Pert(3.6)	2.5	3.8	4.0	[€ct/kWh]
Dollar exchange rate	Pert(1.17)	0.91	1.18	1.36	[€/$]
Natural gas costs	related to the costs of mineral oil				[€/GJ]
Interest rate	Pert(0.06)	0.04	0.06	0.08	[-]
Depreciation period	Uniform(13)	10	-	15	[yr]
Specific CO_2-emission of natural gas	56	-	-	-	[kg CO_2/GJ]

* *Either the (fixed) value of the parameter or the distribution of the parameter based on minimum, maximum and expected value is presented (with the calculation value mentioned in parenthesis). The depreciation period, as well as the required time for RD&D and market introduction should have a round value. The Normal, Pert, Cumul and Uniform distribution are described in the appendix II.*

Table 4.8 *Specific input parameters of the sensitivity and risk analysis on willow wood gasification for SNG production in case of a pressurised O_2-blown gasifier*

Name	@Risk	Minimum	Expected value	Maximum	Unit
Specific investment costs	Pert(485)	386	482	593	[€/kW$_{\text{th}}$]
Required RD&D time	Pert(8)	7	8	9	[yr]
Required time for market introduction	Pert(4)	3	4	5	[yr]
Biomass efficiency:	0.663	-	-	-	[-]
Power requirement	2.7	-	-	-	[MW$_{\text{e}}$]

Table 4.9 *Specific input parameters of the sensitivity and risk analysis on willow wood gasification for SNG production in case of a Battelle gasifier*

Name	@Risk	Minimum	Expected value	Maximum	Unit
Specific investment costs	Pert(440)	340	449	506	[€/kW$_{\text{th}}$]
Required RD&D time	Pert(10)	9	10	11	[yr]
Required time for market introduction	Pert(4)	3	4	5	[yr]
Biomass efficiency:	0.670	-	-	-	[-]
Power requirement	2.8	-	-	-	[MW$_{\text{e}}$]

The required RD&D time for pressurised O_2-blown gasification was determined to be between 7 and 9 years in 2000. As the indirect gasification concept is less developed than a CFB gasification concept, it is assumed that the development of the Battelle gasifier will require an additional 2 years, compared with the pressurised O_2-blown gasification concept. The required time for market introduction is in both cases considered to be between 3 and 5 years. The biomass efficiency, defined as the thermal output of SNG divided by the thermal input of biomass, is based on the Aspen Plus modelling results.

The investment costs are based on the study on integrated gasification systems[76]. A specific breakdown of the investment costs of the pressurised O_2-blown gasifier and the Battelle gasifier is presented in table 4.10. Minimum costs are based on relative cheap advanced systems that will become more or less representative for the gasifiers, whereas maximum costs are based on relative expensive prototype gasification systems.

In case of O_2-blown gasification an additional investment related to the oxygen plant has to be taken into consideration. Polman et al.[2] mention in case of a 100 MW_{th} biomass gasifier an investment cost of M€ 3.85 for an oxygen plant. Furthermore, this oxygen plant will consume approximately 4 MW electricity[2]. In case of the Battelle gasifier the costs of the reactor vessel (at atmospheric conditions) are doubled as the gasification process consists out of two reactor vessels.

As the investment costs of a pressurised gasifier will be higher than of an atmospheric gasifier, a pressure factor[45] is applied, in order to estimate the cost of the CFB-systems at elevated pressure. The pressure factor, pf, is given by:

$$\mathrm{pf} = 2.79 \cdot \mathrm{capacity}^{-0.0597} \tag{4.12}$$

with the capacity in kg per hour (dry and ash free). Furthermore it is assumed that the investment costs related to the feeding system will be twice as high in case of a pressurised system in comparison to an atmospheric system. The gas clean-up in case of the pressurised gasification, on the other hand, will require half the investment costs of the clean-up in case of atmospheric operation. The investment costs related to compression will be lower for pressurised O_2-blown gasification than in case of atmospheric gasification, however, will still be present due to the compression of the oxygen.

Typical costs of methanation in case of O_2-blown gasification process with thermal biomass input of 100 MW_{th} are approximately M€7.0[2]. This includes thorough gas clean-up, methanation, cooling and drying, CO_2 removal and delivery to the gas net. All these costs include cost of engineering, piping etc.

The investment costs of the pressurised O_2-blown CFB gasification process correspond well with the M€ 50.8 mentioned for the investment costs within the study of Polman et al.[2]. Based on the determined distribution of the specific investment costs, as well as on the (distribution of the) parameters presented in tables 4.7 to 4.9, a sensitivity and risk analysis is performed on the SNG production costs, and on the cost per tonne CO_2 avoided. The distributions of the parameters are based on the material balances, obtained from the Aspen Plus simulations, and on expert opinions[46].

The expected SNG production costs amount to €8.5 per GJ in case of pressurised O_2-blown gasification and to €7.8 per GJ in case of Battelle gasification. The distributions of the costs, as well as the uncertainties on these costs are presented in figures 4.11 and 4.12. The figures show that there is a 90% probability that in case of pressurised O_2-blown gasification the SNG production costs will be between €6.6 and €10.2 per GJ. In case of Battelle gasification there is a 90% probability that the costs will be between €6.0 and €9.5 per GJ. The minimum and maximum costs are presented in tables 4.11 and 4.12.

Table 4.10 *Breakdown of the investment costs(in M€) of SNG production process*[76]

	Pressurised O_2-blown CFB			Battelle		
	min	*mean*	*max*	*min*	*mean*	*max*
Pre-treatment	0.7	0.7	0.9	0.7	0.7	0.9
Conveyers	0.4	0.4	0.5	0.4	0.4	0.5
Storage	0.9	1.1	1.2	0.9	1.1	1.2
Feeding systems (2 screw feeders)	0.8	1.0	1.1	0.4	0.5	0.6
Gasifier	2.7	4.2	6.0	3.5	5.5	7.8
Cyclones	1.8	3.2	5.0	1.1	2.1	3.2
Gas cooling	4.1	4.8	5.5	2.6	3.1	3.6
Gas cleanup dry	1.5	1.7	2.0	1.5	1.7	2.0
Gas cleanup scrubber	0.6	1.0	1.6	1.1	2.1	3.2
Oxygen plant	3.9	3.9	3.9	---	---	---
Compressor	0.6	0.8	1.0	1.9	2.6	3.4
Instrumentation & control	0.5	0.7	1.0	0.6	0.9	1.1
Buildings	0.2	0.2	0.3	0.2	0.3	0.4
Grid connections	0.5	0.7	0.9	0.6	0.8	1.1
Site preparation	0.1	0.1	0.1	0.1	0.1	0.1
Civil works	1.2	1.7	2.2	1.4	1.9	2.6
Electronics	0.7	1.0	1.4	0.8	1.2	1.6
Piping	0.5	0.7	0.9	0.6	0.8	1.1
Engineering	3.2	4.2	5.3	2.8	3.9	5.2
Building int. 1st year	0.5	0.7	0.9	0.5	0.6	0.9
Building int. 2nd year	1.6	2.1	2.7	1.4	1.9	2.6
Project contingency	2.2	2.8	3.6	1.8	2.6	3.4
Fees / overheads / profits	1.7	2.2	2.8	1.5	2.1	2.8
Start-up costs	0.9	1.1	1.4	0.7	1.0	1.4
Methanation section	7.0	7.0	7.0	7.0	7.0	7.0
Total investment	38.6	48.2	59.3	34.0	44.9	50.6
Specific investment costs [€/kW$_{th}$]	386	482	593	340	449	506

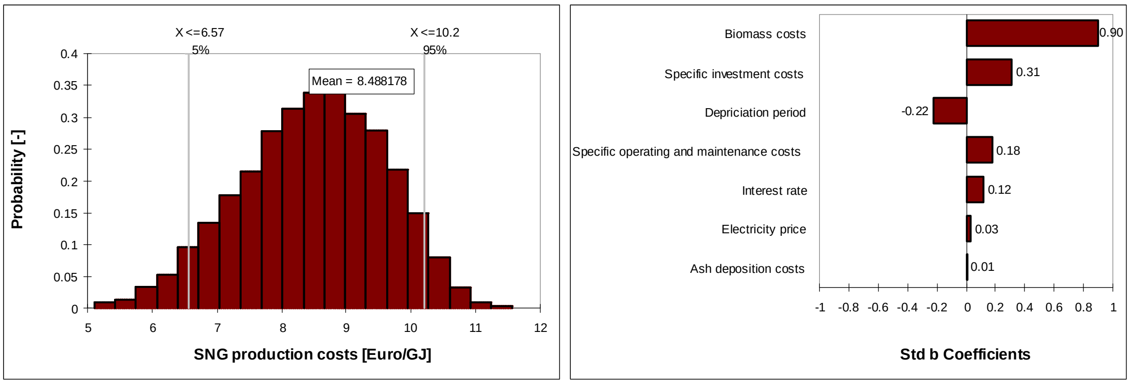

Figure 4.11 *Probability distribution as well as the Tornado graph of the sensitivity analysis of SNG production costs in case of pressurised O_2-blown gasification*

Table 4.11 *SNG production potential until 2020 and SNG production costs in case of pressurised O_2-blown gasification*

		Minimum	Mean	Maximum
SNG production costs	[€/GJ]	5.1	8.5	11.6
SNG production capacity until 2020	[PJ]	0	157	509
Maximum SNG production capacity a year	[PJ/yr]	0.0	14.9	47.1

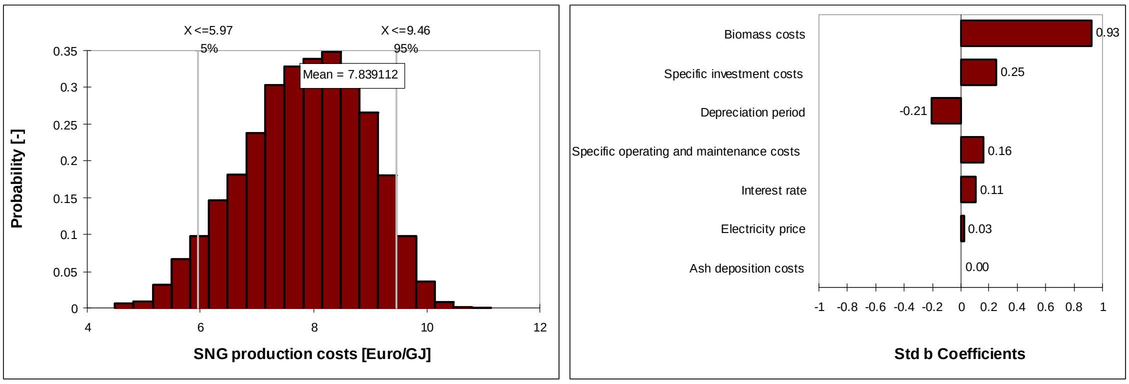

Figure 4.12 *Probability distribution as well as the Tornado graph of the sensitivity analysis of SNG production costs in case of Battelle gasification*

Table 4.12 *SNG production potential until 2020 and SNG production costs in case of Battelle gasification*

		Minimum	Mean	Maximum
SNG production costs	[€/GJ]	4.5	7.8	11.1
SNG production capacity until 2020	[PJ]	0	128	410
Maximum SNG production capacity a year	[PJ/yr]	0.0	15.1	46.5

The Tornado graphs (figures 4.11 and 4.12) show the result of the sensitivity of the SNG production costs to the different input parameters. It can be concluded, that the uncertainty in biomass costs has the largest influence on the uncertainty in SNG production costs, followed by the uncertainties in specific investment costs and the depreciation period. The Tornado graphs also show whether the costs are positively or negatively related to a certain parameter. The biomass costs, for example, have a positive correlation to the SNG production costs, whereas the depreciation period is negatively correlated to the costs. In other words, higher biomass costs will increase the costs of SNG production, whereas a higher depreciation period will decrease the costs.

Beside SNG production costs, also the potential of SNG production until 2020 and the SNG production in 2020 are presented in tables 4.11 and 4.12 as well as in figures 4.13 and 4.14. The SNG production within one year (in this case 2020) represents the potential SNG production per year after the gasification process is completely developed and implemented.

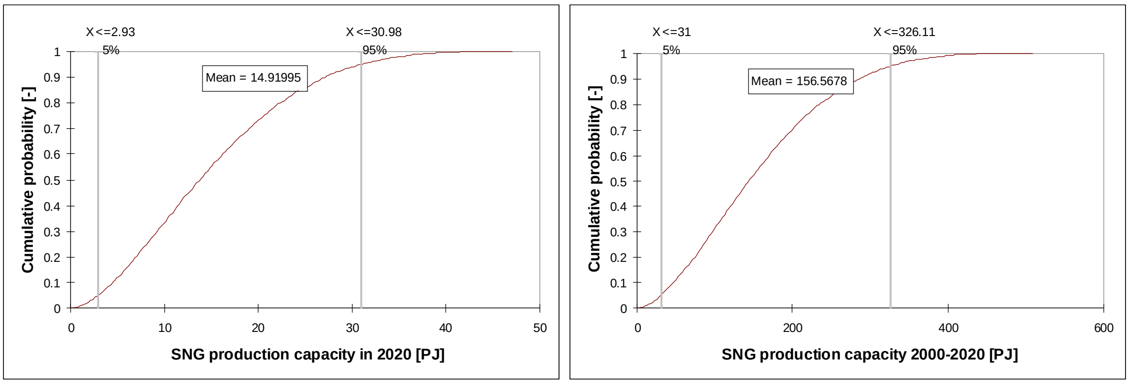

Figure 4.13 *Cumulative probability distributions of the potential SNG production until 2020 as well as the potential SNG production within one year in case of pressurised O_2-blown gasification*

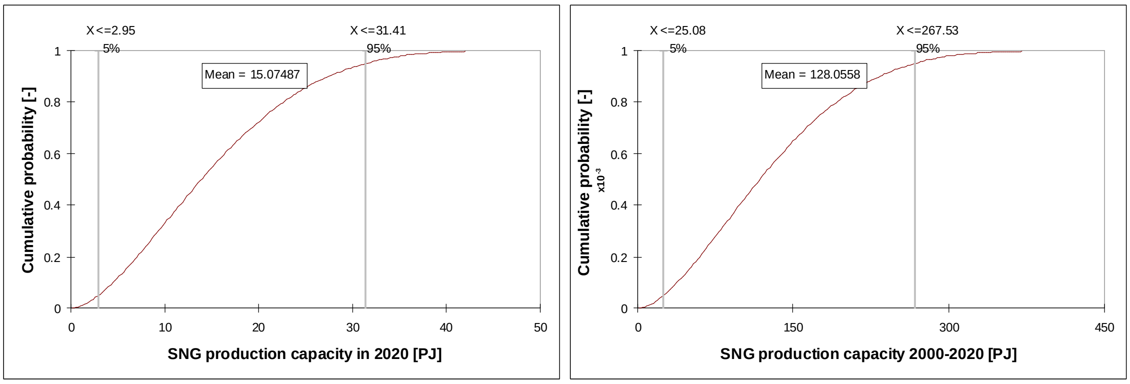

Figure 4.14 *Cumulative probability distributions of the potential SNG production until 2020 as well as the potential SNG production within one year in case of Battelle gasification*

With a potential SNG production capacity of approximately 15 PJ a year, SNG production by either pressurised O_2-blown gasification or Battelle gasification has the potential to fulfil approximately 5% of the Dutch renewable energy target in 2020, or 20% of the biomass related target (see table 2.4). The SNG production capacity of 15 PJ complies with approximately 4.5% of the current total domestic demand for natural gas in the Netherlands (see table 4.13).

Table 4.13 *Natural gas consumption for each province in the Netherlands in 1999*[47]

Province	Citizens [-]	Households [-]	Natural gas consumption* [PJ]
Drenthe	469,800	203,509	9.9
Flevoland	317,200	137,405	6.7
Friesland	624,500	270,522	13.1
Gelderland	1,919,200	831,363	40.4
Groningen	562,600	243,708	11.8
Limburg	1,141,200	494,347	24.0
Noord-Brabant	2,356,000	1,020,577	49.6
Noord-Holland	2,518,400	1,090,926	53.0
Overijssel	1,077,600	466,797	22.7
Utrecht	1,107,800	479,879	23.3
Zeeland	371,900	161,100	7.8
Zuid-Holland	3,397,700	1,471,823	71.5
The Netherlands	15,863,900	6,871,957	334

* *The total domestic natural gas consumption in 1999 amounted to 334 PJ. Based on this amount and the distribution of the Dutch population the natural gas con-sumption per province has been determined*

4.6 Ecological analysis

By producing SNG a certain amount of CO_2 emission is avoided due to the replacement of natural gas by SNG. This amount of avoided CO_2 emission can be calculated from the amount of SNG produced. The specific environmental costs involved can be determined by relating the additional production costs to the avoided CO_2 emission.

The expected costs per tonne CO_2 avoided are equal to €95 per tonne in case of pressurised O_2-blown gasification and €83 per tonne in case of Battelle gasification. The distributions of the costs, as well as the uncertainties on these costs are presented in figures 4.15 and 4.16. The figures shows that there is a 90% probability that the costs per tonne CO_2 avoided will be between €59 and €127 per tonne in case of pressurised O_2-blown gasification and between €48 and €115 per tonne in case of Battelle gasification. The minimum and maximum costs are presented in tables 4.14 and 4.15.

Based on the potential SNG production until 2020 an expected potential reduction of CO_2 emission of 840 ktonne a year has been determined (tables 4.14 and 4.15, as well as figures 4.17 and 4.18).

Table 4.14 *Potential CO_2 emission avoided until 2020 and costs per tonne CO_2 avoided in case of pressurised O_2-blown gasification*

		Minimum	Mean	Maximum
Costs per tonne CO_2 avoided	[€/tonne]	26.8	94.8	150.2
Avoided CO_2 emission until 2020	[Mtonne]	0.0	8.8	28.5
Maximum avoided CO_2 emission a year	[Mtonne/yr]	0.00	0.84	2.63

Table 4.15 *Potential CO_2 emission avoided until 2020 and costs per tonne CO_2 avoided in case of Battelle gasification*

		Minimum	Mean	Maximum
Costs per tonne CO_2 avoided	[€/tonne]	7.5	83.2	147.5
Avoided CO_2 emission until 2020	[Mtonne]	0.0	7.2	23.0
Maximum avoided CO_2 emission a year	[Mtonne/yr]	0.00	0.84	2.60

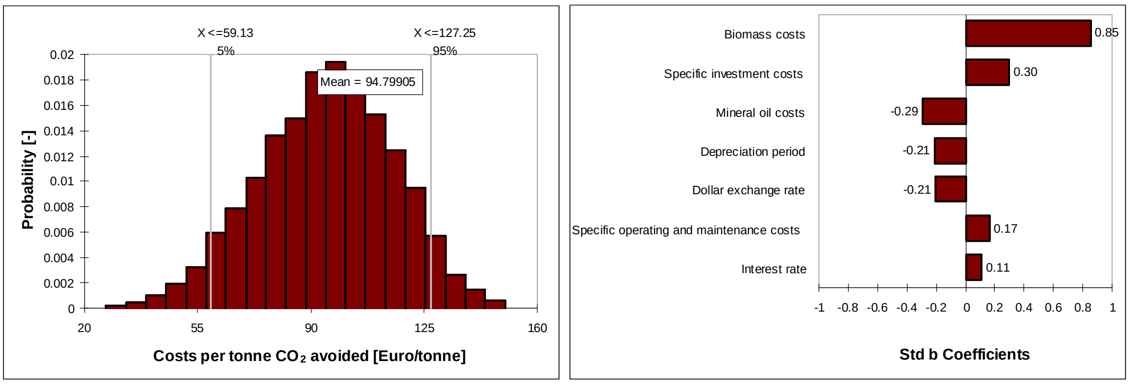

Figure 4.15 *Probability distribution as well as the Tornado graph of the sensitivity analysis of costs per tonne CO_2 avoided in case of pressurised O_2-blown gasification*

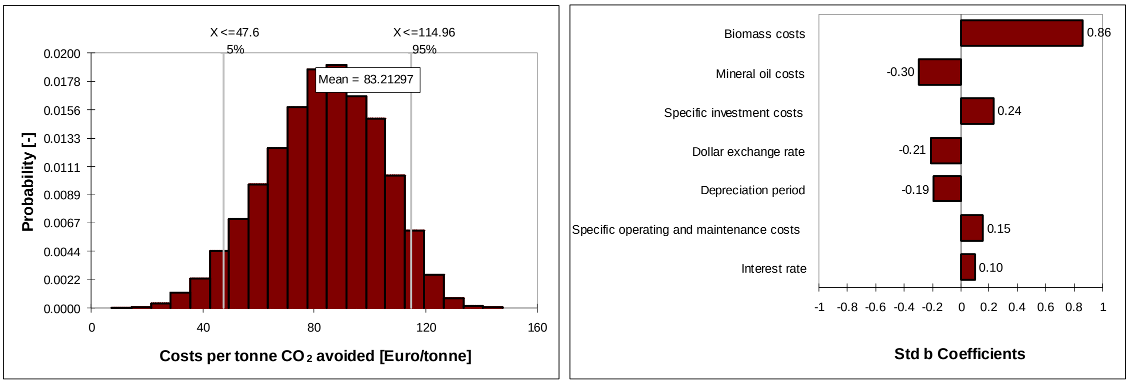

Figure 4.16 *Probability distribution as well as the Tornado graph of the sensitivity analysis of costs per tonne CO_2 avoided in case of Battelle gasification*

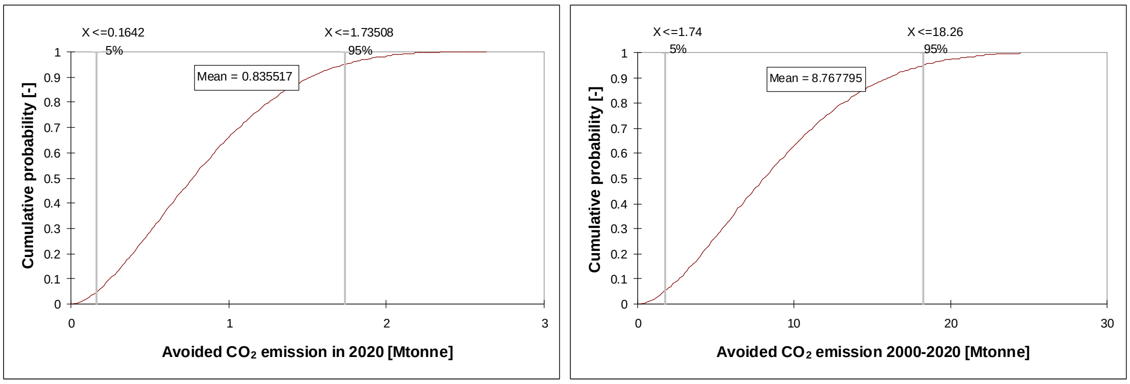

Figure 4.17 *Cumulative probability distributions of the* CO_2 *emission potentially avoided until 2020 as well as the potential* CO_2 *emission avoided within one year in case of pressurised* O_2*-blown gasification*

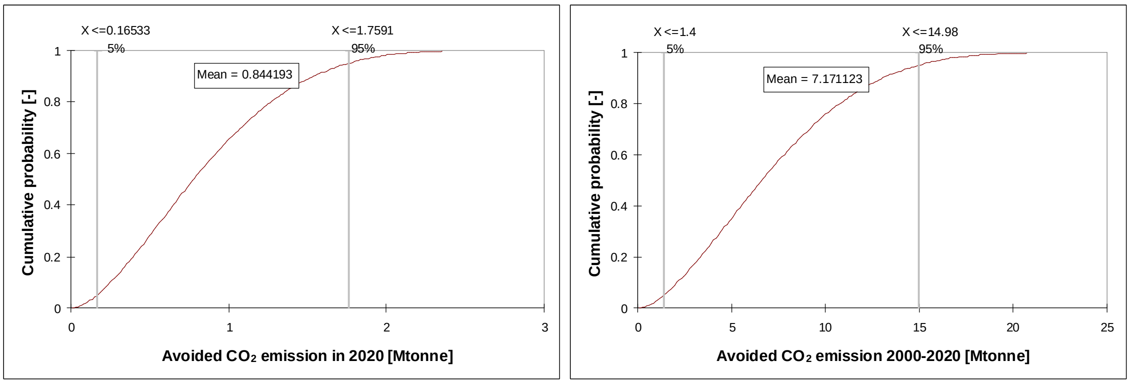

Figure 4.18 *Cumulative probability distributions of the* CO_2 *emission potentially avoided until 2020 as well as the potential* CO_2 *emission avoided within one year in case of Battelle gasification*

The Tornado graphs in figures 4.15 and 4.16 show the result of the sensitivity of the costs per tonne CO_2 avoided to the different input parameters. It can be concluded that, again, the uncertainties in costs of biomass and specific investment costs have a large influence on the uncertainty of the costs per tonne CO_2 avoided. The uncertainty in mineral oil costs, however, now also has a large influence as the natural gas costs, hence the additional SNG production costs, are related to these mineral oil costs.

4.7 Conclusions

The maximum amount of SNG that can be produced within one year is in case of pressurised O_2-blown gasification as well as Battelle gasification approximately 15 PJ. The associated amount of CO_2 emission that is avoided in case of this production is equal to 0.84 Mtonne per year. The costs involved with this CO_2 emission reduction depends on the gasification concept that is used. In case of pressurised O_2-blown gasification the SNG production costs amount to 8.5 €/GJ (hence, costs per tonne CO_2 avoided equal to €95) and in case of Battelle gasification 7.8 €/GJ (costs per tonne CO_2 avoided equal to €83).

Although the CO_2 emission reduction costs are lower, the total amount of CO_2 emission that can be avoided until 2020 is however less in case of the Battelle gasification concept (7.2 Mtonne compared to the 8.8 Mtonne in case of pressurised O_2-blown gasification). As the indirect gasification concept is less developed than a CFB gasification concept, the development of the Battelle gasifier is assumed to require an additional 2 years compared with the pressurised O_2-blown gasification concept, hence, less SNG can be produced until 2020.

4.8 A Dutch SNG production initiative

An interesting opportunity for the production of SNG from biomass and organic wastes in the Netherlands is the co-production at the 253 MW_e Willem-Alexander coal gasification combined cycle (IGCC) power plant at Buggenum. Various options have been studied, with the objective to optimise the economics of the plant to become a strong competitor on the liberalised power market. In the present situation, the plant is operated at partial load (approximately, 57%) during off-peak hours, since off-peak electricity production is not cost-effective. However, by maintaining the gasification unit at full load (460 MW_{th}) during off-peak hours, a surplus of syngas may be produced, which may be converted into liquid or gaseous energy carriers.

De Lange et al.[79] have examined four options for off-peak co-production from a technical and economic point of view, viz. co-production of hydrogen, methanol, Fischer-Tropsch[17] transportation fuels, and SNG. The produced hydrogen or SNG would be supplied into the existing natural gas network. It is assumed that the co-production section will be in operation during off-peak hours only, and involves 200 of the 460 MW total thermal plant capacity. In addition, it has been assumed that the gasifier is operated on a mixture of 24% "green fuels" (biomass[18] with a price of €0.42/GJ), 21% "non-green" alternative fuels[19] (€0.22/GJ) and 55% coal (€2.04/GJ). The flexibility of the SNG co-production option is limited by the methanation section, which cannot be hold on a hot stand-by. Its minimum capacity, however, amounts to 10%.

All co-production options, except for Fischer-Tropsch transportation fuels, are found to be financially viable. Additional costs and additional income are compared with the base case in which only electricity is produced, as described above. The co-production of Fischer-Tropsch transportation fuels was not financially viable at the assumed fossil crude oil price of 15 US$/barrel; a price level of at least 23-29 US$/barrel was required. For the produced SNG a price of €2.54/GJ was required for a zero yearly income, while an SNG market price of €4.54/GJ would lead to a net yearly income of M€4.0.

Based on the above-mentioned co-production ideas, the Dutch Research Institute for Environment, Energy and Process Innovation (TNO-MEP) and NUON have recently started a Gave[20]-project for SNG production at the Willem-Alexander power station at Buggenum[80]. For this purpose the plant will be extended with a unit for biomass delivery and supply, and an SNG plant[80]. The plant feedstock would change from 100% coal to 50 wt.% biomass (372 ktonne with 28% of the total thermal input) and 50 wt.% coal (with 72% of the total thermal input). This will result in the production of 210 MW electricity and 44 ktonne of SNG[81]. The produced "green gas" can replace the natural gas in the distribution network, as well as in natural gas vehicles[80].

As mentioned before, this initiative is an interesting opportunity for demonstration of SNG production from biomass and organic wastes in the Netherlands. However, the entrained flow Shell gasifier at Buggenum results in a product gas with a very high CO concentration (about 60 mol.%) and no methane or C_{2+} fractions. Therefore, it is not the optimal technology for SNG production.

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[17] by once-through synthesis process

[18] chicken manure, plastic/paper pellets (25% green), wood fraction of vegetable/fruits/garden waste, industrial wastewater treatment sludge, sewage sludge[79]

[19] petroleum cokes, plastic/paper pellets (75% non-green), sea harbour waste oil[79]

[20] The objective of the Dutch Gave-programme is to stimulate developments in the field of sustainable, climate neutral energy carriers for replacement of the conventional fossil derived alternatives.

5. SNG PRODUCTION BY BIOMASS HYDROGASIFICATION PROCESS

5.1 Background

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[48], from which SNG production by biomass hydrogasification[21] 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 (figure 5.1).

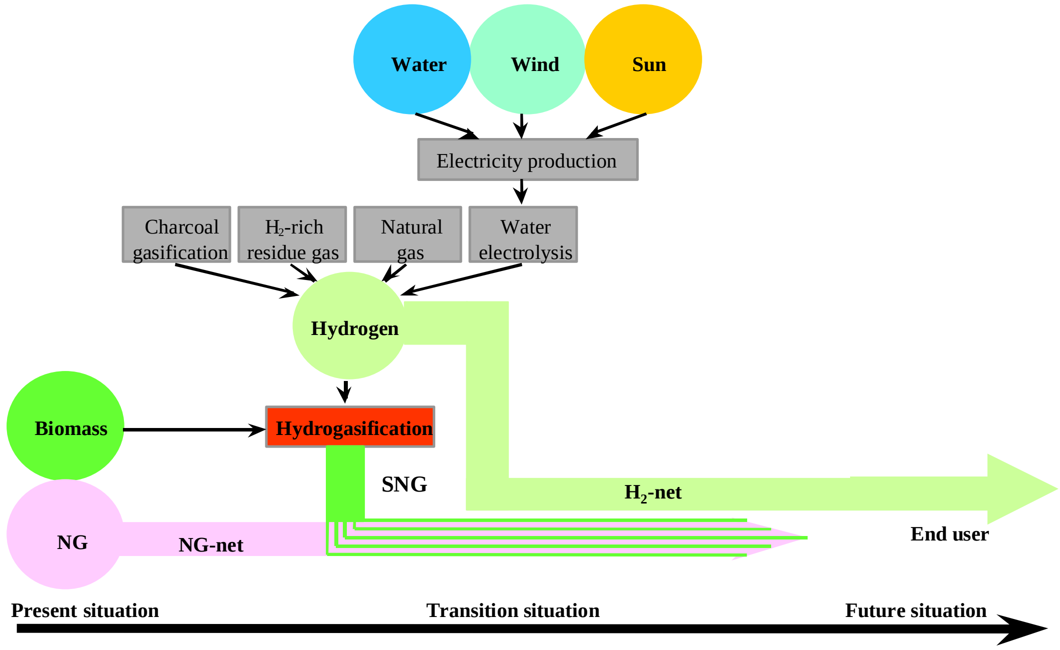

Figure 5.1 *SNG production by biomass hydrogasification with an external hydrogen source*

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[21] Gasification of carbon-containing feedstocks in a hydrogen atmosphere is called hydrogasification.

5.2 State of the art

Hydrogasification of coal has been investigated since the 1930s in Germany, Great Britain and the United States[49]. 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 tonne/day plant in Germany. In the early 1980s, Steinberg et al.[49] 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 over 90% were achieved.

Several processes have been developed for production of methane-rich gases from coal, biomass, or organic solid wastes[50][51][52][53]. The required hydrogen in these processes is produced within the process[3], e.g. by gasification of residual char from the hydrogasifier. The use of an external hydrogen source is new[4], 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.

Figure 5.2 shows a simplified flowsheet of the ECN concept for SNG production by biomass hydrogasification with an external hydrogen source. Hydrogen and biomass are fed to a hydrogasifier, operating at 30 bar and 800-850°C. Due to exothermic reactions taking place in the hydrogasifier, this reactor has the potential to be operated autothermally. The produced gas, with high methane and low carbon monoxide concentrations, passes a gas clean-up section for removal of contaminants, followed by a final methanation step for conversion of residual carbon monoxide. Removal of water from the product gas of the methanation step results in SNG as the final product.

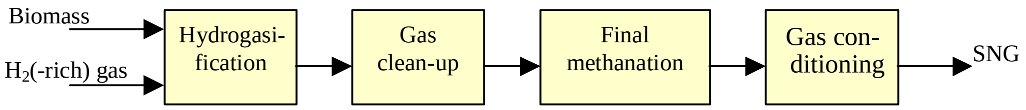

Figure 5.2 *SNG production by biomass hydrogasification with an external hydrogen source*

5.3 Experimental work[5]

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[54]. 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 5.3 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:

$$CO + 3\,H_2 \leftrightarrow CH_4 + H_2O \qquad (5.1)$$

$$CO_2 + H_2 \leftrightarrow CO + H_2O \qquad (5.2)$$

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:

$$C_2H_4 + H_2 \leftrightarrow C_2H_6 \qquad (5.3)$$

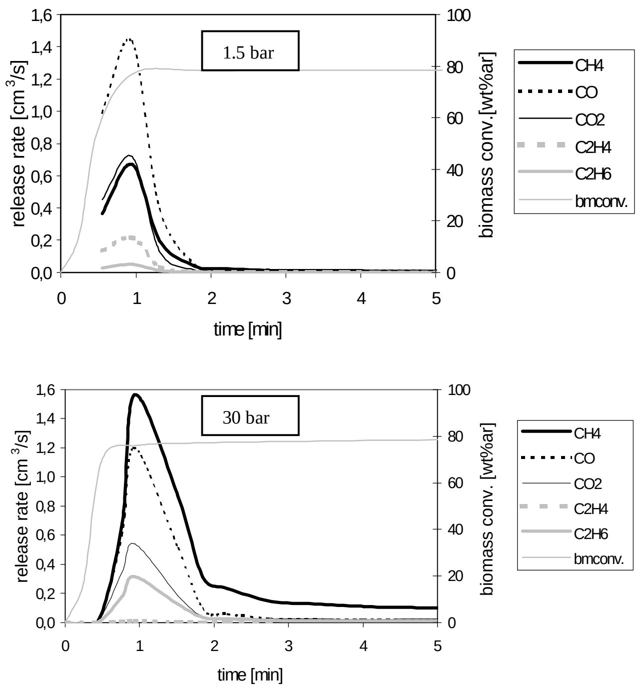

Figure 5.3 *Biomass conversion and release rate of main carbon-containing components in the product gas at 1.5 bar and 30 bar (T = 850ºC)*

At 850ºC 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 hydrogasification of char. Figure 5.4 presents the mean concentration of the main carbon-containing components in the product gas as a function of pressure. Figure 5.5 presents the

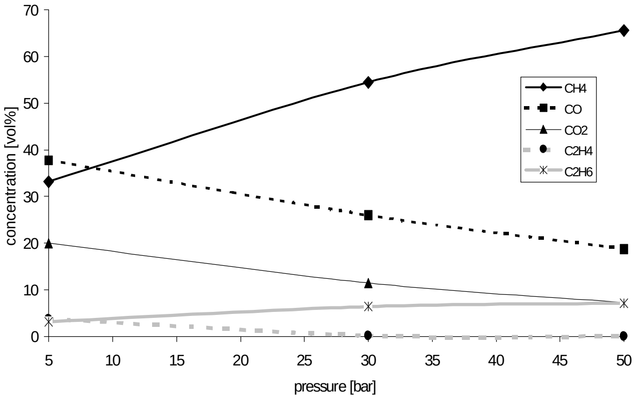

Figure 5.4 *Mean concentration of the main carbon-containing components in the product gas at 850ºC as a function of pressure*

conversion of biomass and carbon as a function of time, for different operating 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.

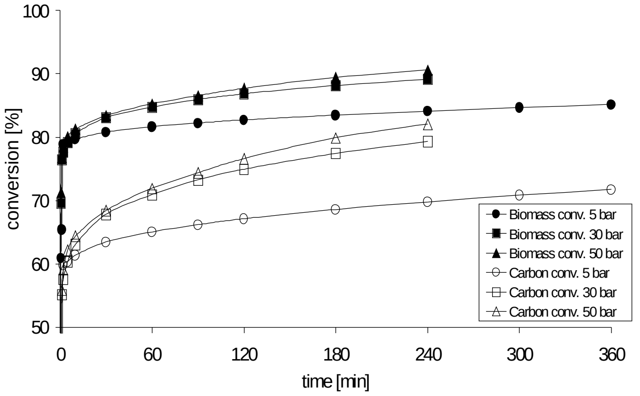

Figure 5.5 *Biomass and carbon conversions at 850ºC as function of time for different operating pressures*

Char gasification with steam resulted in the highest conversion rate, followed by CO_2 and H_2 gasification. Based on these results, 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:

$$C + 2\,H_2 \leftrightarrow CH_4 \qquad (5.4)$$
$$C + CO_2 \leftrightarrow 2\,CO \qquad (5.5)$$
$$C + H_2O \leftrightarrow CO + H_2 \qquad (5.6)$$

Beside the thermobalance experiments, also some tests were carried out at 5 bar and 800°C in the DMT pressurised fluidised bed (PFB) facility[55]. 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.

It should, however, be mentioned that 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 recycled, 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.

5.4 Reactor modelling[7]

A kinetic model has been taken from a study performed by Dong and Borgwardt concerning the biomass reactivity in gasification by the Hynol process[56][57]. 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 5.1, 5.2, and 5.3, whereas the heterogeneous reaction 5.4 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:

$$\frac{dX_1}{dt} = k_1\left(X_C - X_1\right) \qquad (5.7)$$

$$\frac{dX_2}{dt} = k_2\left(1 - X_C - X_2\right) \qquad (5.8)$$

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:

$$X_C = a + b\ T \qquad (5.9)$$

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[58][59][60], 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):

$$F(t) = 1 - \left(1 - C_A\right) \cdot X_{ar} \qquad (5.10)$$

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:

$$\mathrm{M}(t) = \Phi_{m,bm} \cdot \int_0^t F(t)\ dt \qquad (5.11)$$

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:

$$S_{reactor} = \frac{\Phi_{v,g}}{u} \qquad (5.12)$$

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:

$$H_{reactor} = \frac{M_{bm}}{\rho_{bm} S_{reactor} (1 - \alpha_{sp})(1 - \varepsilon)} \qquad (5.13)$$

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 one 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 0.5 mm. The results of the reactor modelling are presented in table 5.1.

Table 5.1 *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

5.5 System modelling

In order to determine the mass and energy balances of the biomass hydrogasification process an Aspen Plus model has been developed, as shown in figure 5.6. Hydrogen and pre-treated biomass (dried and reduced in size) are fed to the hydrogasifier. A CO_2 stream is used to pressurise the biomass to the hydrogasification operating pressure. The produced methane-rich syngas, after gas clean-up, is used for the production of SNG as main product. The heat generated at various points in the process is used for steam and electricity generation in a steam cycle, in order to satisfy the demand within the system. The system pressure is 30 bar, from hydrogasification through methanation. Below follows a brief description of the main parts of the model.

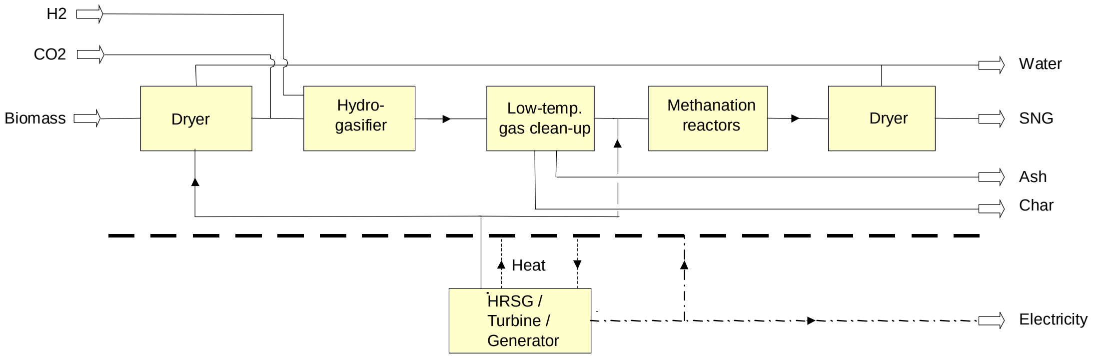

Figure 5.6 *Aspen Plus model for SNG production by combined biomass hydrogasification/ methanation process*

Dryer

The biomass is dried to achieve a water content suitable for hydrogasification (5 wt.%). Heat is supplied to the dryer by condensing steam from the steam system. The water evaporated from the biomass is condensed using cooling water. The condensed water is destined for water-treatment.

Hydrogasifier

The dried biomass, with a moisture content of 5 wt.%, is fed to the gasifier using carbon dioxide as a transportation gas. The gasifier is operated at 30 bar. External hydrogen is compressed to meet the gasification pressure. The gasifier is modelled as a restricted equilibrium reactor with a temperature approach equal to –161.8°C. This temperature approach is set, in order to obtain a carbon conversion of 80% at an operating temperature of 850°C (in accordance with the determined gasification kinetics).

Gas clean-up section
The gas is cleaned at a temperature of 30°C before being preheated to the inlet temperature of the methanation section. The gas clean-up section, however, is not modelled in detail.

Methanation
The syngas is methanated in two serial adiabatic methanation reactors according to the reactor concept applied in the Lurgi coal-to-SNG process (§4.2.3.1). The reactors are modelled as equilibrium reactors and are not restricted by a temperature approach. The operating temperature of the methanation reactors is restricted between 260°C and about 450°C by adding a part of the fresh feed directly to the second methanation reactor, as well as by intercooling and by gas recycling to the first methanation reactor. Simultaneously, the Wobbe-index of the SNG product gas is set to approximately 44.0 MJ/Nm³.

Cooling and drying SNG
The product gas is cooled step by step to 10°C. The first cooler is a heat source for the steam drum. The second cooler pre-heats the boiling feed water. In the third cooler, cooling water is used to bring the gas to 10°C. Finally, an electric cooler is used for further cooling in order to condense water in the product stream, and to obtain SNG product gas with a dew-temperature of –10°C. The condensed water is destined for water-treatment. The reheated SNG product gas is destined for the natural gas grid. The conditions are 30 bar and 20°C.

Steam system
Boiler feed water is pre-heated by cooling the SNG product stream, before entering the steam drum. The heat sources for evaporation are the convective syngas cooler and the product coolers of the two methanation reactors. The steam is then superheated in the radiant syngas cooler. The 40 bar steam is expanded in a steam turbine, driving a generator in order to produce electric power.

Simulation results
The simulation is based on willow wood as biomass. The specification of willow wood is obtained from Phyllis[41], and is presented in table 4.4. (for detailed specification see appendix I). The amount of biomass will correspond to a thermal input of 100 MW. The scale effect of hydrogasification will be discussed later on.

The amount of pressurisation gas (CO_2) is related to the amount of biomass, fed to the gasifier (i.e. the biomass with 5 wt.% moisture content), by 0.195 kg CO_2 per kg biomass. By regulating the amount of (pure) hydrogen fed to the gasifier, an autothermal operation of the hydrogasifier is achieved. It is assumed, that the hydrogasifier has a heat loss equal to 2% of the biomass thermal input. Taking this heat loss into account, the hydrogasifier is considered to be autothermal, in case of a simulated heat production equal to 2% of the biomass thermal input. The main mass and energy flows, as well as the specifications of the SNG product gases are presented in table 5.2.

In order to obtain a dew-temperature of the SNG below –10°C, water in the product stream is condensed by cooling the SNG. The Wobbe-index of the gas is brought within the G-gas specification, by allowing a percentage of the synthesis gas to flow directly to the second methanator, and by recycling a part of the gas, exiting the second methanator, to the first. The temperature in the methanators should thereby remain between approximately 260 and 450°C.

Table 5.2 *Mass & energy flows and SNG product gas specification*

MASS & ENERGY FLOW		Mass kg/s	Energy MW
In:	Biomass	6.5	100.0
	Hydrogen	0.8	94.8
	CO_2	1.2	-
	Power	-	6.2^*
Out:	SNG	3.9	154.1
	Char	0.5	17.5
	Ash	0.1	-
	wastewater	4.0	-
	Power	-	10.7
SNG-to-biomass ratio:		0.630	1.5408
SNG-to-hydrogen ratio:		4.929	1.6262
Carbon conversion:		80.1%	
SNG efficiency:		79.1%	
SNG SPECIFICATION			
CH_4	[mol.%]	82.97	
H_2	[mol.%]	8.02	
CO_2	[mol.%]	8.37	
C_2^+	[mol.%]	0.00	
N_2	[mol.%]	0.53	
O_2	[mol.%]	0.00	
H_2O	[mol.%]	0.01	
CO	[mol.%]	0.09	
LHV	[MJ/kg]	39.57	
LHV	[MJ/Nm³]	30.67	
HHV	[MJ/kg]	43.98	
Wobbe	[MJ/Nm³]	44.03	

* A part of the power, generated within the process.

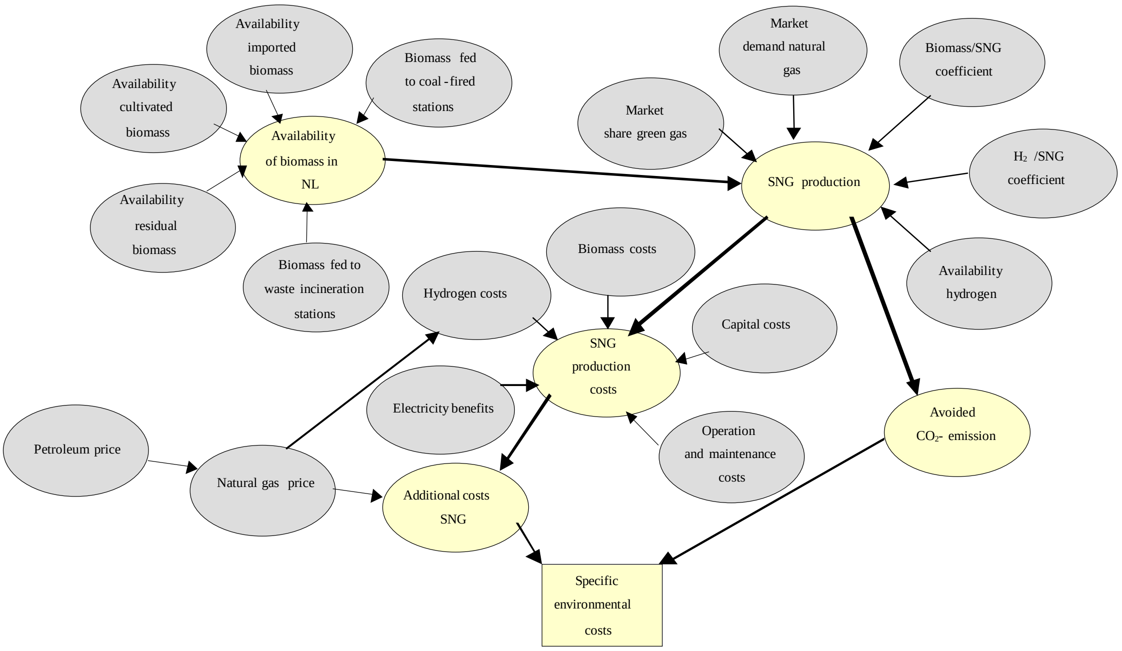

Figure 5.7 *Economic feasibility diagram*

5.6 Economic analysis

In order to determine the economic feasibility of the process, a model has been constructed based on SNG production costs, as well as specific environmental costs (costs per tonne CO_2 avoided, see §5.7). The diagram for the determination of the economic feasibility is presented in figure 5.7.

Based on the availability of biomass and hydrogen in the Netherlands, as well as the specific process parameters (Aspen Plus simulations), the amount of produced SNG is determined.

The SNG production costs are determined by taking into consideration the capital investment costs, the operating and maintenance costs, electricity generation benefits, as well as the costs of hydrogen and biomass. Considering the costs of natural gas, the “additional production costs” of the SNG can be determined.

5.6.1 Availability and costs of resources

Biomass

See §4.5.1.

Hydrogen

Based on an inventory of hydrogen(-rich) residue gases[61] and personal communication[62], the total availability of hydrogen(-rich) residue gases in the Netherlands is estimated to amount to 2884 million Nm³ (19.66 PJ of hydrogen). A list of the different locations, where hydrogen can be obtained as a residue gas, is presented in table 5.3. In order to take into account the market share of the hydrogasification process with regards to the available hydrogen, an uncertainty factor (with expected value of 50%) will be used within the feasibility study (table 5.5).

Due to the small scale, the potential of the hydrogasification based on hydrogen residue gases at Bergen op Zoom and Hengelo is limited. Therefore, these locations will not be taken into consideration within the feasibility study. Based on the remaining five locations, a cumulative distribution of hydrogen availability is constructed (appendix II, as well as table 5.5).

Table 5.3 *The availability of hydrogen(-rich) residue gases in the Netherlands*

Company	Location	Amount of gas	Amount of H_2
Akzo Nobel Base Chemicals BV	Hengelo	22 mln Nm³ hydrogen a year	0.24 PJ
Akzo Nobel Base Chemicals BV	Delfzijl	38 mln Nm³ hydrogen a year	0.41 PJ
Akzo Nobel Base Chemicals BV	Rotterdam	110 mln Nm³ hydrogen a year	1.18 PJ
Carbon Black Nederland BV	Botlek	500 mln Nm³ gas a year (16% H_2)	0.86 PJ
General Electric Plastics BV	Bergen op Zoom	14 mln Nm³ hydrogen a year	0.15 PJ
Corus	IJmuiden	1000 mln Nm³ gas a year (60% H_2)	6.47 PJ
Methanor	Delfzijl	1200 mln Nm³ gas a year (80% H_2)	10.35 PJ
Total amount of hydrogen gas	the Netherlands	2884 mln Nm³ H_2-rich residue gases a year	19.66 PJ

Due to large differences in the availability of hydrogen, the implemented scale of a hydrogasification process at each location (hence the specific investment costs) will show a large variation. In order to take into account the correlation between availability of hydrogen and implemented scale of the process, the hydrogen availability and the specific investment costs will be (negatively) correlated to each other. The applied scales of implementation of the hydrogasifcation process will also take into consideration a market share of the hydrogasification process, with regards to the available hydrogen, of 50%[46].

As a result of the application of hydrogen(-rich) residue gases, the costs of hydrogen are equal to the costs of natural gas, required as a replacement within the current application of the hydrogen residue gases. The natural gas costs are related to the cost of mineral oil and, therefore, take into account the uncertainty of mineral oil costs and dollar exchange rate. The commodity price of gas in €ct/Nm³ is equal to[63]:

$$\frac{37.4}{1100} \cdot P - 0.36 \qquad (5.14)$$

The coefficient P represents the costs of crude mineral oil in €/tonne.

5.6.2 SNG production cost

In order to determine the production costs of SNG, several assumptions have been made and presented in table 5.5.

The implemented scale of the hydrogasification process has to be directly related to the amount of hydrogen available at a certain location. As this amount, at the five locations considered, varies from approximately 0.40 PJ to 10.4 PJ, the scale of the process, hence the specific investment cost, will have a relatively large uncertainty. Based on a market share of the hydrogasification process of 50%[46], 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 might be constructed. The expected value of the thermal input in case of hydrogasification based on hydrogen residue gas is thought to be 50 MW.

The minimal specific investment costs are obtained in case of the 160 MW_{th} hydrogasification, with maximum and expected value resulting from the 5 and 50 MW_{th} hydrogasification. The costs are based on the study on integrated gasification systems[76]. By means of scaling rules, the costs of the 5, 50 and 160 MW_{th} BFB-systems are determined.

$$\frac{Costs_{size"2"}}{Costs_{size"1"}} = \left(\frac{Size"2"}{Size"1"}\right)^x \qquad (5.15)$$

with x between 0 and 1 (in general around 0.6, according to the Williams 0.6 scaling rule).

In addition, a pressure factor is applied, in order to estimate the cost of the BFB-systems at a pressure of 30 bar. The pressure factor, pf, is given by equation 4.12.

The costs of the methanation reactors are based on 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. Therefore, the methanation will be different from the methanation used within the hydrogasification process. 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.

Typical costs of methanation in case of O_2-blown gasification process with thermal biomass input of 100 MW_{th} are approximately M€7.0[2]. This includes thorough gas clean-up, methanation, cooling and drying, CO_2 removal and delivery to the gas net. All costs include cost of engineering, piping etc. The investment costs are determined by downscaling. The breakdown of the specific investment costs is presented in table 5.4.

In order to compare the investment costs of hydrogasification with the pressurised O_2-blown and the Battelle gasifier (chapter 4) a cost breakdown for a 100 MW_{th} hydrogasification process is also presented in table 5.4.

Table 5.4 *The breakdown of the specific investment costs (in M€) of willow wood hydrogasification*

	Thermal biomass input			100 MW$_{th}$ hydrogasification		
	5 MW	*50 MW*	*160 MW*	*min*	*mean*	*max*
Pre-treatment	0.7	4.5	11.3	5.3	7.7	10.8
Conveyers	0.0	0.2	0.6	0.4	0.4	0.5
Storage	0.2	0.7	1.5	0.9	1.1	1.2
Feeding systems (2 screw feeders)	0.0	0.5	1.6	0.8	1.0	1.1
Gasifier	0.6	2.7	5.7	2.7	4.2	6.0
Gas cooling	0.7	3.1	6.5	4.1	4.8	5.5
Gas cleanup dry	0.2	1.1	2.4	1.5	1.7	2.0
Gas cleanup scrubber	0.1	0.6	1.4	0.6	1.0	1.6
Compressor	0.1	0.4	1.2	0.6	0.8	1.0
Instrumentation & control	0.4	0.8	1.1	0.7	0.9	1.2
Buildings	0.0	0.2	0.4	0.2	0.3	0.4
Grid connections	0.5	0.8	1.0	0.6	0.9	1.2
Site preparation	0.0	0.2	0.1	0.1	0.1	0.1
Civil works	0.3	0.8	2.9	1.5	2.1	2.8
Electronics	0.5	0.1	1.5	0.9	1.3	1.7
Piping	0.1	1.4	1.2	0.6	0.9	1.1
Engineering	0.7	1.1	6.0	3.2	4.4	5.8
Building int. 1st year	0.1	0.5	1.0	0.5	0.7	1.0
Building int. 2nd year	0.3	1.4	3.0	1.6	2.2	2.9
Project contingency	0.5	1.9	4.0	2.1	2.9	3.9
Fees / overheads / profits	0.4	1.5	3.2	1.7	2.3	3.1
Start-up costs	0.2	0.7	1.6	0.9	1.2	1.5
Methanation section	0.9	3.4	6.9	5.2	5.2	5.2
Total investment	7.7	30.8	66.1	36.8	48.4	61.8
Specific investment costs [€/kW$_{th}$]	1541	616	413	368	484	618

Based on the determined distribution of the specific investment costs, as well as on the (distribution of the) parameters presented in table 5.5, a sensitivity and risk analysis is performed on the SNG production costs, and on the cost per tonne CO_2 avoided (discussed in §5.7). The distributions of the parameters are based on the material balances, obtained from the Aspen Plus simulation, and on expert opinions[46].

Table 5.5 *Input parameters of the sensitivity and risk analysis on willow wood hydrogasification*

Name	@Risk*	Minimum	Expected value	Maximum	Unit
Availability residual biomass	Linear	159 (in 2000)	-	196 (in 2020)	[PJ]
Uncertainty factor residual biomass	Normal(1)	normal distribution with μ=1 and σ=0.1			[-]
Availability cultivated biomass	Uniform(25)	0	-	50	[PJ]
Availability imported biomass	Uniform(1000)	0	-	2000	[PJ]
Biomass fed to coal-fired stations	Pert(25.8)	5	25	50	[PJ]
Biomass fed to waste incineration stations	40	12	-	40	[PJ]
Availability hydrogen gas	Cumul(7.1)	0	-	20	[PJ]
Heating value hydrogen gas	120	-	-	-	[MJ/kg]
Costs hydrogen gas	related to the natural gas costs				[€/GJ]
Biomass-efficiency**	1.5408	-	-	-	[-]
Hydrogen-efficiency**	1.6262	-	-	-	[-]
Required RD&D time***	Pert(8)	7	8	9	[yr]
Required time for market introduction	Pert(4)	3	4	5	[yr]
Market share with regards to available H_2	Pert(0.5)	0	0.5	1	[-]
Plant capacity factor	0.9	-	-	-	[-]
Specific investment costs****	Pert(819)	413	616	1,541	[€/kW$_{th}$]
Specific operating and maintenance costs	Pert(0.05)	0.03	0.05	0.07	[-]
Biomass costs	Pert(2.3)	0	2.5	3.6	[€GJ]
Mineral oil costs	Pert(20.2)	15	20	26	[$/Barrel]
Dollar exchange rate	Pert(1.17)	0.91	1.18	1.36	[€/$]
Natural gas costs	related to the costs of mineral oil				[€/GJ]
Electricity benefits (green)[64]	Pert (10.3)	9.3	10.6	10.8	[€ct/kWh]
Ash/carbon processing[44]	Pert(68)	61	68	75	[€/tonne]
Interest rate	Pert(0.06)	0.04	0.06	0.08	[-]
Depreciation period	Uniform(13)	10	-	15	[yr]
Specific CO_2-emission of natural gas	56	-	-	-	[kg CO_2/GJ]

* Either the (fixed) value of the parameter or the distribution of the parameter based on minimum, maximum and expected value is presented (with the calculation value mentioned in parenthesis). The depreciation period, as well as the required time for RD&D and market introduction should have a round value. The Normal, Pert, Cumul and Uniform distribution are described in the appendix II.

** The biomass and hydrogen efficiencies are defined as the thermal output of SNG, divided by either the thermal input of biomass, or the thermal input of hydrogen.

*** Starting-point within the feasibility study has been the year 2000.

**** The specific investment costs are directly related to the availability of hydrogen gas, as in case of low availability of hydrogen gas the scale of the hydrogasifier might be restricted by the availability, hence the specific investment costs becoming relatively high.

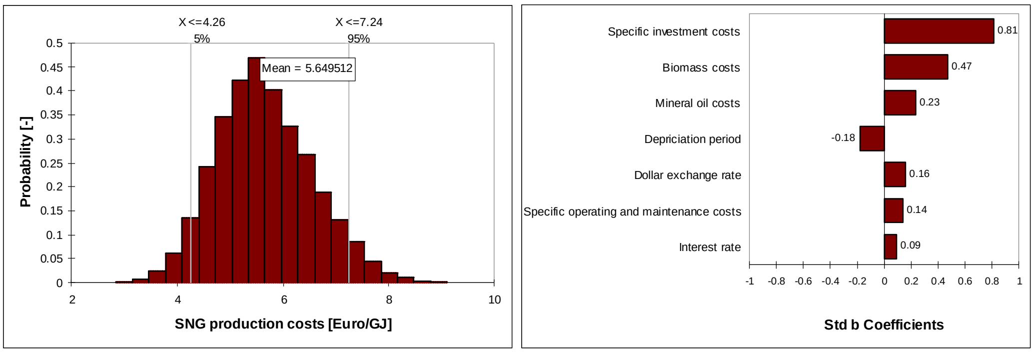

Figure 5.8 *Probability distribution as well as the Tornado graph of the sensitivity analysis of SNG production costs*

The expected SNG production costs are equal to €5.6 per GJ. The distribution of the costs, as well as the uncertainties on these costs are presented in figure 5.8. The figure shows, that there is a 90% probability that the SNG production costs will be between €4.3 and €7.2 per GJ. The minimum and maximum costs are presented in table 5.6.

Table 5.6 *SNG production potential until 2020 and SNG production costs in case of hydrogasification of willow*

		Minimum	Mean	Maximum
SNG production costs	[€/GJ]	2.8	5.6	9.1
SNG production capacity until 2020	[PJ]	0	60	353
Maximum SNG production capacity a year	[PJ/yr]	0	5.7	30.7

The Tornado graph (figure 5.8) shows the result of the sensitivity of the SNG production costs to the different input parameters. It can be concluded, that the uncertainty in specific investment costs has the largest influence on the uncertainty in SNG production costs, followed by the uncertainties in costs of biomass and of mineral oil. The latter can be explained by the coupling of hydrogen costs to mineral oil costs through the natural gas costs. The Tornado graph also shows whether the costs are positively or negatively related to a certain parameter. The mineral oil costs, for example, have a positive correlation to the SNG production costs, whereas the depreciation period is negatively correlated to the costs. In other words, higher mineral oil costs will increase the costs of SNG production, whereas a higher depreciation period will decrease the costs.

Beside the SNG production costs, also the potential of SNG production until 2020 and the SNG production in 2020 are presented in table 5.6 as well as in figure 5.9. The SNG production within one year (in this case 2020) represents the potential SNG production per year after that hydrogasification process is completely developed and implemented.

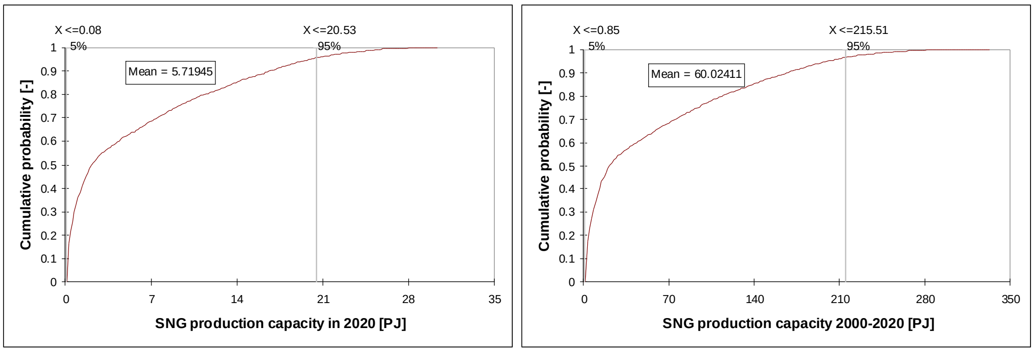

Figure 5.9 *Cumulative probability distributions of the potential SNG production until 2020 as well as the potential SNG production within one year (i.e. in 2020)*

With a potential SNG production capacity of 5.7 PJ a year, the hydrogasification process has the potential to fulfil approximately 2% of the Dutch renewable energy target in 2020, or ±7.5% of the biomass related target (see table 2.4). The SNG production capacity of 5.7 PJ complies with approximately half the current domestic demand for natural gas within the province of Groningen (see table 4.13). This is much lower than the SNG production capacity of pressurised O_2-blown gasification or indirect gasification, as described in §4.5.2.

5.7 Ecological analysis

By producing SNG a certain amount of CO_2 emission is avoided due to the replacement of natural gas by SNG. This amount of avoided CO_2 emission can be calculated from the amount of SNG produced. The specific environmental costs involved can be determined by relating the additional production costs to the avoided CO_2 emission. However, due to the application of hydrogen residue gases, normally used for e.g. firing purposes, these gases will have to be replaced by fossil-based fuel in its current application. As a result, the produced SNG will only be partly CO_2 neutral.

The expected costs per tonne CO_2 avoided are equal to €115 per tonne. The distributions of the costs, as well as the uncertainties on these costs are presented in figure 5.10. The figure shows that there is a 90% probability that the costs per tonne CO_2 avoided will be between €53 and €188 per tonne. The minimum and maximum costs are presented in table 5.7. Based on the potential SNG production until 2020 an expected potential reduction of CO_2 emission of 123 ktonne a year has been determined, as is also presented in table 5.7 and in figure 5.11.

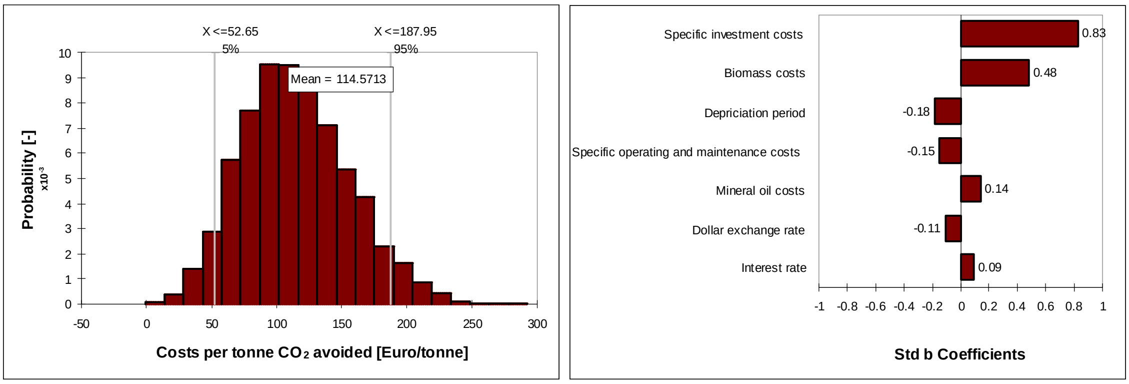

Figure 5.10 *Probability distribution as well as the Tornado graph of the sensitivity analysis of costs per tonne CO_2 avoided*

Table 5.7 *Potential CO_2 emission avoided until 2020 and costs per tonne CO_2 avoided in case of hydrogasification of willow*

		Minimum	Mean	Maximum
Costs per tonne CO_2 avoided	[€/tonne]	-0.8	115	292
Avoided CO_2 emission until 2020	[Mtonne]	0	1.3	7.6
Maximum avoided CO_2 emission a year	[ktonne/yr]	0	123	663

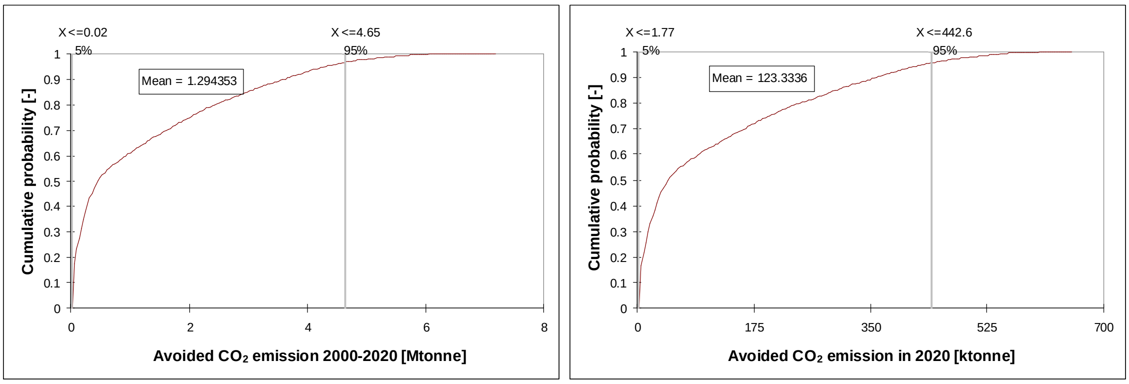

Figure 5.11 *Cumulative probability distributions of the CO_2 emission potentially avoided until 2020 as well as the potential CO_2 emission avoided within one year*

The Tornado graph in figure 5.10 shows the result of the sensitivity of the costs per tonne CO_2 avoided to the different input parameters. It can be concluded that, the uncertainties in specific investment costs and costs of biomass have the largest influence on the uncertainty of the costs per tonne CO_2 avoided.

The costs per tonne CO_2 avoided normally would increase in case the costs of hydrogen increase, however, would decrease if the natural gas costs increase (as the costs per tonne CO_2 avoided are determined by relating the additional production costs to the avoided CO_2 emissions). As hydrogen as well as natural gas costs are positively correlated to the mineral oil costs, an increase of mineral oil costs will on the one hand result in an increase of costs per tonne CO_2 avoided, due to an increasing cost of hydrogen. On the other hand, increasing mineral oil costs result in a decrease of the costs per tonne CO_2 avoided (due to an increase in natural gas costs). The influence of the uncertainty in mineral oil costs on the costs per tonne CO_2 avoided is, therefore, relatively small when compared with its influence on the SNG production costs.

5.8 Conclusions

The maximum amount of SNG that can be produced within one year is equal to 5.7 PJ in case only the application of hydrogen residue gases in the Netherlands is considered. The amount of CO_2 emission that can be avoided is equal to 123 ktonne a year. Considering the RD&D time, and the time required for market introduction, approximately 60 PJ of SNG can be produced until 2020, and 1.3 Mtonne of CO_2 emission can be avoided.

SNG production and specific environmental costs amount to €5.6 per GJ SNG and €115 per tonne CO_2 avoided.

Based on the utilisation of hydrogen residue gases in the Netherlands, the market for hydrogasification plants is limited and, as a result, the total amount of CO_2 emission that can be avoided relatively low. The application of non-residual hydrogen gases, whether or not sustainable, has not been considered within this study, however, will increase the potential number of plants, hence, increase the total amount of CO_2 emission that can be avoided[22]. Especially the application of sustainable hydrogen gases will have a major influence on the amount of CO_2 emission avoided, as the produced SNG can be considered (almost) CO_2 neutral. As a result, sustainable hydrogen, which involves higher hydrogen costs, might still be interesting, in case that specific environmental costs are reduced. Furthermore, higher hydrogen costs might be acceptable in case biomass is considered that normally, for example, is predestined for waste treatment. The application of non-residual (sustainable) hydrogen gases (for example hydrogen from the hydrogen grid, or internally produced hydrogen) has been investigated within the study on the potential of the biomass hydrogasification process in the Netherlands (Novem-NECST programme, project number 249.402-0190)[6].

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[22] however, at higher SNG production costs

6. SNG CO-PRODUCTION BY CONVERSION OF BIOMASS THROUGH FISCHER-TROPSCH / METHANATION PROCESS

6.1 Background

The Fischer-Tropsch (FT) process is used for the synthesis of hydrocarbons and, to a lesser extent, of aliphatic oxygenated compounds by the catalytic hydrogenation of carbon monoxide. The synthesis was discovered in 1923 by F. Fischer and H. Tropsch at the Kaiser Wilhelm Institute for Coal Research in Mülheim, Germany[66]. In the catalytic Fischer-Tropsch synthesis one mole of CO reacts with two moles of H_2 to form mainly aliphatic straight-chain hydrocarbons (C_xH_y). Typical FT catalysts are based on iron or cobalt. As with the methanation process (§4.2) about 20% of the chemical energy is released as heat in this exothermic reaction:

$$CO + 2\,H_2 \rightarrow \text{-}(CH_2)\text{-} + H_2O \qquad (6.1)$$

As follows from this equation, the FT reaction consumes hydrogen and carbon monoxide in a ratio of H_2/CO equal to 2. When the ratio in the feed gas is lower, it can be adjusted with the water-gas shift (WGS) reaction:

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad (6.2)$$

Iron-based FT catalysts show considerable WGS activity and the H_2/CO ratio is adjusted in the synthesis reactor. In the case of cobalt-based catalysts the ratio needs to be adjusted prior to FT synthesis. Typical operation conditions for FT synthesis, when aiming for long-chain products, are temperatures of 200-250°C and pressures of 25-60 bar[67]. The polymerisation-like chain-growth reaction results in a range of products, comprising light hydrocarbons (C_1 and C_2), LPG (C_3-C_4), naphtha (C_5-C_{11}), diesel (C_9-C_{20}), and wax ($> C_{20}$) fractions. The distribution of the products depends on the catalyst and the process operation conditions. With respect to the production of “green” diesel, process conditions can be selected to produce maximum amounts of products in the diesel-range. However, an even higher yield of diesel can be achieved when the FT synthesis is optimised towards production of wax. The wax can be selectively cracked to yield predominantly diesel. For this hydrocracking reaction additional hydrogen is required, which can be produced from a syngas side-stream that is completely shifted to hydrogen via the WGS reaction.

Following World War II, considerable research was conducted in the United States on the iron catalysts. One commercial plant was erected at Brownsville, Texas, in 1948, which used a fluidised bed of mill scale promoted by potash. Because synthetic oil was not competitive with petroleum, the plant was shut down within a few years. In the years since 1960, interest has grown in the United States in catalytic methanation to produce high-calorific gas. In the methanation step, carbon monoxide and carbon dioxide are converted to methane according to:

$$CO + 3\,H_2 \leftrightarrow CH_4 + H_2O \qquad (6.3)$$
$$CO_2 + 4\,H_2 \leftrightarrow CH_4 + 2\,H_2O \qquad (6.4)$$

The methanation process has been discussed extensively in chapter 4 (§4.2). The methanation reaction consumes hydrogen in a ratio of H_2/CO equal to approximately 3. When the ratio in the feed gas is lower, it can be adjusted with the water-gas shift reaction.

The syngas used within the FT and the methanation process can be obtained by biomass gasification. In 2002 a DEN-project has been started, in order to investigate the potential of the cogeneration of FT liquids and SNG, in comparison to stand-alone production of either FT liquids or SNG[72].

In case of cogeneration, a part of the thermal biomass input is converted to liquid fuels by once-through FT-synthesis. The off-gas of the FT-synthesis, containing non-converted CO and H_2, CH_4 produced during gasification, and gaseous hydrocarbons produced during the FT synthesis (C_1 to C_4), could be upgraded to the standard quality of natural gas by methanation and CO_2 removal. A simplified flowsheet of this (cogeneration) process, as well as the alternative stand-alone options, are presented in figure 6.1.

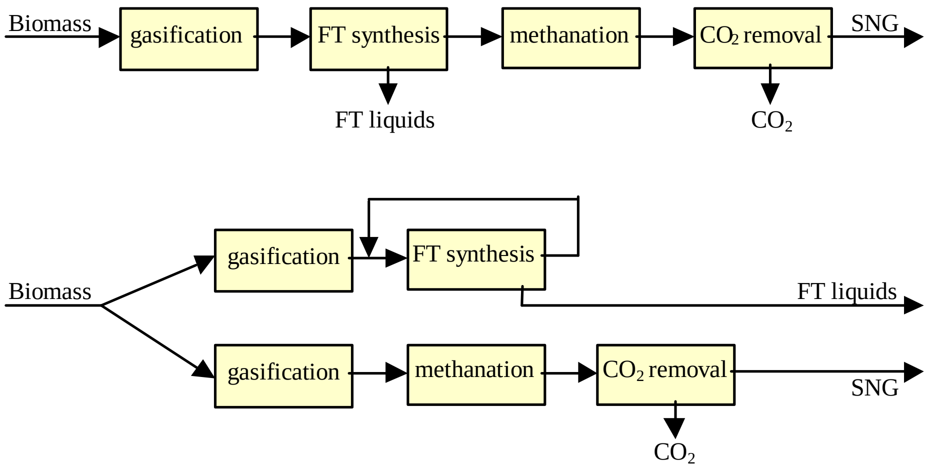

Figure 6.1 *Cogeneration as well as stand-alone production of FT liquids and SNG*

6.2 State of the art

Shell and Sasol are the only two companies that currently operate Fischer-Tropsch gas-to-liquids processes on a commercial scale. Several other corporations including Exxon Mobil, Energy International, Rentech, Statoil, and Syntroleum are currently developing catalysts and processes for gas-to-liquid conversion based on the Fischer-Tropsch process. While Shell operates on synthesis gas obtained from natural gas, to a host of middle-distillate products and waxes, Sasol uses synthesis gas obtained from coal gasification. Recent reviews[68][71] have shown that for the Fischer-Tropsch process to be economically viable, it needs to be operated on a large scale.

Three main types of reactors are either currently employed in industry, or in research studies on the improvement of the Fischer-Tropsch process. Multi-tubular fixed bed reactors have been used by Sasol, since the 1950s, and are currently being operated by Shell in its Middle Distillate Synthesis (SMDS) process at Bintulu in Malaysia[67][71], since 1993. Fluidised bed reactors are employed by Sasol and known as the Sasol Synthol process[67][71]. A slurry reactor has been developed by Exxon (now Exxon Mobil) at its R&D facility with an internal diameter of 1.2 m and a height of 21 m, while Sasol commissioned its internally cooled slurry reactor called the Sasol Slurry Bed Reactor (SSBR) in 1993. This reactor is of 5 m diameter and 22 m height. According to Sie and Krishna[71], the SSBR technology is considered by Sasol to be commercially proven and the design of a 10,000 barrel/day plant was considered.

6.3 System modelling

The cogeneration of Fischer-Tropsch liquids and SNG has been modelled within the software package MS-Excel, in accordance to the general flowsheet presented in figure 6.2. The selected process variables are five bio-syngas compositions from five gasifiers (§6.3.1), CO conversion in the FT-synthesis (§6.3.2), and the FT α-parameter (§6.3.2).

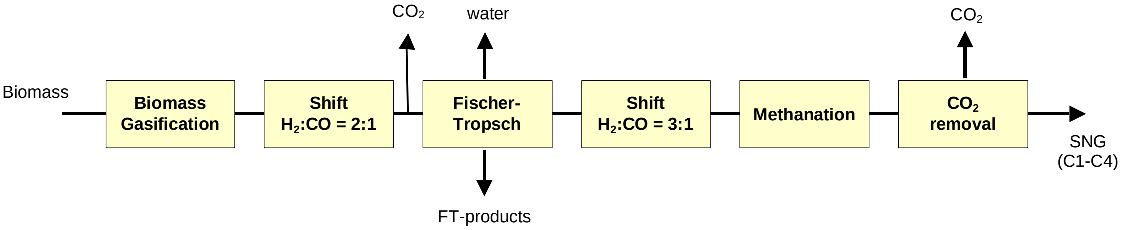

Figure 6.2 *General flowsheet of the cogeneration of FT-liquids and SNG from biomass*

6.3.1 Biomass gasification

The gasifiers that are considered within the modelling study are an indirect gasifier (Battelle), an oxygen-blown CFB gasifier (operated at either atmospheric conditions or at elevated pressure), an entrained flow gasifier and an atmospheric air-blown CFB gasifier. The different bio-syngas compositions for these five gasifiers, as well as their cold gas efficiencies, are presented in table 6.1. In order to be able to produce SNG, the gasifiers that operate at elevated pressure use CO_2 as pressurisation gas. Carbon dioxide, unlike nitrogen, can be removed quite easily from the product gas, hence enabling the adjustment of the quality of the product gas to the standard quality of natural gas.

Table 6.1 *Bio-syngas compositions for five different gasifiers*

		Battelle	atm.CFB (O_2)	pres.CFB (O_2)[23]		Entrained flow	atm.CFB (air)
CO	[vol.%]	26.3	18.1	10.5	10.5	38.4	16.6
H_2	[vol.%]	14.3	22.3	11.9	11.9	22.2	13.4
CO_2	[vol.%]	7.6	20.1	23.1	30.6	22.4	12.9
H_2O	[vol.%]	36.5	31.9	33.6	33.6	16.7	13.3
CH_4	[vol.%]	10.3	4.7	8.8	8.8	0.0	3.6
N_2/Ar	[vol.%]	0.0	0.5	8.0	0.5	0.3	38.3
C_2	[vol.%]	3.4	1.6	2.9	2.9	0.0	1.2
C_3	[vol.%]	0.0	0.0	0.0	0.0	0.0	0.0
C_4	[vol.%]	0.0	0.0	0.0	0.0	0.0	0.0
CGE[24]	[%]	80.4	80.6	79.1	79.1	77.9	79.1

6.3.2 Fischer-Tropsch synthesis

Equation 6.1 is the Fischer-Tropsch reaction, which leads to the formation of products, consisting mainly of paraffins (alkanes) and minor amounts of olefins (alkenes). The methylene (–(CH_2)–) group, shown in this equation, is used to represent the wide range of products formed during the Fischer-Tropsch reaction, which are actually of different chain lengths. The chain length distribution can be described by means of the Anderson-Schulz-Flory (ASF) equation, which is represented as[67]:

$$\log \frac{W_n}{n} = n \cdot \log \alpha + \log \frac{(1-\alpha)^2}{\alpha} \qquad (6.5)$$

where W_n is the mass fraction of a product, consisting of n carbon atoms, and α the chain growth probability factor. Higher values of α give more high molecular weight products. The value of α is characteristic of the particular catalyst employed in the Fischer-Tropsch process and, depending

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[23] The composition of the syngas from the pressurised oxygen-blown gasifier is presented twice, one resulting from the application of N_2 as compression gas, one resulting from CO_2 as compression gas.

[24] The Cold Gas Efficiency is defined as the thermal output of syngas divided by the thermal biomass input of the gasifier.

on the needs of a particular production process, catalysts can be tailored towards the production of predominantly low or higher molecular weight hydrocarbons. In practice, there is often a deviation from the ideal ASF distribution with regards to the lower hydrocarbon yields. C_1 yields are usually higher than predicted, whereas C_2 (as well as C_3 and C_4) yields are often lower than predicted, as shown in figure 6.3[69]. In order to correct for this deviation, the C_1 to C_4 yields are calculated according to modified equations[72], whereas the yields of the higher hydrocarbons (C_{5+}) are calculated according to the ASF distribution (equation 6.5).

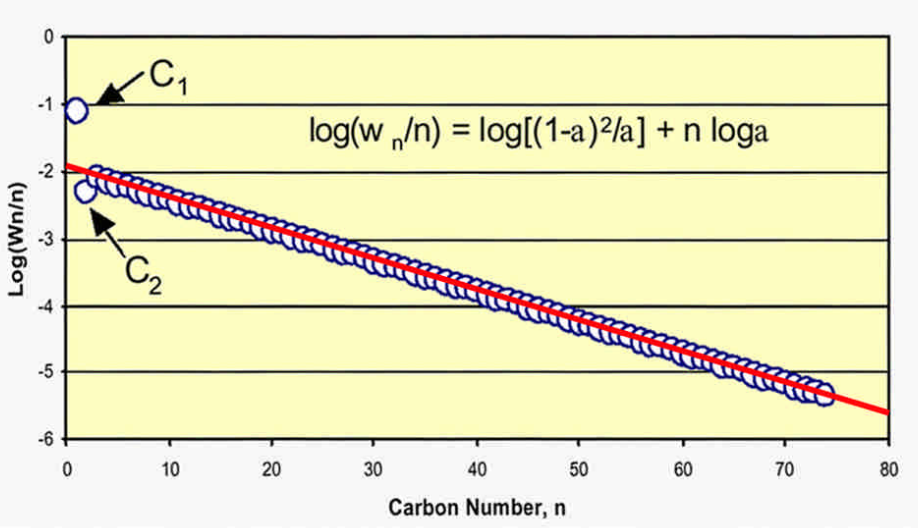

Figure 6.3 *ASF as well as experimental distribution of a FT product yields*

The Fischer-Tropsch synthesis is evaluated for α varying from 0.5 to 0.95 and a CO-conversion to Fischer-Tropsch (η_c) of 0 to 0.95. As during Fischer-Tropsch synthesis 1 mole of carbon monoxide reacts with approximately 2 moles of hydrogen (equation 6.1), the bio-syngas from the gasifier is shifted (equation 6.2) to a hydrogen-to-carbon-monoxide ratio of two, before entering the Fischer-Tropsch reactor.

6.3.3 Methanation process

In contrast to Fischer-Tropsch synthesis, methanation will require a hydrogen-to-carbon-monoxide ratio of approximately three (equation 6.3). As the conversion efficiencies from a given syngas to SNG and Fischer-Tropsch are almost equal (both approximately 80%), the conversion of CO to FT liquids affords a little bit higher overall efficiency as less energy is lost in the (exothermic) shift.

The methanation process has been modelled only according to equation 6.3; hence production of methane by the reaction of carbon dioxide with hydrogen has not been taken into consideration. During methanation all CO, remaining after the FT-synthesis, is converted to methane. Hydrocarbons, already present in the syngas, are assumed to behave as inert during methanation.

6.3.4 CO_2 removal

In order to produce an SNG with a quality comparable to that of standard Dutch natural gas, i.e. equal Wobbe-index, CO_2 removal takes place after methanation. The Wobbe-index is assumed to be equal to 43,7 MJ/Nm^3. The Wobbe-index of standard Dutch natural gas has a value between 43,5 and 44,4 MJ/Nm^3[7].

6.3.5 Technical feasibility of cogeneration of SNG and FT-liquids from biomass

As mentioned before, a DEN-project has been started in 2002, to investigate the potential of cogeneration of SNG and FT-products[72]. Within that study the overall production efficiency of SNG and FT-products, as well as their sole efficiencies have been determined as a function of the

CO-conversion and the α of the FT-synthesis for the five different gasifiers. The highest production efficiencies of SNG are obtained in case of gasification concepts with high amounts of hydrocarbons (see table 6.1), hence indirect gasification (Battelle) and pressurised oxygen-blown CFB gasification. Due to the high nitrogen fraction in the syngas of an air-blown CFB gasifier, or gasifiers using N_2 as pressurisation gas, these concepts of gasification are not suitable for SNG production and have, therefore, been left out of consideration.

The results of the indirect gasification and the pressurised O_2-blown gasification concepts are presented in figure 6.4 and figure 6.5, with some specific data in case of no FT-synthesis (η_c=0) and maximal FT-synthesis (η_c=0.95, α=0.95) presented in table 6.2. The efficiencies are defined as the thermal output of FT-product and/or SNG, divided by the thermal biomass input. A more comprehensive evaluation of the efficiencies of cogeneration for the different gasification concepts will be performed within the study on high-efficiency cogeneration of SNG and FT-products[72].

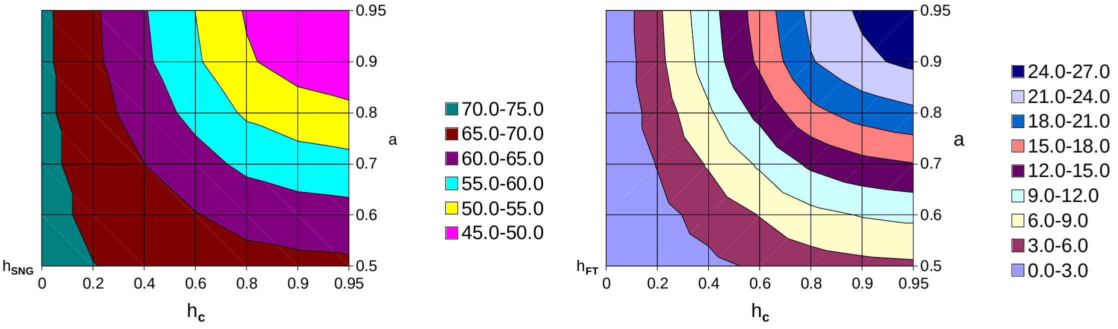

Figure 6.4 *Efficiencies in case of the concept of indirect gasification (Battelle)*

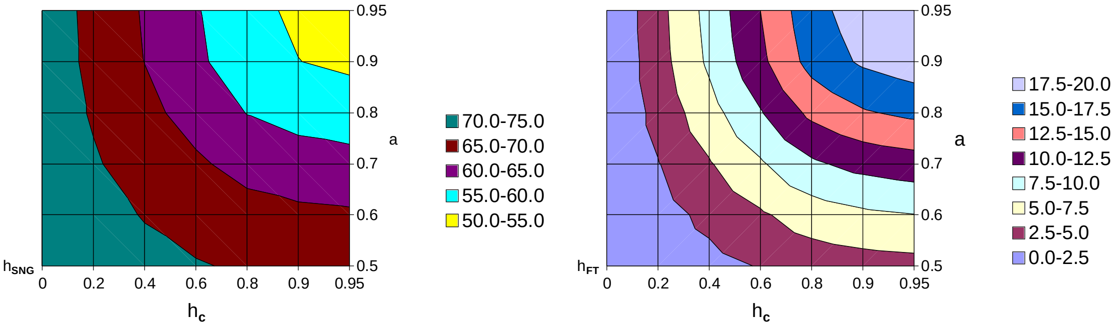

Figure 6.5 *Efficiencies in case of the concept of pressurised O_2-blown CFB gasification*

Table 6.2 *Specific efficiencies of the concepts of indirect gasification (Battelle) and O_2-blown CFB gasification*

	$\eta_{overall}$	η_{SNG}	$\eta_{FT\text{-}products}$
Indirect gasification (Battelle)			
η_c=0	71.2	71.2	0.0
α=0.95, η_c=0.95	71.4	45.5	25.8
O_2-blown CFB gasification			
η_c=0	72.8	72.8	0.0
α=0.95, η_c=0.95	73.0	53.2	19.8

6.4 Conclusions

Although the study on the feasibility of cogeneration of SNG and FT-products is not yet completed, some preliminary conclusions can be drawn. The sole efficiencies of SNG and FT-products show strong fluctuations with CO-conversion and α of the FT-synthesis, whereas the overall efficiency towards combined production of FT-products and SNG shows only a small deviation with CO-conversion and α. This is caused by almost equal conversion efficiency from a given syngas to SNG and FT-products.

From ***technical point of view*** the highest efficiency towards SNG is obtained in case of maximal methanation of the syngas (η_c=0) and the highest efficiency towards FT-products in case of maximal Fischer-Tropsch synthesis (η_c=0.95, α=0.95). Furthermore, pressurised oxygen-blown CFB gasification results in a higher efficiency towards SNG than indirect gasification, although the amount of hydrocarbons in the bio-syngas are higher in case of indirect gasification. The H_2-to-CO ratio within the bio-syngas of the indirect gasifier, however, is lower than in case of pressurised O_2-blown CFB gasification, as a result of which the energy losses during shift are higher. The advantage of a higher fraction of hydrocarbons is completely abrogated by the higher energy losses during shift.

As the assessment of ***economic and ecological feasibility*** has not been carried out yet, the formulation of a final conclusion on the feasibility of cogeneration of SNG and FT-products would be premature. The final evaluation of the technical as well as economic and ecological feasibility of cogeneration of SNG and FT-products will be completed in 2003[72].

7. COMPETITION ANALYSIS

Within this project the bio-(syn)gas being upgraded to SNG is either produced by a digestion process, or by a (hydro)gasification process.

7.1 SNG production by anaerobic digestion processes

As explained in chapter 3, anaerobic digestion is a proven technology being applied for small-scale decentralised conversion of “wet” organic residues at their origin. SNG production in this sector should always compete with the well-known combined heat and power application. The produced heat in a prime mover (at the moment mainly gas engines, in the future the more efficient fuel cell technologies) can be used optimally within the digestion process, and for other purposes such as space heating. Within this study, therefore, the production of SNG through anaerobic digestion has been considered as a reference case, and not as a competing route to SNG production by (hydro)gasification processes.

7.2 SNG production by gasification-based processes

The following gasification-based SNG production routes have been considered within this study:

- Pressurised oxygen-blown CFB gasification followed by methanation.
- Atmospheric indirect gasification followed by methanation.
- Pressurised BFB hydrogasification followed by methanation.
- Pressurised oxygen-blown CFB gasification followed by Fischer-Tropsch synthesis and methanation.
- Atmospheric indirect gasification followed by Fischer-Tropsch synthesis and methanation.

The main results of the technical, economic, and ecological analyses of the different gasification-based SNG production routes are summarised in table 7.1. Below each parameter, presented in table 7.1, will be discussed briefly.

7.2.1 Required time for RD&D and market introduction

The estimated expected RD&D time for pressurised oxygen-blown CFB gasification, as well as for hydrogasification is eight years. For indirect Battelle gasification due to the complicated heat exchange between the gasifier and the combustor, an additional RD&D time of two years (total RD&D time of 10 years) has been considered. For all the SNG production routes a period of four years is assumed to be necessary for the market introduction.

With the year 2000 being as starting point, it is therefore expected that the pressurised options (oxygen-blown CFB gasification and BFB hydrogasification) would be fully introduced in 2012, while fully introduction of the indirect gasification would be realised in 2014.

7.2.2 Efficiency SNG production

The energetic efficiency of SNG production by the Battelle process (67%) is slightly higher than in case of pressurised oxygen-blown CFB gasification (66.3%), due to the higher carbon conversion of the Battelle process (100% versus 93%).

On the other hand, the SNG production efficiencies for Battelle and pressurised oxygen-blown gasification, as calculated within chapter 6, are 4 to 6.5% points higher (71.2 and 72.8%),

Table 7.1 *Evaluation data for gasification-based SNG production routes*

		gasification/methanation		hydrogasification	FT-SNG	
		oxygen-blown	indirect		oxygen-blown	indirect
Assumed thermal input	[MW]					
biomass		100	100	50	10,000	10,000
hydrogen				47		
Required RD&D time	[yr]	8	10	8	8	10
Required time for market introduction	[yr]	4	4	4	4	4
Efficiency SNG production	[%]	66.3	67.0	79.1	72.8 / 53.2**	71.2 / 45.5**
Carbon conversion	[%]	93.3	100	80.1	93.3	100
Specific investment costs*	[€/kW_{th}]	482	449	616	*	*
SNG production costs	[€/GJ]	8.5	7.8	5.6	*	*
SNG production capacity until 2020	[PJ]	157	128	60	*	*
Maximum annual SNG production capacity	[PJ/yr]	14.9	15.1	5.7	*	*
Costs per tonne CO_2 avoided	[€/tonne]	95	83	115	*	*
Avoided CO_2 emission until 2020	[Mtonne]	8.8	7.2	1.3	*	*
Maximum annual avoided CO_2 emission	[Mtonne/yr]	0.84	0.84	0.12	*	*

* The final evaluation of the technical as well as economic and ecological feasibility of cogeneration of SNG and FT-products will be carried out within the Novem project “High efficiency cogeneration of "green" Substitute Natural Gas (SNG) and "green" transport fuels by the Fischer-Tropsch synthesis”[72].

** Efficiencies SNG production for stand-alone SNG, respectively for FT-SNG co-production options

*** Including oxygen plant

compared to efficiencies presented in chapter 4. This is due to the fact, that in chapter 6 the produced tars during the gasification (±4MW_{th}) are assumed to be separated from the product gas by OLGA process, before being recycled and converted within the gasifier with an efficiency of 95%. Taking this assumption into account for the cases considered in chapter 4, it would also be possible to achieve SNG production efficiencies above 70% for those cases.

In chapter 6, it is assumed that the H_2-to-CO ratio before methanation will be adjusted to 3 by the shift reaction. As this ratio in case of the Battelle process is lower (0.54, see table 6.1) than in case of oxygen-blown gasification (1.13, see table 6.1), the energy loss during the exothermic shift reaction will be higher for the Battelle process, resulting in a slightly lower energetic efficiency (71.2%), compared to oxygen-blown gasification (72.8%).

According to table 7.1, the highest energetic efficiency for SNG production has been achieved in case of the hydrogasification process (79.1%). This is due to the fact, that a high percentage of methanation reactions will already take place within the hydrogasifier which requires heat[25]. The much lower CO-content of the product gas from the hydrogasifier, compared to product gas from other gasifiers, requires a much smaller downstream methanation, resulting in less heat production outside the gasifier and, thus, higher SNG production efficiencies.

7.2.3 Carbon conversion

Highest carbon conversion can be achieved in case of indirect Battelle process, by combusting the produced char from the gasifier, in order to generate the required process heat. The lowest carbon conversion belongs to the hydrogasification process. Within this process, the rapid conversion of carbon-containing volatiles will be followed by the slow hydrogasification of char, resulting in the assumed maximum carbon conversion of 80% at 850°C. The oxygen-blown gasification with a carbon conversion above 93% lies between the Battelle and hydrogasification processes.

7.2.4 Specific investment costs

The specific investment costs for Battelle and oxygen-blown gasification processes are based on a biomass input of 100 MW, while in case of hydrogasification process it is based on a biomass input of 50 MW, due to the limited availability of the required hydrogen. This has resulted in the highest specific investment costs for hydrogasification process. The higher specific investment costs for oxygen-blown gasification, compared to the Battelle process, is mainly due to the requirement of an oxygen plant.

7.2.5 SNG production costs

Lowest SNG production costs can be achieved in case of hydrogasification process (5.6 €/GJ). Concerning the biomass gasification/methanation routes, the SNG production costs of the Battelle process are lower (7.8 €/GJ) than in case of pressurised oxygen-blown gasification (8.5 €/GJ), due to the lower investment costs, as well as the higher carbon conversion of the Battelle process (100% versus 93%).

In order to stimulate the demand for sustainable (green) energy, the Dutch government has taken some measures, by which the difference between the price of energy from fossil fuels and green energy will become smaller. One of the measures is, that for sustainable energy no ecotax (REB) has to be paid. For small consumers of natural gas (up to 5,000 Nm^3) the ecotax in 2002 amounted to 12.40 €ct/Nm^3[64] or 3.9 €/GJ[26]. Another stimulating measure is the so-called ecotax-transfer (REB-doorsluizing), which is an additional compensation for the producers/ suppliers of green gas. For 2002 the ecotax-transfer amounted to 5.79 €ct/Nm^3[64] or 1.8 €/GJ[24].

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[25] In oxygen-blown or indirect gasification a part of biomass carbon has to be combusted for heat supply to the gasifier.

[26] 1Nm^3 Groningen natural gas has a net calorific value of 31.669 MJ[70].

As shown in figure 7.1, the small consumers of gas will pay the same price for natural gas or green gas. In case of natural gas supply, the gas distribution companies will transfer the received ecotax from the consumers to the government. However, in case of green gas supply, the difference between the price of green gas and natural gas (equal to ecotax) will belong to the gas distribution companies.

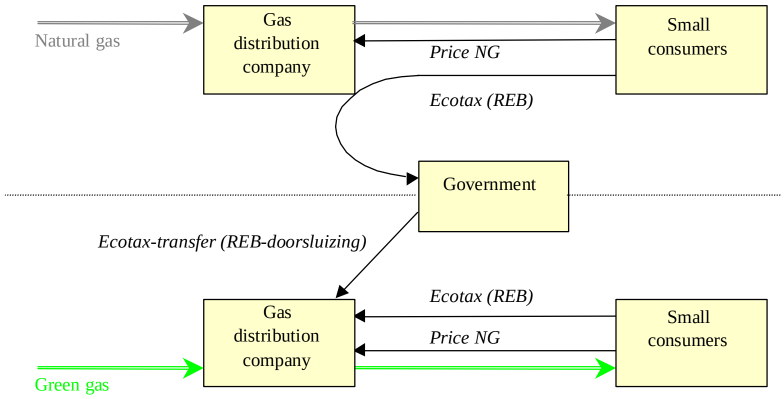

Figure 7.1 *Market price of green gas (SNG)*

Based on the above-mentioned measures, the market price for SNG is based on the natural gas price for large consumers (2.7-3.2 €/GJ[6]), plus the ecotax for small consumers (3.9 €/GJ), plus the ecotax-transfer (1.8 €/GJ). On this basis, the market price for green gas will be 8.5-8.9 €/GJ. For an SNG that is only partly green, the market price will be equal to:

Market price SNG = percentage green * (8.5 – 8.9 €/GJ) + (1- percentage green) * (2.7-3.2 €/GJ)

Table 7.2 presents the production cost, as well as the market price of SNG for different cases. As can be seen, although SNG production by biomass hydrogasification has resulted in the lowest SNG production costs (5.6 €/GJ), the market price for SNG from hydrogasification process is the lowest of all (4.9-5.4 €/GJ). This is due to the fact, that only a part of the produced SNG through this process is considered to be green (38.5% see table 5.2). For other two cases, the produced SNG will for 100% be green, resulting in promising market prices for both cases. At higher thermal inputs these cases will become even more promising. For hydrogasification, however, higher biomass inputs are not possible, due to the limited availability of the required hydrogen.

Table 7.2 *Production cost and market price of SNG for different cases*

		gasification/methanation		hydrogasification
		oxygen-blown	Indirect	
Assumed thermal input biomass hydrogen	[MW]	 100	 100	 50 47
Production cost	[€/GJ]	8.5	7.8	5.6
Market price	[€/GJ]	8.5 – 8.9	8.5 – 8.9	4.9 – 5.4

7.2.6 SNG production capacity until 2020

For oxygen-blown gasification the SNG production capacity until 2020 is 157 PJ, which is higher than the capacity for indirect gasification (128 PJ), as the latter requires a longer RD&D time. For hydrogasification process, again the limited availability of the applied hydrogen has resulted in the lowest capacity potential (60 PJ) until 2020.

7.2.7 Maximum annual SNG production capacity

The maximum annual SNG production capacity for indirect gasification is slightly higher than for oxygen-blown gasification (15.1 respectively 14.9 PJ), because of higher carbon conversion (100% versus 93.3%). The lowest annual SNG capacity with 5.7 PJ belongs to hydrogasification process.

With a potential SNG production capacity of approximately 15 PJ a year, SNG production by either pressurised O_2-blown gasification or Battelle gasification has the potential to fulfil approximately 5% of the Dutch renewable energy target in 2020, or even 20% of the biomass related target (see table 2.4). The SNG production capacity of 15 PJ complies with approximately 4.5% of the total domestic demand for natural gas in the Netherlands (see table 4.13). For hydrogasification option, the SNG production capacity of 5.7 PJ complies with approximately half the domestic demand for natural gas within the province of Groningen (see table 4.13).

7.2.8 Costs per tonne CO_2 avoided

Hydrogasification process results in the highest costs per tonne avoided CO_2 emissions (115 €/tonne). This is due to the fact, that the produced SNG through this process is only partly CO_2 neutral, and the amount of fossil-based hydrogen has to be taken into account.

For Battelle process lower costs per tonne avoided CO_2 emissions has been achieved (83 €/tonne), compared to oxygen-blown gasification (95 €/tonne). This is due to the lower SNG production costs and the higher SNG production efficiency of the Battelle process (see figure 4.9).

7.2.9 Avoided CO_2 emission until 2020

As a result of limited availability and the origin of the applied hydrogen in biomass hydrogasification process, only 1.3 Mtonne CO_2 emissions can be avoided until 2020. This is much less than the potential of the Battelle process (7.2 Mtonne) or oxygen-blown gasification route (8.8 Mtonne). A lower potential for Battelle, compared to oxygen-blown gasification, is based on the assumption that indirect gasification requires a somewhat longer development time than the pressurised oxygen-blown gasification.

7.2.10 Maximum annual avoided CO_2 emission

The maximum annual avoided CO_2 emission for both the indirect and the oxygen-blown gasification is with 0.84 Mtonne 7 times higher than in case of hydrogasification process (0.12 Mtonne).

7.3 Candidate SNG production processes

7.3.1 Gasification (pressurised oxygen-blown/atmospheric indirect) followed by Fischer-Tropsch synthesis and methanation

The overall energetic efficiency of the FT-SNG co-production options is practically equal to the energetic efficiency of the stand-alone SNG options (see table 6.2). Advantages of a co-production option over a stand-alone SNG option will therefore be completely determined by the economy of the considered options. Higher expected specific investment costs of a combined FT-SNG, compared to a stand-alone SNG, will probably result in higher SNG production costs. This would make a combined FT-SNG option less interesting than a stand-alone SNG option. However, as the assessment of economic and ecological feasibility of the cogeneration option has not been completed yet, the formulation of a final conclusion on the feasibility of this option would be premature. The final evaluation of the technical as well as economic and ecological feasibility of cogeneration of SNG and FT-products will be carried out within the Novem project “High efficiency cogeneration of "green" Substitute Natural Gas (SNG) and "green" transport fuels by the Fischer-Tropsch synthesis”[72].

7.3.2 Pressurised BFB hydrogasification followed by methanation

The energetic efficiency of SNG production by biomass hydrogasification is higher, and the production costs are lower than in the case of SNG production by biomass gasification/ methanation routes. However, the potential of the SNG production, as well as the avoided fossil-based CO_2 emissions until 2020 are much higher for biomass gasification / methanation routes, and the costs per tonne avoided CO_2 emissions are lower than in the case of SNG production by biomass hydrogasification. This is due to the limited availability (until 2020), as well as the origin (fossil-based) of the applied hydrogen. Fossil-based hydrogen lowers the market price for SNG from hydrogasification process, as only a part of the produced SNG can be considered green.

Based on these results it can be concluded, that production of SNG from hydrogasification of biomass is less interesting than upstream pressurised oxygen-blown or indirect gasification with downstream methanation.

In 2002 several hydrogasification experiments have been performed at Twente University[77]. These experiments have been carried out in a bench-scale pressurised fixed-bed gasifier, constructed by order of ECN, as a part of the technical feasibility of the hydrogasification process. Detailed information concerning the design and construction of this test facility is presented in appendix III.

Experiments were performed at T = 600, 800 and 850°C, P = 5, 10, 15, 20, 25 and 30 bar, using hard wood sawdust particles of 0.15 – 0.5 mm and 0.5 – 1.0 mm. The biomass / hydrogen ratio was 2.5 – 15 kg/kg. Residence time of biomass in the reactor, and of hydrogen in the hot reactor zone, was 0.4 – 50 min and 15 – 160 s respectively.

Based on the experimental results Kronberg[77], among others, has made the following conclusions, which also give cause to prefer upstream pressurised oxygen-blown or indirect gasification with downstream methanation above hydrogasification.

- The hydrogasification process cannot be operated in ‘once-through’ mode. Recirculation of hydrogen is required.
- The overall hydrogasification process is exothermal. However, the energy release is too low and insufficient for autothermal operation.

As mentioned in §5.3, recirculation of a part of hydrogen in the product gas from the hydrogasifier will result in higher costs and process complexity. However, in case that the process could not be operated autothermally, it would not be technically feasible. It should however be mentioned, that according to available literature there have been some processes which are based on hydrogasification of coal, biomass, or organic wastes[49][50][51][52][53]. Hydrogasification of brown coal with autothermal operation is even demonstrated in a 240 tonne/day plant in Germany[85].

7.3.3 Gasification (pressurised oxygen-blown/atmospheric indirect) followed by methanation

The energetic efficiency of SNG production by the Battelle process is higher and the SNG production costs and the costs per tonne avoided CO_2 emissions are lower than in case of pressurised oxygen-blown CFB gasification. This is, among others, due to lower investment costs and/or higher carbon conversion of the Battelle process.

On the other hand, the potential of the SNG production as well as the avoided fossil-based CO_2 emissions until 2020 are higher for pressurised oxygen-blown CFB gasification than in the case of SNG production through the Battelle process. The reason for this is the assumption that indirect gasification would require a somewhat longer development time than the pressurised oxygen-blown CFB gasification.

In other words after development, the indirect gasification scores better with respect to all aspects, compared to pressurised oxygen-blown CFB gasification, and therefore it is more promising for SNG production.

7.4 RD&D trajectory for candidate SNG production processes

In order to determine the possibility of the candidate processes for SNG production (indirect gasification / pressurised oxygen-blown gasification, followed by methanation) becoming a technical success, a diagram containing development stages, as well as possible problems has been constructed, as presented in figure 7.2.

The development of these processes is expected to take about 10 years for indirect gasification and 8 years for pressurised oxygen-blown gasification, and can be divided into three stages:
- determination of the Proof of Principle (PoP);
- determination of the Proof of Concept (PoC);
- demonstration of a complete integrated process on pilot scale.

Each development stage is subjected to uncertainties of certain parameters, for example of the problem of biomass feeding being solved, or of agglomeration becoming problematic, or of a successful upscaling. After the development stages, a period of four years is expected to be necessary for the market introduction.

7.4.1 Determination of the Proof of Principle

In the PoP-stage (this project) a gasification technology has to be selected for further development for green gas production. The preconditions which have to be taken into account in this stage are, among others, production of a tar-free, low-nitrogen, and high-hydrocarbon content synthesis gas, and the possibility of upscaling of the technology to a commercial scale for the long term.

Indirect gasification, producing a medium-calorific and almost nitrogen-free product gas with high concentrations of methane and C_2-fractions, offers very promising perspectives for, among others, SNG , power, or CHP production. The tar formed within this process, is expected to be removed from the gasifier product gas by the ECN OLGA process. The upscaling potential of this technology is expected to be less than the pressurised oxygen-blown gasification, due to the complicated heat exchange between the gasifier and the combustor. This makes the technology mainly suitable for decentralised SNG or CHP production. The fact that this technology does not require an oxygen plant is another positive aspect of this technology for decentralised applications.

Also pressurised oxygen-blown gasification results in a medium-calorific and either a low-nitrogen content (when using nitrogen as pressurisation gas), or almost a nitrogen-free (when using CO_2 as pressurisation gas) product gas with high concentrations of methane and C_{2+}

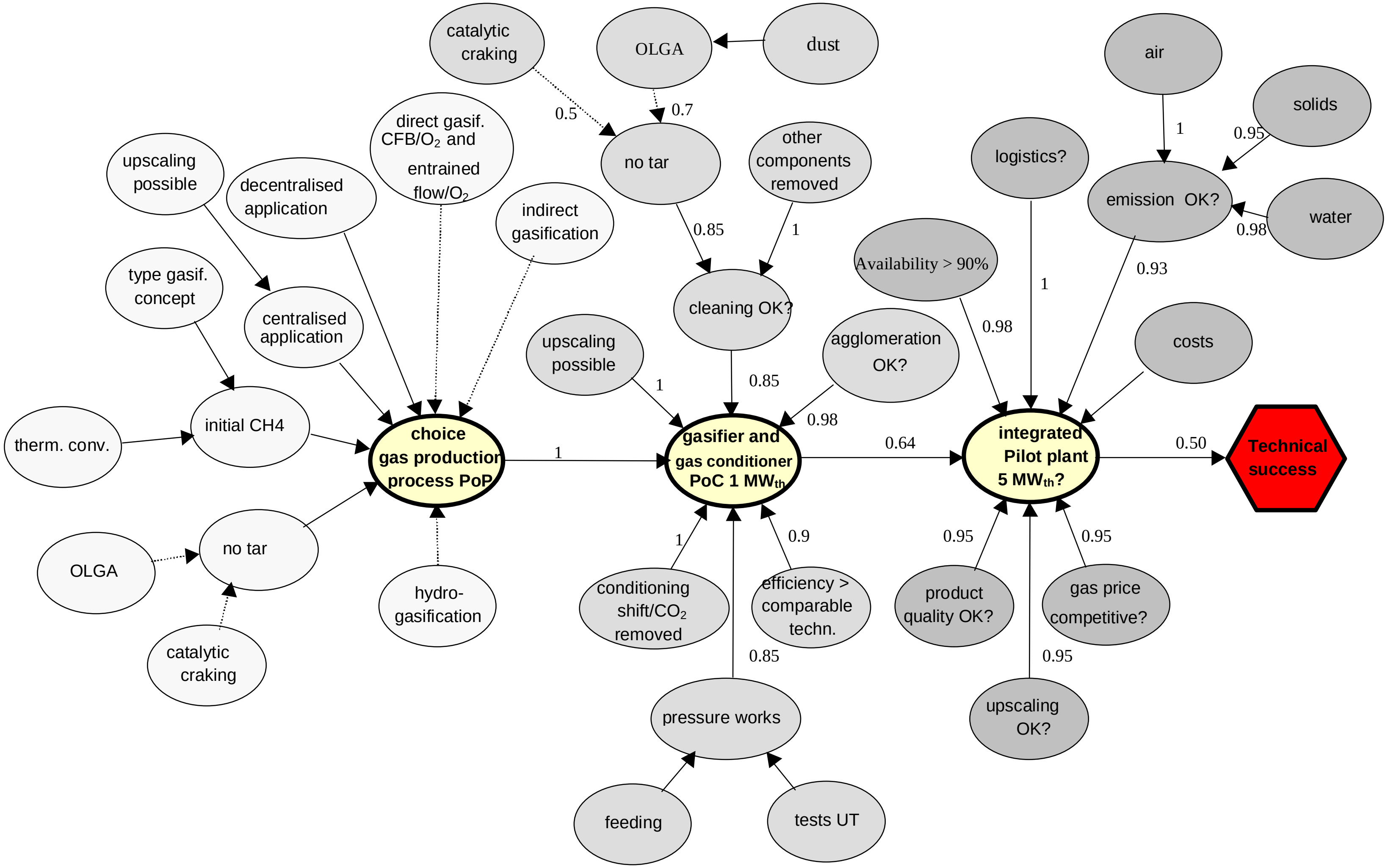

Figure 7.2 *Technical success diagram of processes for green gas (SNG) production*[87]

fractions. Therefore, this technology is also suitable for SNG production from biomass. Again the tar formed within this process, is expected to be removed from the gasifier product gas by the ECN OLGA process. The upscaling potential of this technology makes it suitable for centralised SNG production. Also the requirement of an oxygen plant, which can only be economic at large-scale applications, is another reason to consider the pressurised oxygen-blown gasification as a promising option for centralised SNG production.

7.4.2 Determination of the Proof of Concept

After determining the proof of principle, the proof of concept should be determined. Depending on the interest of the gasifier producers, it will be decided whether ECN independent, or in cooperation with a producer will go on with technology development. At the same time it will be determined, whether the technology development will be limited to gas clean-up and gas conditioning, or it will also include the gasifier development. It is of course preferred to come to a collaboration with a gasifier producer, whereby ECN would bring his knowledge in the technology development trajectory, and the gasifier producer would finally take care for the actual marketing of the jointly developed technology.

In the PoC-stage a bench-scale gasification/gas clean-up installation should be realised at ECN, with which a synthesis gas can be produced that satisfies the specifications for downstream methanation. An extensive R&D programme will be coupled to the realisation of this installation, mainly in the field of analysis of the “optimal” gasification conditions (among others agglomeration behaviour) and gas clean-up (removal of tar and other components).

The PoC-stage of SNG production will start in 2003. As the first step in realisation of a bench-scale gasification/gas clean-up installation, a lab-scale gasification facility is being constructed at ECN. This new test facility, the Milena (**M**ultipurpose **I**ntegrated **L**ab-unit for **E**xplorative and i**N**ovative **A**chievements in biomass gasification) has a thermal input of about 17 kW, corresponding to a biomass input of about 3.5 kg/h. The installation will be suitable for indirect gasification experiments. Several R&D aspects of indirect gasification such as agglomeration aspects, tar formation / tar reduction, and removal of other components can be studied in this test facility. At the same time operating of such a system will generate the required information for the assessment of the existing commercial indirect gasification processes like Battelle and Güssing. Moreover, the facility will also be suitable for agglomeration studies under oxygen-blown gasification conditions[65].

7.4.3 Demonstration of a complete integrated process on pilot scale

In the demonstration stage a complete integrated concept should be realised, that finally can be upscaled to commercial scale on the long term. The R&D aspects that should be analysed in this phase are, among others, the availability of the integrated concept, logistics aspects, ecological aspects (emissions to air and water, quality of solid residues), the quality and price of the product.

As the key technology development within the cluster polygeneration of the ECN business unit Biomass is the gasification / gas clean-up technology, the downstream methanation and gas conditioning of the syngas to SNG are not included within the technology development scope of this cluster. Therefore, for the final implementation of the complete integrated concept industrial collaborations will be required.

8. CONCLUSIONS & RECOMMENDATIONS

8.1 Conclusions

1. Although substitute natural gas (SNG) has not yet been incorporated as an alternative fuel in the Dutch and the European renewable energy policies, it can play an important role in realisation of the Dutch and the European climate and renewable energy targets. The produced SNG can most suitably be used for heat (and power) production in the domestic (households) sector, followed by decentralised CHP application in the industrial (and services) sectors. Besides, when the transport sector (at EU-level) would be so far to apply natural gas as an alternative transportation fuel, it should also be possible to use SNG as a substitute for natural gas within the transport sector.

2. Anaerobic digestion is a proven technology being applied for small-scale decentralised conversion of “wet” organic residues at their origin. SNG production in this sector should always compete with the well-known combined heat and power application. The produced heat in a prime mover (at the moment mainly gas engines, in the future the more efficient fuel cell technologies) can be used optimally within the digestion process, and for other purposes such as space heating. Within this study, therefore, the production of SNG through anaerobic digestion has been considered as a reference case, and not as a competing route to SNG production by (hydro)gasification processes.

3. The overall energetic efficiency of the FT-SNG co-production options is practically equal to the energetic efficiency of the stand-alone SNG options. Advantages of a co-production option over a stand-alone SNG option will therefore be completely determined by the economy of the considered options. Higher expected specific investment costs of a combined FT-SNG, compared to a stand-alone SNG, will probably result in higher SNG production costs. This would make a combined FT-SNG option less interesting than a stand-alone SNG option. The final evaluation of the technical as well as economic and ecological feasibility of cogeneration of SNG and FT-products will be carried out within the Novem project “High efficiency cogeneration of "green" Substitute Natural Gas (SNG) and "green" transport fuels by the Fischer-Tropsch synthesis”.

4. The energetic efficiency of SNG production by biomass hydrogasification is higher, and the production costs are lower than in the case of SNG production by biomass gasification/ methanation routes. However, the potential of the SNG production, as well as the avoided fossil-based CO_2 emissions until 2020 are much higher for biomass gasification / methanation routes, and the costs per tonne avoided CO_2 emissions are lower than in the case of SNG production by biomass hydrogasification. This is due to both the limited availability (until 2020), and the origin (fossil-based) of the applied hydrogen. Fossil-based hydrogen lowers the market price for SNG from the hydrogasification process, as only a part of the produced SNG can be considered green. Based on these results it can be concluded, that production of SNG from hydrogasification of biomass is less interesting than upstream pressurised oxygen-blown or indirect gasification with downstream methanation.

5. The energetic efficiency of SNG production by an indirect gasification process is higher, and the SNG production costs, and the costs per tonne avoided CO_2 emissions are lower than in case of pressurised oxygen-blown CFB gasification. This is, among others, due to lower investment costs and/or higher carbon conversion of an indirect gasification process. On the other hand, the potential of the SNG production as well as the total avoided fossil-based CO_2 emissions until 2020 are higher for pressurised oxygen-blown CFB gasification than in the case of SNG production through an indirect gasification process. The reason for

this is the assumption that indirect gasification would require a somewhat longer development time than the pressurised oxygen-blown CFB gasification. In other words after development, indirect gasification will score better with respect to all aspects, compared to pressurised oxygen-blown CFB gasification, and therefore it is more promising for SNG production.

6. The RD&D trajectory for candidate SNG production processes is expected to take about 10 years for indirect gasification and 8 years for pressurised oxygen-blown gasification, and can be divided into three stages:
   - determination of the Proof of Principle (PoP);
   - determination of the Proof of Concept (PoC);
   - demonstration of a complete integrated process on pilot scale.

   In the PoP-stage (this project) the indirect gasification technology has been selected for further development for green gas production. The preconditions which have been taken into account in this stage are, among others, production of a tar-free, low-nitrogen and high-hydrocarbon content synthesis gas, and the possibility of upscaling of the technology to a commercial scale on the long term.

   In the PoC-stage a bench-scale gasification / gas clean-up installation should be realised at ECN, with which a synthesis gas can be produced that satisfies the specifications for downstream methanation. An extensive R&D programme will be coupled to the realisation of this installation, mainly in the field of analysis of the “optimal” gasification conditions (among others agglomeration behaviour) and gas clean-up (removal of tar and other components). As the first step in realisation of a bench-scale gasification / gas clean-up installation, a lab-scale gasification facility is being constructed at ECN. This new test facility, the Milena (**M**ultipurpose **I**ntegrated **L**ab-unit for **E**xplorative and i**N**ovative **A**chievements in biomass gasification) has a thermal input of about 17 kW, corresponding to a biomass input of about 3.5 kg/h. The installation will be suitable for indirect gasification experiments. Moreover, the facility will also be suitable for agglomeration studies under oxygen-blown gasification conditions.

   In the demonstration stage a complete integrated concept should be realised, that finally can be upscaled to commercial scale on the long term. The R&D aspects that should be analysed in this phase are, among others, the availability of the integrated concept, logistics aspects, ecological aspects (emissions to air and water, quality of solid residues), the quality and price of the product.

   After the development stages, a period of four years is expected to be necessary for the market introduction.

8.2 Recommendations

1. According to a first estimation[87], the realisation of the PoC-stage of the RD&D trajectory will require a budget of about M€5.5. Main part of this amount should be financed by the industrial partners and different RD&D programmes (DEN/NEO/EU). This requires an intensive acquisition activity. For further technology development also joining international consortiums through EU-projects is recommended.

2. Contacts with developers / producers of indirect gasification technologies (Battelle and Güssing), as well as industries providing downstream methanation (among others Lurgi) and gas conditioning are recommended.

3. Within this study no attention has been paid to developments regarding the super critical biomass gasification (at about 600°C and 300 bar) for SNG production. It seems, however,

that high concentrations of methane and other hydrocarbons could be achieved in the product gas resulting from this process. Therefore, it is recommended to study the potential of super critical gasification for production of SNG from biomass; especially from “wet” raw organic materials.

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APPENDIX I: COMPOSITION SHEET OF WILLOW WOOD[41]

Results of: Willow
For: Gasification

component		mean value	max. value	min. value	standard deviation in %	number of references
water	wt%wet	15.0	43.5	2.39	80.7	19
C	wt%daf	49.7	51.0	48.2	1.83	19
H	wt%daf	6.06	6.74	5.83	3.18	19
O	wt%daf	43.6	45.0	41.6	2.27	19
N	wt%daf	0.63	1.44	0.10	49.0	19
S	wt%daf	0.06	0.13	0.03	47.3	14
Cl	wt%daf	0.01	0.03	0.00	52.0	14
F	wt%daf	0.00	0.01	0.00	157	4
Al	mg/kg dry	59.5	100	30.0	53.9	4
As	mg/kg dry	1.26	1.40	1.00	12.0	5
B	mg/kg dry	9.75	12.0	8.80	15.5	4
Ba	mg/kg dry	4.00				1
Ca	mg/kg dry	5720	7700	4000	23.4	5
Cd	mg/kg dry	2.35	3.00	1.70	39.1	2
Co	mg/kg dry	0.60	0.90	0.30	35.4	5
Cr	mg/kg dry	13.7	45.0	2.90	129	5
Cu	mg/kg dry	13.2	44.0	3.60	132	5
Fe	mg/kg dry	67.5	110	39.0	51.0	4
Hg	mg/kg dry	0.05				1
K	mg/kg dry	2894	4058	2000	27.4	7
Mg	mg/kg dry	524	700	360	25.1	5
Mn	mg/kg dry	9.68	13.0	7.90	23.2	5
Mo	mg/kg dry	0.50				1
Na	mg/kg dry	210	510	37.0	72.9	7
Ni	mg/kg dry	26.2	78.0	4.90	114	5
P	mg/kg dry	708	860	640	12.4	5
Pb	mg/kg dry	238	340	135	61.0	2
Sb	mg/kg dry	2.50	4.00	1.00	84.9	2
Se	mg/kg dry	1.00	1.00	1.00	0.00	2
Si	mg/kg dry	618	1800	88.0	115	5
Sn	mg/kg dry	1.65	3.00	0.30	116	2
Sr	mg/kg dry	14.0				1
Te	mg/kg dry	1.00				1
Ti	mg/kg dry	3.98	9.00	1.30	86.1	4
V	mg/kg dry	0.28	0.60	0.20	63.9	5
Zn	mg/kg dry	97.4	130	62.0	34.2	5
volatiles	wt%daf	83.1	86.1	80.3	2.24	18
ash	wt%dry	1.91	4.59	0.45	50.9	22
HHV	kJ/kg daf	19798	20868	18315	3.19	18
LHV calc.	kJ/kg daf	18489	19596	17011	3.35	18

Legend:
daf: dry and ash
HHV: higher heating
LHV: lower heating value

APPENDIX II: @RISK[73]

@RISK, a software system for the analysis of business and technical situations impacted by risk, brings the techniques of sensitivity and risk analysis to the industry's standard spreadsheet packages Microsoft Excel and Lotus 1-2-3. @RISK clarifies the impact of uncertain model parameters on the model results.

Used distributions:

Within the work described in this report two probability distributions functions have been implemented. The RiskPert function is used to specify a PERT distribution with a minimum and maximum value as specified. The shape parameter is calculated from the defined most likely value. As an example, the RiskPert function with minimum of 0, maximum of 10 and expected value of 2 is presented in figure II.1.

The RiskUniform function is used to specify a UNIFORM distribution with a minimum and maximum value. The probability of each value between minimum and maximum is the same, whereas the probability of values above the maximum and below the minimum is zero. As an example, the RiskUniform function with minimum of 5 and maximum of 10 is also presented in figure II.1.

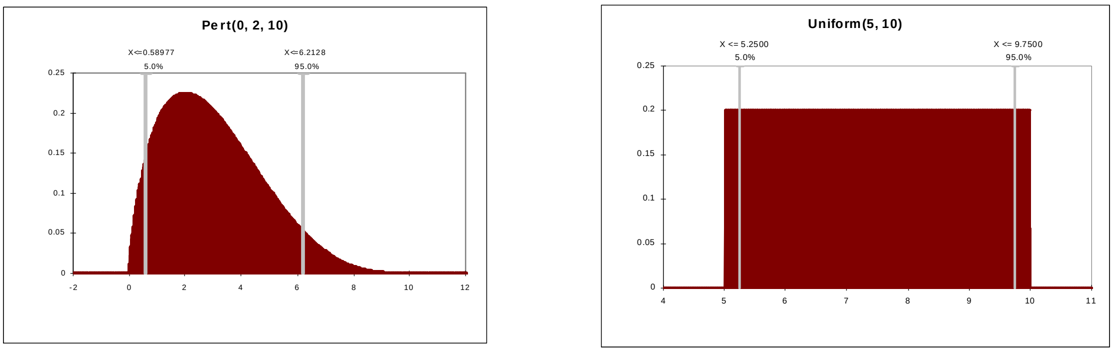

Figure II.1 *The RiskPert(0,2,10) and RiskUniform(5,10) functions as an example if Pert and Uniform distributed parameters*

Sensitivity analysis:

Based on the probability distributions of the model input parameters a quantitative measurement of the strength of the relationship between the model output and the model input parameters could be determined. Rank order correlation calculates the relationship between two data sets by comparing the rank of each value in a data set.

The rank order correlation value returned by @RISK can vary between -1 and 1. A value of 0 indicates there is no correlation between variables, they are independent. A value of 1 indicates a complete positive correlation between the two variables, a value of -1 a complete inverse correlation. Other correlation values indicate a partial correlation; the output is affected by changes in the selected input, but may be affected by other variables as well.

Risk analysis:

In a broad sense, Risk Analysis is any method – qualitative and/or quantitative – for assessing the impacts of risk on decision situations. The goal of any of these methods is to help the decision-maker choose a course of action, given a better understanding of the possible outcomes that could occur. In this study risk analyses are performed in order to obtain a probability distribution of the total SNG production costs for the different scenarios.

@RISK uses simulation, sometimes called Monte Carlo simulation, to do a risk analysis. Simulation in this sense refers to a method whereby the distribution of possible outcomes is generated by letting a computer recalculate the model over and over again, each time using different randomly selected sets of values of the input parameters. In effect, the computer is trying all valid combinations of the values of input variables to simulate all possible outcomes.

Risk analysis results in probability distributions of the model output parameter(s). In the cumulative curve, the point of 50% cumulative probability represents the median value of the output parameter.

APPENDIX III: DESIGN OF A BIOMASS HYDROGASIFICATION REACTOR AND THE EXPERIMENTAL SET UP[77]

DESIGN OF A HYDROGASIFICATION REACTOR

Pressurised fluidised bed reactor

Based on the available experience and recommendations, a continuous operating bubbling fluidized bed was originally chosen as a reactor for hydrogasification experiments. After extensive experimental and modeling study such a reactor was designed. Unfortunately, manufacturing of the reactor turned out to be impossible within the project, due to the technical difficulties and its high cost, as would be discussed below.

One of the most serious problems in the design of fluidised bed reactors emerges from the fact that the ratio of the biomass and gas volume flow rates is very high, especially at high pressures. This can be explained as follows. To have a good fluidisation, certain gas velocity is necessary. Therefore, if the pressure increases, the gas mass flow rate increases proportionally. In the hydrogasification process the mass flow rates of hydrogen and biomass are related and, consequently, increasing of pressure demands also proportional increase in the biomass flow rate, for a given volume flow rate of hydrogen. This makes biomass feeding in the form of a diluted suspension, which is necessary to prevent biomass agglomeration, difficult. Therefore, agglomeration and tube blockage could be expected.

On the other hand, manufacturing a fluidised bed reactor appeared to be too costly. The high cost was due to the severe operating conditions, use of hydrogen and technical problems with biomass feeding. In particular, the reactor must have rather thick wall and could only be made from a bar of very expensive steel. No tubes are available from which the reactor body with a large hole in the tube wall for biomass supply can be made; at $T = 800\text{-}850^{o}C$ and $P = 30$ bar the tube becomes unsafe, due to the hole. Feeding biomass through the reactor top or bottom can hardly be achieved. The required high flow rate of biomass is one of the reasons for that.

New hydrogasification reactor concept and design

To overcome the problems with the pressurised fluidised bed reactors several different reactor concepts had been considered, from which a co-current growing fixed bed reactor appeared to be the most suitable for the experiments. Figure III.1 presents a schematic drawing of the reactor. Biomass and hydrogen are supplied into the reactor through the top cover. The product gas is removed from the bottom. The produced char is continuously accumulated on the supporting porous plate. The principle of the reactor operation is explained in figure III.2.

The growing fixed-bed reactor has several advantages compared to the fluidised bed reactor. These advantages are:

- Feed flow rate can be arbitrary, whereas in the fluidised bed reactor it is dictated by the gas velocity required for a good fluidisation. This means that experiments could be done at a lower biomass flow rate. As a result the pressurised feeding system is significantly cheaper.
- A very wide range of the gas residence time can be studied by varying the feed flow rate.
- The biomass particle size can vary in a wide range. In the fluidised bed the particle size cannot exceed 0.5 mm, otherwise very high fluidisation velocities, and as a consequence high biomass flow rates and large reactor volume will be necessary. Thus the internal transport limitations could also be studied.
- There is no problem with char removal from the reactor. In the fluid bed the necessary entrainment rate of the char is difficult to control. Without removing of char the fluidised bed reactor will be overfilled in a few minutes, due to the very high biomass flow rate.
- Heat transfer in a fixed bed is rather poor, especially at low gas flow rates. Therefore, heat losses will be much less than in the fluidised bed reactor. Measuring of the temperature along the reactor axis provides information on the reaction heat effect.

- Since the bed height continuously increases during experiments, information on the entire history of the hydrogasification process conditions can be acquired from the same experimental run. In particular, the measurements of the product composition at different moments of time or for different bed heights is equivalent to the measurements of the concentration distribution over the height of a co-current fixed-bed reactor. This is further explained in figure III.2.

The disadvantage of the growing fixed-bed reactor, compared to the fluidised bed reactor, is that conditions in the reactor change along the height. As a result interpretation of the experiments in quantitative terms is more difficult than for most of the fluidised bed reactors.

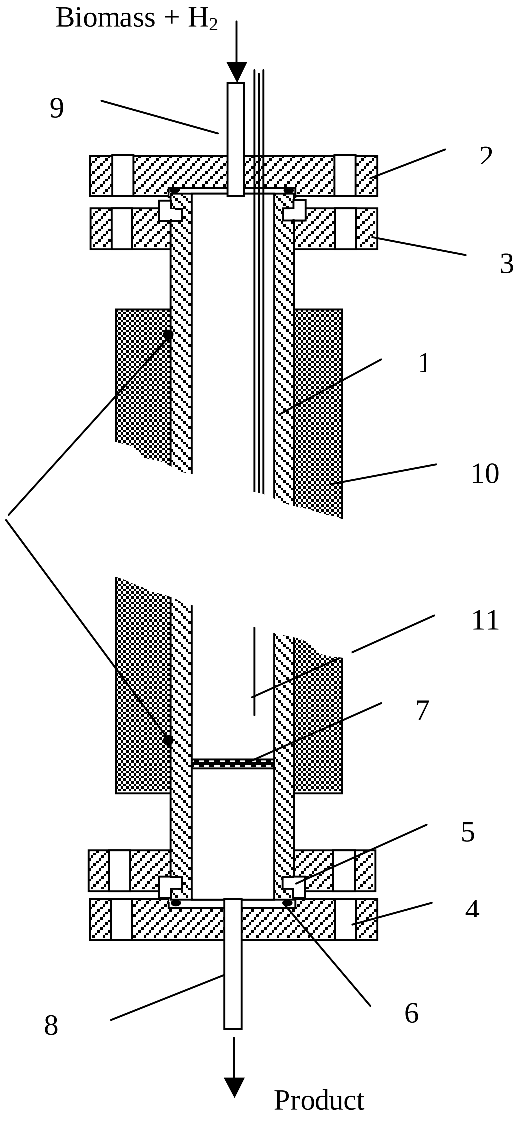

Figure III.1 *Co-current growing fixed-bed reactor (1: reactor tube; 2, 3: flanges; 4: hole for bolt; 5: half-moon positioning ring; 6: sealing ring; 7: supporting perforated and porous plates; 8: product outlet tube; 9: hydrogen and biomass feeding tube; 10: heater; 11: 3 thermocouples inside the reactor; 12: thermocouples at the reactor wall)*

The main reactor characteristics are:
- Material of the reactor tube: Incoloy Allow 800HT.
- Material of the flanges: stainless steel AISI 316.
- Reactor tube 1: length 700 mm, outside diameter 48.26 mm; inside diameter 33.98 mm.

- Tubes 8 and 9: outside diameter 13.72 mm; inside diameter 7.7 mm.
- Height of the heating section: 400 mm.
- Maximum temperature of the heated part of the reactor 1000^{o}C at pressure of 40 bar.
- Maximum pressure 40 bar at temperature of 1000^{o}C.

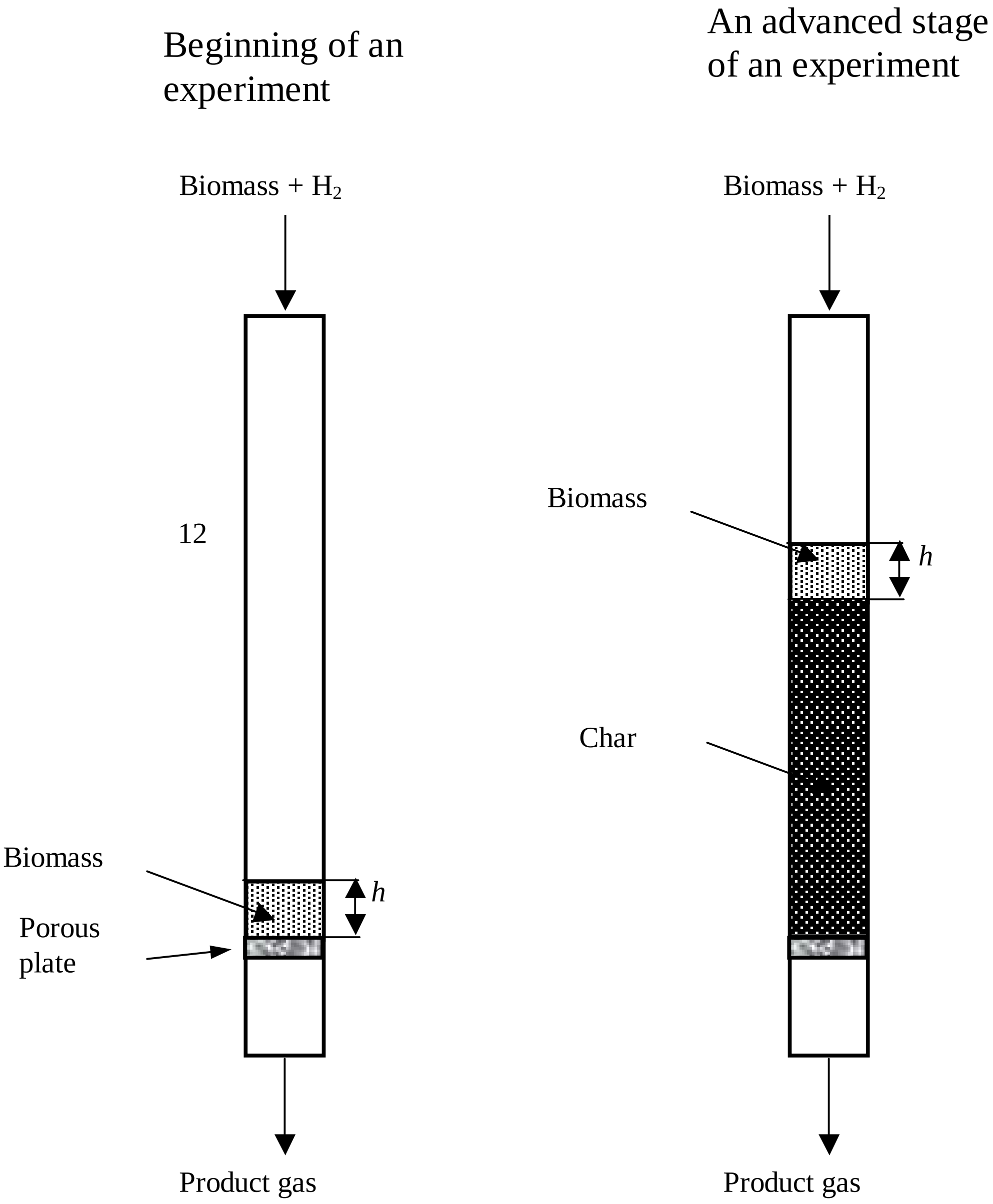

Figure III.2 *Operating principle of the co-current growing fixed-bed reactor. During experiments the bed height grows steadily by introducing fresh material through the top. The upper layer of the bed remains the same, whereas the lower part shows more advanced stage of the reaction. The growing fixed-bed reactor operates similar to the co-current moving-bed industrial gasifier. The difference is that the moving-bed gasifier operates at steady state. This is achieved by removing the solid phase from the reactor bottom at a required flow rate. If the longitudinal mixing can be neglected, product composition at the outlet of the growing fixed-bed reactor should be the same as gas composition in the co-current moving-bed reactor at the distance from the top of the reactor, equal to the height of the growing fixed bed.*

The flanges of the reactor were connected to the reactor tube without welding and were not heated during the experiments in order to prevent hydrogen leakage. Only the central part of the reactor of 400 mm height was heated by an electrical heater.

Three thermocouples were inserted into the reactor through the top cover using a Conax Buffalo sealing gland, which is suitable for temperatures up to 870°C and pressures up to 690 bar. Positions of the thermocouples in the reactor are shown in figure III.3.

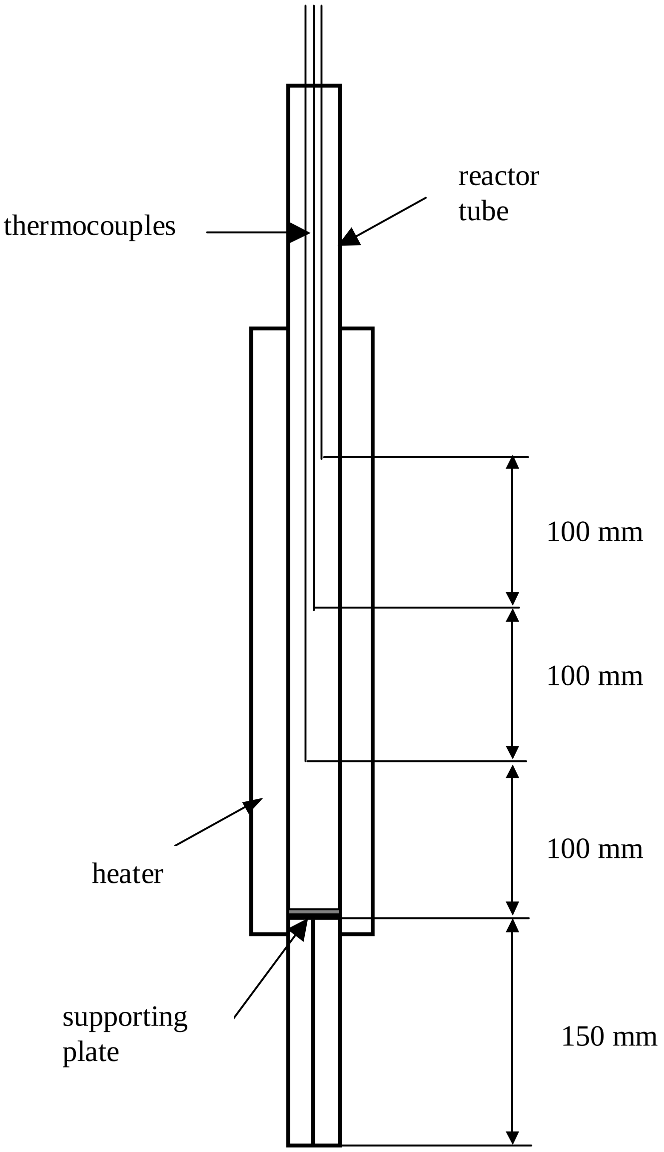

Figure III.3 *Position of the thermocouples in the reactor*

Two thermocouples were connected to the outer surface of the reactor tube near the top and bottom of the heated section, see figure III.1.

The supporting plate (thickness: 3 mm, free area: 5 %, diameter of holes: 1 mm) was made from the same material as the reactor tube. To prevent char withdrawal from the reactor, a glass porous disk (thickness: 3 mm, pore size: 10 µm) was placed on the supporting plate and sealed along the circumference.

Reactor operating parameters

The available experimental data on gasification of wood at 30 bar and 750-950°C with a hydrogen-rich gas[56] shows, that most of the biomass conversion (about 80% on weight basis) occurs during the rapid-reaction stage (devolatilisation and pyrolysis reaction of volatile matter in biomass with hydrogen) in less than 0.2-0.3 min. After that a slow reaction of residual carbon with hydrogen takes place. The biomass conversion increases from 85 to 90% when gasification time extends from 20 to 150 min. These data indicate the characteristic time scale of the hydrogasification experiments and the residence times of hydrogen and biomass in the reactor.

The hydrogen flow rates were chosen so that the residence time of hydrogen in the heated reactor section was in the range of 10-200 s. Higher flow rates would lead to additional difficulties due to:

- heating of the feed;
- excessive pressure drop (several bars) over the bed of biomass and char and over the supporting glass porous plate;
- large volume of a pressurised biomass storage vessel and complicated biomass feeding system.

The biomass flow rate of approximately 1 kg per 75-100 mole of hydrogen was taken according to the calculations of the conditions at which the reactor operation in autothermal mode seems possible[5].

Heating of biomass and hydrogen in the reactor can easily be calculated. The Reynolds number of the hydrogen stream in the reactor is:

$$\mathrm{Re} = \frac{\Phi_{H_2} d_r}{S_r \mu(T)} \qquad \text{(III.1)}$$

where Φ_{H2} is the mass flow rate of hydrogen, $\mu(T)$ is hydrogen viscosity, d_r is inner diameter of the reactor, and S_r is cross sectional area of the reactor.

At the maximum hydrogen flow rate of about 1 g/min ($1.667 \cdot 10^{-3}$ kg/s), the Reynolds number is less than 30. Therefore, the hydrogen flow is laminar. Assuming constant temperature of the reactor wall (T_w), the average temperature (T) of hydrogen-biomass stream at distance x from the inlet can be calculated as:

$$\theta = \frac{T_w - T}{T_w - T_{in}} = 4\sum_{n=1}^{\infty} \frac{1}{\lambda_n^2} \exp\left(-4\lambda_n^2 \frac{x}{d_r} \frac{1}{\mathrm{Re}\,\mathrm{Pr}} \right) \qquad \text{(III.2)}$$

where T_{in} is the inlet temperature, $\mathrm{Pr} = \mu(T)C(T)/\lambda(T)$ is the Prandtl number, C(T) is the mass heat capacity of hydrogen-biomass mixture, $\lambda(T)$ is the thermal conductivity of hydrogen and λ_n is the n^th^ root of the equation:

$$J_0(\lambda) = 0 \qquad \text{(III.3)}$$

where J_0 is the Bessel function of the first kind.

The presence of biomass in the hydrogen stream is taken into account via the heat capacity of the stream, because heating of the biomass particles occurs very quickly.

Figure III.4 shows the dimensionless temperature (θ), calculated for typical operating conditions as a function of the distance (in meters) from the inlet. At a distance of about 0.2 m from the inlet, the temperature of the biomass-hydrogen stream is almost equal to the wall temperature. Therefore, for the chosen flow rates the feed will be heated very quickly in the reactor, and heating of hydrogen before the reactor is not necessary.

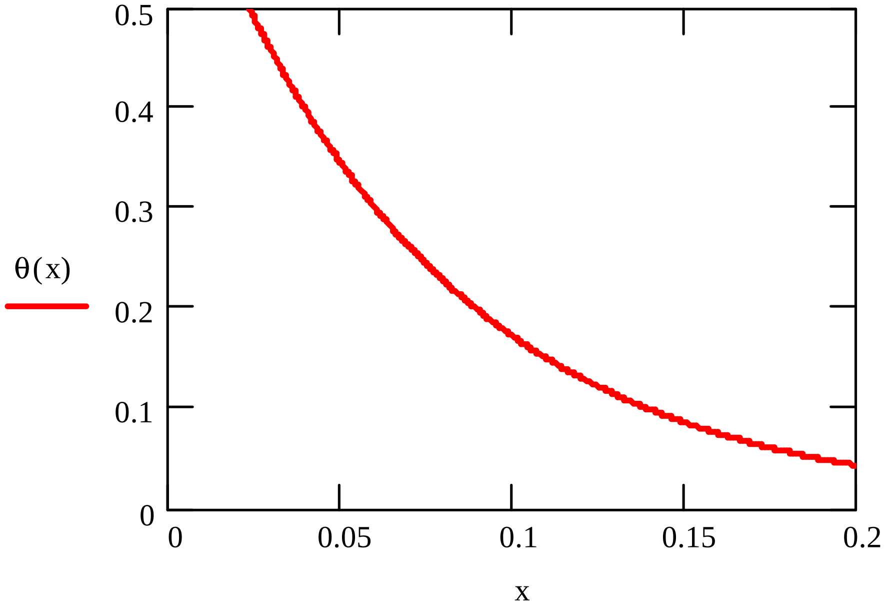

Figure III.4 *Dimensionless temperature (equation III.2) of the biomass-hydrogen stream as a function of the distance x (m) from the reactor inlet (hydrogen flow rate 8.12 Nl/min, biomass/hydrogen ratio 8 kg/kg)*

Estimates of the power, required for heating of hydrogen and biomass in the reactor, for evaporation of water contained in biomass, and for compensation of energy losses to surroundings and the reactor flanges showed, that a heater of 2 kW would be sufficient. In the experiments a heater of 3.8 kW was used.

In order to check whether biomass gasification is limited by the heat transfer, calculations of the biomass particle heating were made. These calculations show, that the heat transport resistances should not influence the gasification of the biomass particles less than 1 mm. The thermal equilibration time is about 0.2 s.

Biomass feeding system

Several different systems for biomass feeding into the pressurised reactor were considered. Finally a screw feeder was chosen. Its drawing is shown in figure III.5. Biomass is stored in a pressurised bunker (inner diameter: 106 mm, height: 200 mm). The biomass is transported by a screw. The transport of biomass from the bunker is supported by a small stream of pressurised hydrogen, supplied through the top cover of the feeder. The biomass transport in the feed supplying tube is facilitated by hydrogen stream, going to the reactor. To avoid biomass bridging in the bunker, the feeder was equipped with a rotating wheel, driven by the screw. The feeder was designed for the maximum pressure of 60 bar. With the used screw and motor, the biomass flow rate was in the range of 0.5-5.0 g/min. The volume of the bunker was about 1.5 l. About 400 g of wood saw dust could be stored in the bunker. Thus at the supplied biomass flow rates of about 2-3 g/min during the experiments, the feeder could continuously operate for several hours.

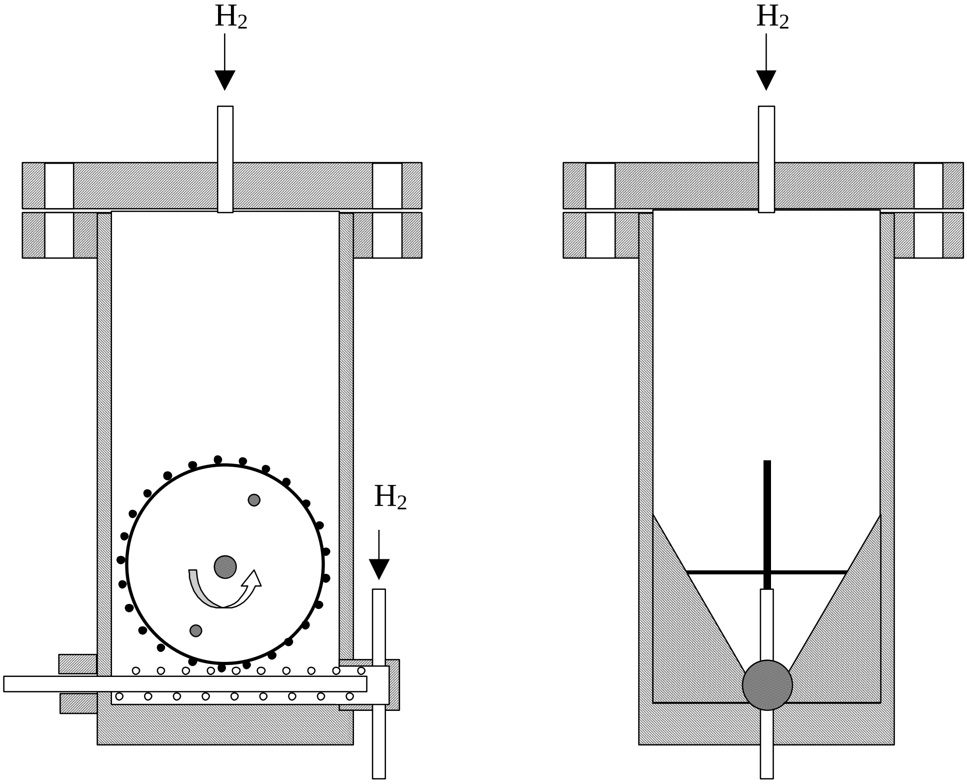

Figure III.5 *Biomass feeder (views from the two sides)*

Experimental set up

Figure III.6 shows the flowsheet of the designed and built experimental set up. The main components of the set up are: reactor, biomass feeder, heater, gas cooler, and tar trap. Complete experimental set up also included a gas product analysing system for methane, carbon monoxide, carbon dioxide, hydrogen and oxygen. For safety reasons the set up was placed inside a concrete bunker and operated from the outside. The photographs of the experimental set up and its main constituents are given in figures III.7 to III.10.

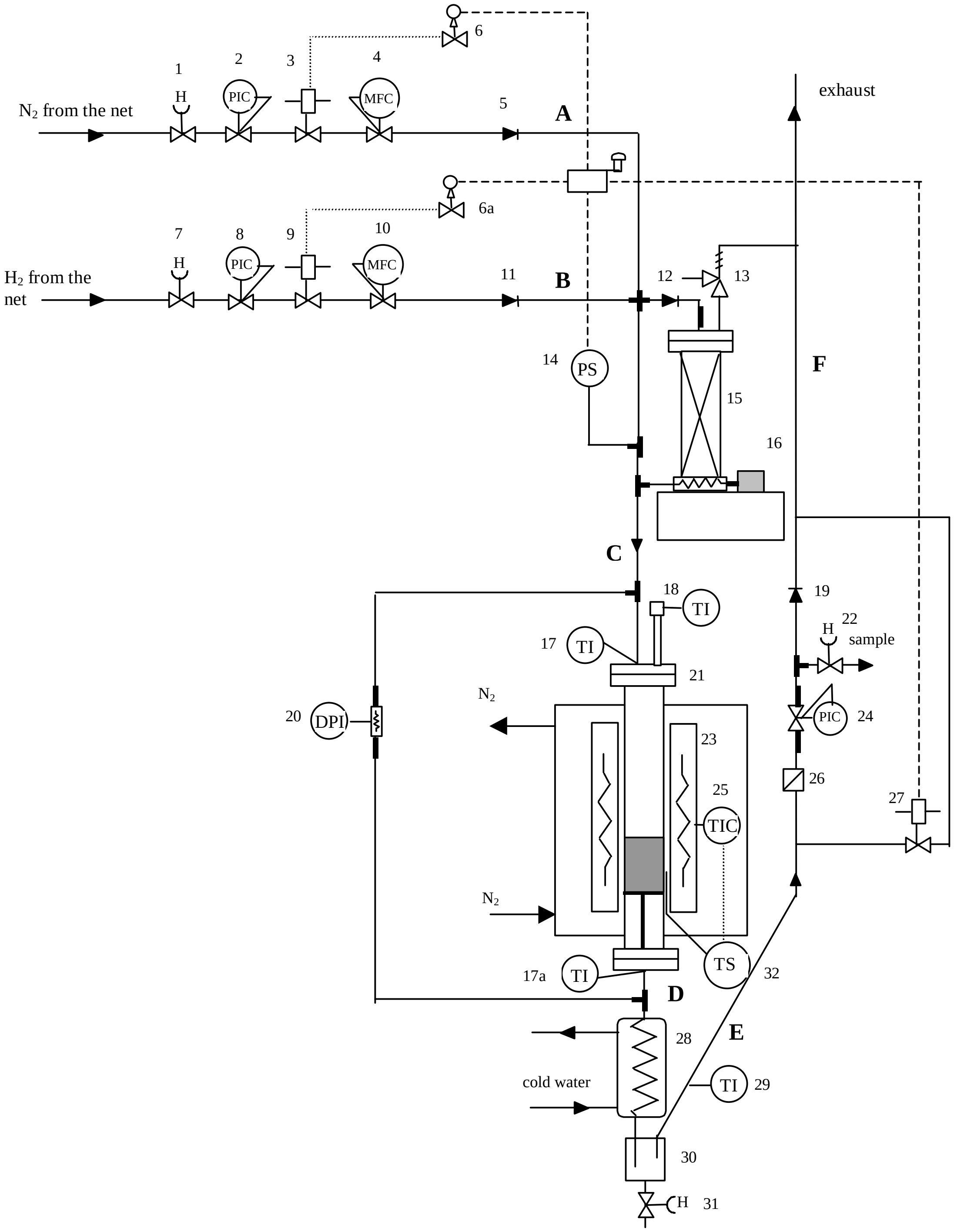

Figure III.6 *Flowsheet of the experimental set up*

List of the equipment

1, 7, 22, 31	ball valve, 2-way
2, 8	pressure reduction valve
3, 9	magnet valve
4, 10	mass flow controller
5, 11, 12, 19	check valve
6, 6a	pneumatic valve
13	safety valve
14	pressure transmitter
15	biomass storage vessel with screw feeding
16	electrical motor
17, 17a, 18, 29	temperature transmitter (thermocouple)
20	differential pressure meter
21	reactor
23	electrical heater
24	back pressure valve
25	heater temperature control
26	filter
27	solenoid valve
28	water cooler
30	tar trap
32	temperature switch

Mass- and energy streams

Stream	Medium	Max. pressure bar	Temperature ^oC	Piping[*]	Max. flow rate Nl/min
A	nitrogen, N_2	10	ambient	RVS, 10, 2	100
B	hydrogen, H_2	40	ambient	RVS, 10, 2	10
C	hydrogen, Biomass (wood)	40	ambient	RVS, 10, 2 and flexible hose, 10, 2	10 3.8 g/min
D[**]	H_2 : 24.4 vol.% CH_4 : 35.2 vol.% H_2O : 29.8 vol.% CO_2 : 7.1 vol. % CO : 3.1 vol.% N_2 : 0.4 vol.%	40	350	RVS, 10, 2	6
E	H_2 : 24.4 vol.% CH_4 : 35.2 vol.% H_2O : 29.8 vol.% CO_2 : 7.1 vol. % CO : 3.1 vol.% N_2 : 0.4 vol.%	40	30	RVS, 10, 2	6
F	Idem E	0	ambient	RVS, 10, 2	6

[*] Material, outer diameter (mm), wall thickness (mm)

[**] according to calculations of ECN

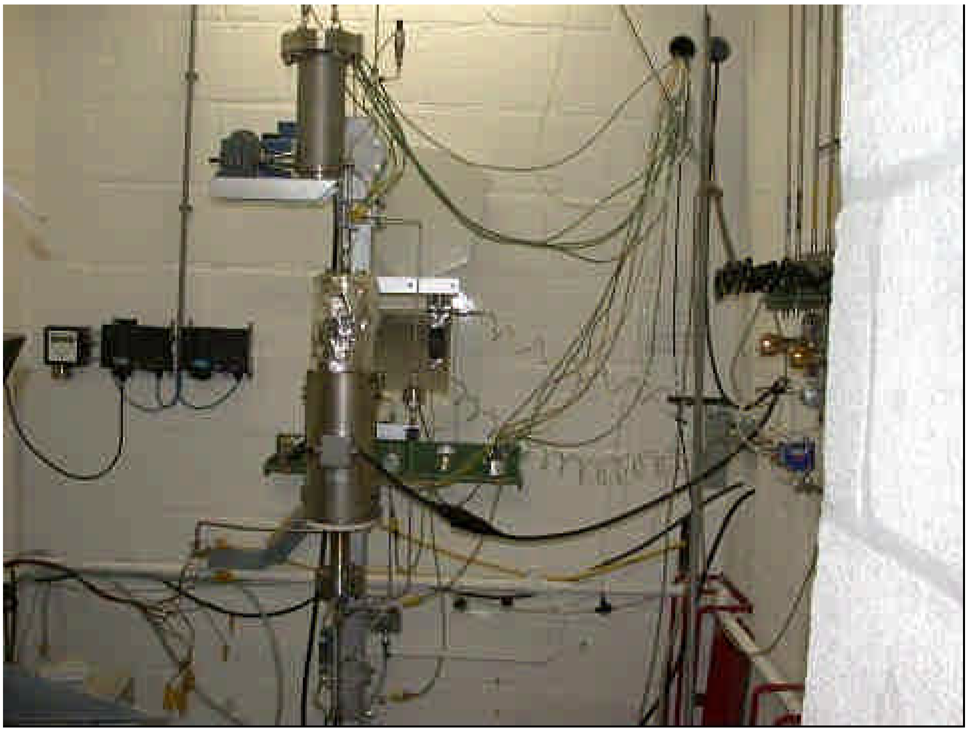

Figure III.7 *Experimental set up in the bunker*

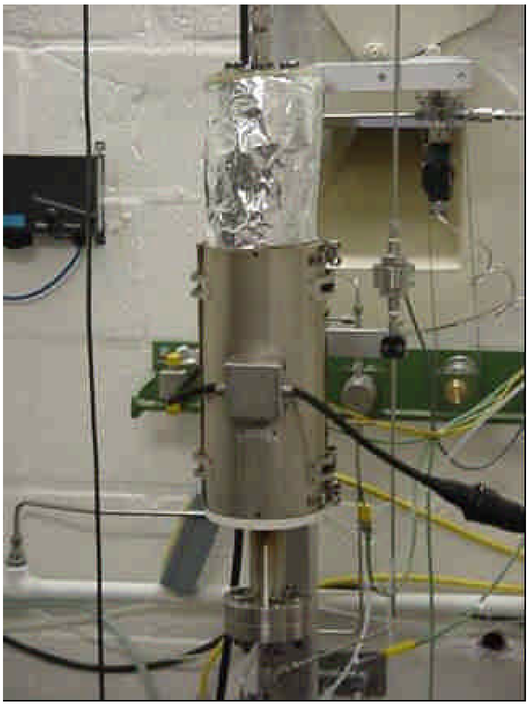

Figure III.8 *Hydrogasification reactor with the electrical heater. Upper part of the reactor is insulated to decrease heat losses.*

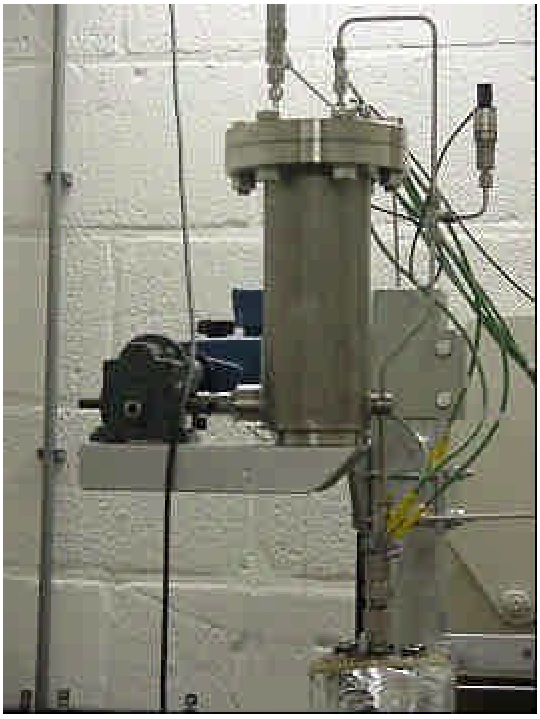

Figure III.9 *Biomass feeder*

Figure III.10 *Product gas cooler and tar trap*