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High efficiency co-production of Fischer-Tropsch (FT) transportation fuels and Substitute Natural Gas (SNG) from biomass

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Justification

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Abstract

The technical, economic, and ecological feasibility was studied of the co-production of 50 PJ of Fischer-Tropsch (FT) transportation liquids and 150 PJ of substitute natural gas (SNG) per year (*i.e.* 10% of the 2001 Dutch consumption). In the co-production concepts part of the SNG is produced by methanation of the FT off-gas, which already contains significant amounts of C_1-C_4 SNG compounds. The additional required SNG is produced by dedicated methanation of part of the gasification product gas. Co-production results in higher biomass-to-fuel efficiencies, lower biomass input requirements, and less negative net present values (NPVs) compared to the case of complete separate production of both fuels. Co-production concepts based on pressurised oxygen-blown and indirect gasification result in the lowest CO_2 emission reduction costs of about 100 €/tonne. Co-production of “green” Fischer-Tropsch transportation fuels and “green” SNG will be an economic feasible process in the Netherlands, when both energy carriers receive the same tax exemptions as currently is given to green electricity or at crude mineral oil prices above 56 $/bbl.

Keywords

Biomass, Gasification, Fischer-Tropsch synthesis, Methanation, Transportation fuels, Substitute Natural Gas (SNG), Co-production, Probability analysis, Technical analysis, Economic analysis, Ecological analysis, Entrained flow gasification, Indirect gasification, circulating fluidised bed (CFB) gasification.

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EXECUTIVE SUMMARY

Introduction

Biomass is one of the most promising renewable energy sources to replace fossil fuels and has the unique characteristic that it is a feedstock for the production of chemicals as well as gaseous and liquid fuels. In the Dutch situation, especially natural gas and transportation fuels are important products with a total current consumption of approximately 2000 PJ/year. Production of "green" substitute natural gas (SNG) and transportation fuels will allow the use of the existing natural gas and transportation fuels infrastructure and, hence, make a gradual transition from the present fossil fuel-based energy supply system to a complete biomass-based economy possible. Gasification of biomass generates a product gas or synthesis gas that can be used to produce SNG and transportation fuels by methanation and Fischer-Tropsch (FT) synthesis, respectively.

During the conduction of a study to develop gas cleaning for integrated biomass/FT processes, it was recognized that typical off-gases from FT synthesis, due to the high amount of CH_4 and higher hydrocarbons, resemble the composition of Groningen natural gas. The idea was postulated that by upgrading this off-gas FT transportation fuels and SNG could be co-produced and with probably a higher overall efficiency compared to two separate production processes. However, the technical and economic bases for this concept were far from well investigated.

The objective of the underlying study was to determine the technical and economic feasibility of large-scale systems of co-production of “green” FT transport fuels and “green” SNG from biomass. The systems are assessed assuming a targeted annual production of 50 PJ of Fischer-Tropsch transportation fuels and 150 PJ of SNG, which equals 10% of the current (2001) Dutch consumption of these energy carriers. Co-production of 50 PJ/yr of FT transportation fuels and 150 PJ/yr of SNG leads to an annual CO_2 emission reduction of approximately 12.5 Mtonne.

System definition

The evaluated overall system comprises the whole chain of biomass collection, transport, syngas production by gasification, gas cleaning, and FT and SNG synthesis, similar to a previous study on the feasibility of large-scale synthesis gas production from biomass (*i.e.* virgin wood) imported from the Baltic States.

In case of co-production, part of the thermal biomass input is converted to liquid fuels via FT-synthesis and the FT off-gas is methanated to afford SNG. The amount of SNG produced will depend on both the chosen gasification concepts and the operating conditions of the FT-synthesis. If additional SNG is required to meet the objective of replacing 10% of the annual Dutch natural gas consumption, this needs to be produced in a separate dedicated process. In integrated co-production concepts part of the product gas is used for Fischer-Tropsch synthesis and the other part for SNG synthesis, whereas in parallel co-production concepts two different gasification processes are used.

For all evaluated systems an Aspen$^+$ model is constructed in order to determine the mass, heat and work balance of the processes. Six combinations are considered of gasifier type, operating pressures, and pressurisation gas. The Fischer-Tropsch synthesis is operated at a temperature of 220°C and a partial pressure of the syngas components (H_2 and CO) of 40 bar. The FT feed gas is shifted to a H_2/CO ratio of two and all CO_2 is removed. The FT-off-gas is shifted to a H_2/CO ratio of three, necessary for the methanation (at 66 bar). The produced SNG has a Wobbe-index of 43.7 MJ/m_n^3 (regulated by either CO_2 removal after methanation or by adding a part of the CO_2 removed before the FT-synthesis).

The economic assessment is based on the Net Present Value (NPV) and the Internal Rate of Return (IRR). The economical and ecological feasibility is determined based on costs for the CO_2 emission reduction by the replacement of fossil-based fuels.

Feasibility of co-production

In the co-generation concept, 50 PJ/yr of Fischer-Tropsch liquids is produced with SNG as “spin-off” product from FT synthesis. The assessment is concentrated on the four gasification technologies:

- Atmospheric oxygen-blown CFB gasification [A-CFB(O_2)];
- Pressurised (with carbon dioxide) oxygen-blown CFB gasification [P_C-CFB(O_2)];
- Indirect gasification (atmospheric) [indirect];
- Entrained-flow gasification (pressurised and oxygen-blown) [EF].

Overall efficiencies for a given gasification option are effectively independent of the energy carrier produced (*i.e.* FT-liquids or SNG). This means that there is no incentive to produce either energy carrier over the other, with respect to optimising the energy efficiency of a process. The efficiencies for the independent energy carriers SNG (η_{SNG}) and FT-liquids (η_{FT}), however, depend strongly on the CO-conversion in Fischer-Tropsch synthesis (η_c) and the chain growth probability factor α, which was demonstrated by lab-scale Fischer-Tropsch synthesis experiments.

The amount of SNG produced in addition to the 50 PJ/yr of Fischer-Tropsch liquids depends on both the chosen gasification concept and the operating conditions of the FT-synthesis and varies from 3 (for entrained flow) to 134 PJ/yr (for pressurised O_2-blown CFB) if the FT synthesis is operated at maximum FT-production conditions (η_c as well as • equal to 0.95). At these preferred conditions the additional SNG required to meet the 150 PJ/yr production target, should be produced by methanation in a separate SNG process.

Integrated and parallel co-production

The amounts of FT transportation liquids and SNG to be produced were both targeted at 10% of the current Dutch consumption of transportation fuels and natural gas, corresponding to 50 and 150 PJ per year for FT-liquids and SNG, respectively. The production of additional SNG can be carried out via integrated or parallel co-production concepts. In these cases either a side-stream of the gasifier is used for methanation or the product gas from a separated gasifier. Five co-production concepts were considered:

- Integrated co-production with indirect gasification;
- Integrated co-production with pressurised oxygen-blown CFB gasification;
- Integrated co-production with entrained flow gasification;
- Co-production with entrained flow gasification and parallel indirect gasification and;
- Co-production with entrained flow gasification and parallel pressurised oxygen-blown CFB gasification.

In the parallel systems, the Fischer-Tropsch liquids production is based on co-production with entrained flow gasification as then (by far) the highest yield to Fischer-Tropsch liquids is achieved. Although in all concepts the same amounts of FT-liquids and SNG are produced, there is a huge variation in the required biomass input due to the different overall energy efficiency. The integrated co-production concepts with indirect and pressurised gasification require approximately 10% less biomass compared to both parallel concepts.

The major criterion for comparing the economic potential is the CO_2 reduction costs. The expected values for the CO_2 emission reduction costs range from 99 to 174 €/tonne (based on a biomass price of 2.3 €/GJ). The NPV is negative for all concepts, which means that co-production of “green” FT transportation fuels and SNG will require some financial (governmental) incentives or tax exemptions. Furthermore, the Internal Rate of Return (IRR) has to be considered as a negative discount rate at which the NPV of that project equals zero, which means that providing a loan would *cost* interest instead of yielding it. Integrated co-production of FT-liquids and SNG by pressurised oxygen-blown CFB gasification, or indirect gasification, is economically more attractive than by entrained flow gasification. Both concepts with parallel SNG production appear less interesting, however, the differences are not significant considering the uncertainties.

On the long term, increasing costs of mineral oil should make the co-production of “green” FT transportation liquids and SNG from biomass competitive without financial (governmental) incentives for energy investments. The break-even mineral oil prices range from 56 to 83 $/barrel - all well above the current average price of 20 $/barrel.

Integrated co-production concepts have higher net energy efficiencies than the parallel co-production concepts, *i.e.* higher biomass-to-fuel efficiencies. Correspondingly, the economic perspectives, expressed as CO_2 emission reduction costs, of the integrated co-production concepts are better.

Conclusions

This study was directed towards the determination of the technical and economic feasibility of systems for the co-production of “green” Fischer-Tropsch liquid transportation fuels and “green” Substitute Natural Gas (SNG) from biomass. The major conclusions with respect to the technical feasibility of producing SNG as “spin-off” product from Fischer-Tropsch synthesis (*i.e.* by co-generation) are:

- There is no incentive to produce either SNG or FT-liquids as for a given gasification option, the conversion efficiencies to both products essentially equal. The product ratio of SNG and FT-liquids can be controlled by the CO-conversion (η_c) and chain growth factor (α).
- The *overall efficiencies* (FT liquids plus SNG) are higher for CFB and indirect gasification concepts compared to EF gasification as already much CH_4 and C_2 compounds are present in the product gas. On the other hand, the efficiency to FT-liquids is much higher for EF gasification resulting from the presence of all the chemical energy in the gas as syngas components (CO and H_2).
- Additional SNG can be produced either by *“Integrated co-production”*, in which a side-stream of the product gas of the gasifier is used for dedicated methanation or by *“Parallel co-production”*, in which part of the biomass is fed to a second (other type of) gasifier coupled to a dedicated stand-alone methanation.
- Integrated or parallel SNG co-production is preferred over changing the FT synthesis conditions, as it is optimal to operate the FT synthesis at maximum α (*i.e.* α= 0.95), *i.e.* due to the smaller gas volume flows and the better ‘quality’ of the liquid FT products.

The major conclusions of the economic evaluation with respect to producing fixed amounts of 50 and 150 PJ of Fischer-Tropsch transportation fuels and SNG, respectively, are:

- The integrated co-production concepts have generally higher net energy efficiencies compared to the parallel co-production concepts.
- None of the co-production concepts is economically feasible at current conditions and with a biomass price of 2.3 €/GJ, as follows from the *negative* net present values (NPV). The concepts would become economic feasible at mineral oil prices (well) above 56 $/barrel crude oil (in the study a crude oil price of 20 $/barrel is assumed).
- The corresponding CO_2 emission reduction costs range from 99 to 175 €/tonne. The trend in CO_2 costs corresponds to the trend in efficiencies.
- In general, pressurised oxygen-blow CFB gasification [P_C-CFB(O_2)] and Indirect gasification are the most suitable technologies for co-production (see figure 1), with CO_2 reduction costs in the range of the energy tax exemption for “green power” in the Netherlands of 100 €/tonne.

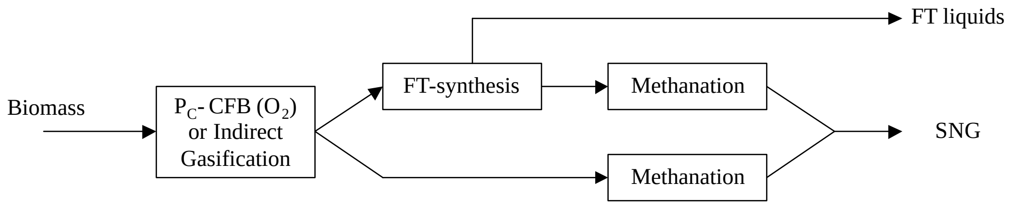

Figure 1 *Optimal system for co-production of "green" Fischer-Tropsch transportation fuels and "green" substitute natural gas (SNG)*

The main conclusion of the study is that the co-production of Fischer-Tropsch transportation fuels and substitute natural gas (SNG) from biomass is economically more feasible than the production of energy carriers in separate processes. Co-production of "green" Fischer-Tropsch transportation fuels and "green" SNG will become an economic feasible process in the Netherlands, when both energy carriers receive the same tax exemptions as currently is given to green electricity.

1. INTRODUCTION

1.1 Introduction

Worldwide energy supply is currently mainly based on fossil fuels. Scenario studies on future energy supply predict a decreased use of these fossil fuels and an increase of the application of renewable fuels [1,2]. Reasons for these expected trends are the finiteness of fossil fuel resources and the environmental problems, especially the emission of greenhouse gases, associated with the use of fossil fuels. Biomass is one of the most promising renewable energy sources considered for replacing fossil fuels. An important aspect of biomass is that besides electricity and heat, chemicals as well as gaseous and liquid fuels can be produced from this renewable source.

In the Dutch situation, especially Substitute Natural Gas (SNG) and transportation fuels are important products in this respect. With a total current consumption of natural gas and transportation fuels in the Netherlands of approximately 2000 PJ/year [3,4] the substitution of a part of the natural gas and transportation fuels market will already lead to a significant contribution towards the sustainability of the energy supply. Furthermore, production of "green" SNG and transportation fuels will allow the use of the existing natural gas and transportation fuels infrastructure and, hence, make a gradual transition from the present fossil fuel-based energy supply system to a complete biomass-based economy possible.

Various technologies are under development for converting biomass into chemicals and transportation fuels. Examples comprise biochemical conversion by fermentation of agricultural residues (*i.e.* to produce bio-ethanol) and chemical conversion by esterification of vegetable oils (*i.e.* to produce bio-diesel). Another technology is thermal conversion by gasification in which the solid biomass is converted into a product gas or synthesis gas [5]. This route has two major advantages, *viz.* almost all biomass materials are suitable for gasification and synthesis gas being a possible universal intermediate in many synthesis processes. Also SNG and transportation fuels can be produced from syngas by methanation and Fischer-Tropsch (FT) synthesis, respectively.

1.2 Project background

The production of SNG and Fischer-Tropsch liquids from biomass are important topics in the ECN Biomass research programme. In a recently finalised joint ECN Biomass and Shell study, co-financed by the Agency for Research in Sustainable Energy (SDE), the necessary gas cleaning and optimal systems designs were developed for integrated biomass/FT processes [6,7,8]. In the catalytic Fischer-Tropsch synthesis one mole of CO reacts with two moles of H_2 to form a hydrocarbon chain extension -(CH_2)-. This polymerisation-like chain-growth reaction results in a range of products, comprising light gaseous hydrocarbons (C_1-C_4), naphtha (C_5-C_{10}), diesel (C_{11}-C_{20}), and wax ($>C_{20}$) fractions. All the liquid compounds, *i.e.* the C_5+ hydrocarbons, are products applicable as transportation fuel, whereas the C_1-C_4 compounds together with the unconverted CO and H_2 form the reactor off-gas. To optimise the product yield, the off-gas of the FT synthesis unit (containing unconverted CO and H_2, inerts, and light hydrocarbons) can be recycled to the syngas unit (figure 2).

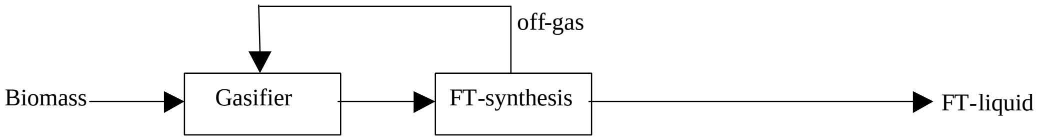

Figure 2 *System for the production of FT-liquids from biomass*

During the conduction of the before-mentioned project it was recognized that the hydrocarbon composition of typical experimentally obtained Fischer-Tropsch off-gases resemble the composition of the Groningen natural gas [9], *i.e.* the gas standard for the Netherlands. These off-gases can be upgraded to Groningen gas quality by methanation of the little CO present and removing CO_2 to adjust the Wobbe-index to the right value. Based on this observation the idea was postulated that, if both "green" FT liquids and "green" SNG are desired products, it is probably more efficient to synthesize SNG from the FT off-gas (figure 3) rather than in a separate process. Or in other words: a higher biomass-to-fuel efficiency will be obtained by co-production of liquids (FT) transportation fuels and SNG.

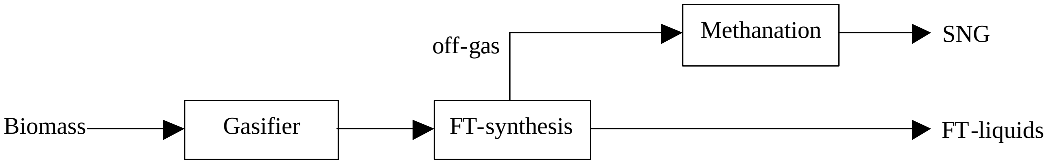

Figure 3 *System for the co-production of liquid FT transportation fuels and SNG*

1.3 Problem definition

Due to the high amount of CH_4 and higher hydrocarbons in the off-gas of FT-synthesis, this gas seems to be highly suitable for upgrading to SNG, and by intuition this increases the overall efficiency of the process. However, the technical and economic bases for this concept are far from well investigated.

Furthermore, in the co-production concept (figure 3) the amount of SNG that can be produced is determined by the Fischer-Tropsch synthesis conditions. Typically, the yield of SNG will be less than 10% at conditions for optimised production of C_5+ FT-liquids. In the current Dutch situation the consumption of transportation fuels and SNG is approximately 1:3 on energy basis. With this co-production concept, this product ratio can only be achieved by operating the FT synthesis at sub-optimal conditions (*i.e.* resulting in effective lower overall efficiencies).

Alternatively, to have the flexibility to produce "green" FT-liquids and "green" SNG in different ratios, the additional required amount of SNG can be produced from a syngas side stream of the gasifier. In this integrated co-production system, the methanation step is assumed to use part of the syngas from the gasifier.

Product gases from different types of gasifiers, however, have very different compositions and correspondingly are more or less suitable for methanation. Therefore, it might be more efficient to produce the additional required amount of SNG in a parallel system. In this so-called parallel co-production concept, the parallel system will be optimised for the separate production of SNG and allows the application of another gasifier being ideal for SNG production.

1.4 Objective

The objective of the study is to determine the technical and economic feasibility of systems of co-generation of “green” Fischer-Tropsch liquid transport fuels and “green” Substitute Natural Gas from biomass. The study is limited to large-scale production, as this is required to make a significant contribution to the Dutch renewable energy targets. The main questions to be answered are: which route of co-generation has the highest efficiency and which route has the highest economic potential? The systems are assessed assuming a targeted annual production of 50 PJ of Fischer-Tropsch transportation fuels and 150 PJ of SNG, which equals 10% of the current Dutch consumption of both these energy carriers. This corresponds to the expected EU targets of 10% renewable energy carriers in 2020. Co-production of 50 PJ/yr of FT transportation fuels and 150 PJ/yr of SNG leads to an annual CO_2 emission reduction of approximately 12.5 Mtonne.

1.5 Approach

In order to fulfil the stated objective, first the overall technical system of gasification, Fischer-Tropsch synthesis, and methanation and the boundaries are defined. Within the system six combinations of gasifier-type and gasification conditions are evaluated. Based on an initial evaluation of the technical aspects, the most promising concepts are evaluated in more detail on their economic and ecological feasibility.

2. SYSTEM DEFINITION

2.1 Overall System

The overall system evaluated within this study comprises the whole chain of biomass collection, transport, syngas production by gasification, gas cleaning, and FT and SNG synthesis. The same system was evaluated in a previous study on the “technical and economic feasibility of large-scale synthesis gas production in the Netherlands from imported biomass feedstock”. ECN Biomass, Shell, BTG, University of Utrecht, and Ecofys jointly carried out this study in 2002 [5]. In principle, in the current study several assumptions and conclusions are taken from that study, as they will not be different for the syngas production part in this study (selected data are presented in appendix A).

In the overall system (figure 4), the biomass is collected in a number of production locations and transported to collection facilities (A), from where it will be transported to and stored at a central port (B). From the central port, the biomass is shipped (C) to the location of the syngas manufacturing facility (D). At the manufacturing facility, biomass will initially be stored before being pre-treated and gasified (E).

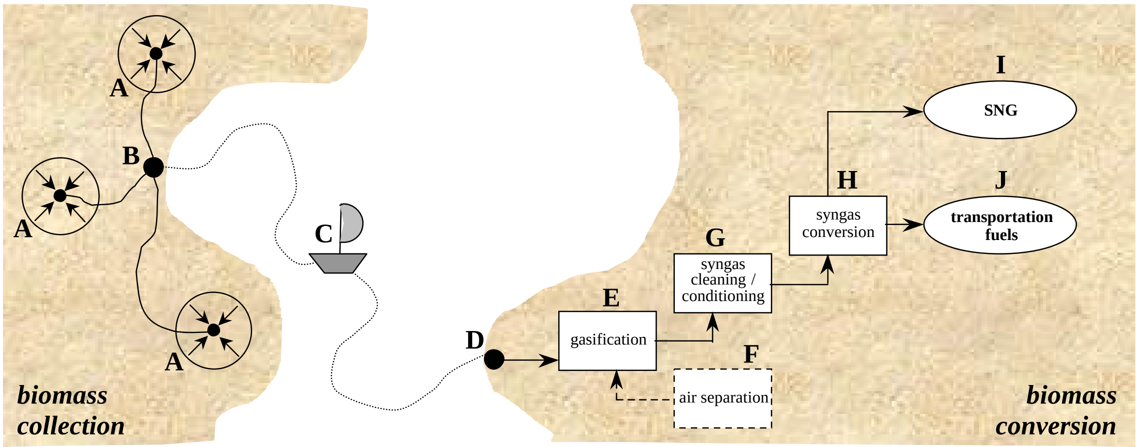

Figure 4 *Overview of the overall system for the large-scale production of FT transportation fuels and SNG from imported biomass*

The biosyngas from the gasifier is cleaned and conditioned (G) to the specifications of the syngas conversion facility (H). The two conversion facilities considered within this study are the Fischer-Tropsch (FT) synthesis in order to produce transportation fuels and methanation in order to produce SNG.

Feedstock. As biomass source, sustainable produced virgin wood (both logs and forestry residues like branches and chips) has been chosen. It can be efficiently collected in the large quantities required for co-generation of SNG and transportation fuels and does not require a major change in land use. Furthermore, its production will lead to a consistent quality of the biomass and offers a realistic alternative for fossil feedstocks, taking into account conversion efficiencies and biomass price. In the process evaluation, biomass is obtained from the Baltic States for reasons of the Baltic States being included within the EU in the near future and the EU striving for less dependency on external energy sources. In addition, the Baltic States have large forestry acreage and reliable biomass contracts with Baltic suppliers are expected to be feasible. The amount of biomass required for syngas production, however, exceeds the current production capacities of the Baltic States, but it is assumed that on a "global" basis biomass import will also be possible from other areas. In this study an average biomass price of 2.3 €/GJ is taken for the biomass delivered to the collection facility. This value corresponds to a biomass mixture of 1:1 logs and residues at prices of 0.6 and 4.0 €/GJ, respectively.

Product yields. Within the study the amounts of Fischer-Tropsch transportation fuels and SNG to be produced were fixed. These amounts were (arbitrarily) set to equal 10% of the current Dutch consumption of both energy carriers. With the current annual consumption being ~500 and ~1500 PJ for transportation fuels [4] and natural gas [3], respectively, the targeted production levels in this study are 50 PJ/year of FT fuels and 150 PJ/year of SNG.

2.2 Co-production options

SNG and FT transportation fuels can be produced by co-production, integrated co-production, or by parallel co-production. In case of co-production 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 (as well as smaller amounts of C_2 hydrocarbons) produced during gasification and gaseous hydrocarbons produced during the FT-synthesis (C_2 to C_4), is upgraded to the standard quality of natural gas by methanation of the remaining CO. A simplified flow sheet of this co- production process is presented as first option in figure 5. Due to the high amount of CH_4 and higher hydrocarbons in the off-gas of the FT-synthesis, the off-gas is highly suitable for the relatively simple upgrading to SNG. The amount of SNG that can be produced will, however, depend on both the chosen gasification concept and the operating conditions of the FT-synthesis.

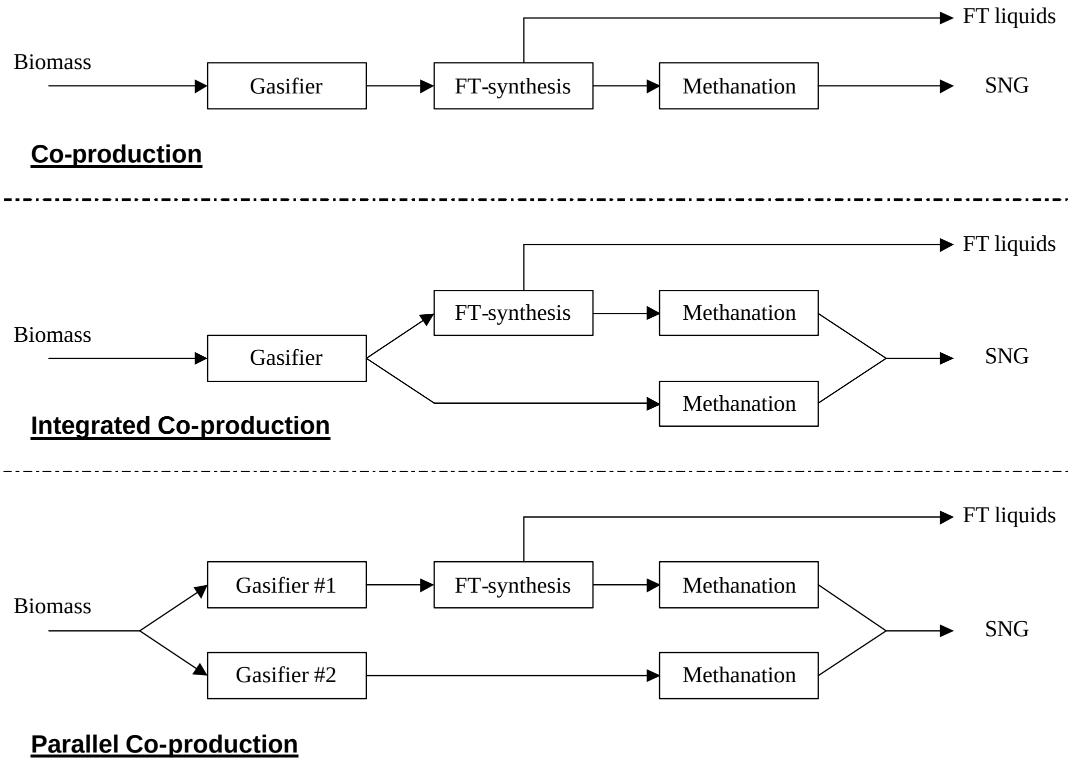

Figure 5 *"Co-production", "Integrated Co-production, as well as " Parallel Co-Production" of FT-liquids and SNG*

Although upgrading of the FT off-gas to "spin-off" SNG is most likely the most attractive concept of co-generation, the amount of SNG that can be produced is limited and fixed by the Fischer-Tropsch synthesis conditions. To be able to meet the objective of replacing 10% of the annual Dutch natural gas consumption, additional SNG needs to be produced. This is realized with a separate dedicated SNG production process, either by integrated or parallel co-production (see second and third concepts in figure 5, respectively). In the integrated concept the product gas of the gasifier is used partly for Fischer-Tropsch synthesis and partly for SNG synthesis; whereas in the parallel concept two different gasification processes are used.

2.3 Technical system modelling

The co-generation of Fischer-Tropsch liquids and SNG has been modelled in accordance to the general flow sheet presented in figure 6.

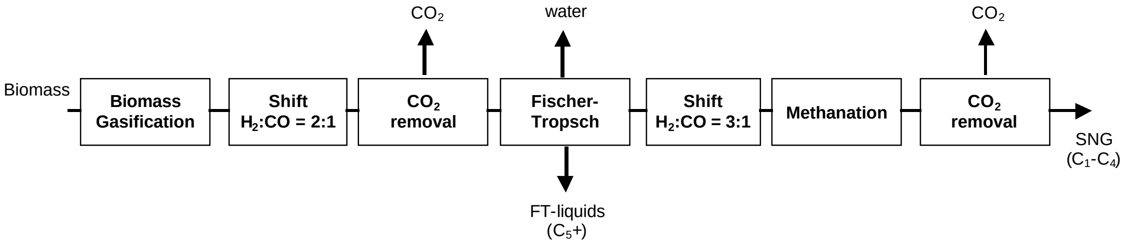

Figure 6 *The general flow sheet of the co-production of FT-liquids and SNG from biomass*

2.3.1 Biomass gasification

Three gasifier types are considered for gasification: circulating fluidised bed (CFB), entrained flow (EF), and indirect gasifiers. Additionally, for the CFB gasifier, air and oxygen-blown gasification as well as atmospheric and pressurised gasification are considered. The atmospheric air-blown CFB gasification technology is included, as it is the preferred current technology for biomass integrated gasification combined cycle systems (BIG-CC). However, the disadvantage is that the product gas is diluted with nitrogen. Alternatives are technologies that produce a non-diluted syngas, hence either oxygen-blown CFB gasification and indirect gasification. Oxygen-blown gasifiers, however, have the disadvantage of additional capital costs associated with the oxygen plant.

Pressurised gasification is included as this holds the advantage of only having to compress the gasification medium instead of the product gas with a larger volume. However, disadvantages are the higher capital costs for the gasifier and the necessity to use inert pressurisation gas. In this study pressurised CFB gasifiers operate at 20 bar. For the CFB both the cases with N_2 and CO_2 as pressurisation medium are considered.

Whereas the CFB and indirect gasifiers are operated at 850°C, the entrained flow (EF) gasifier operates at 1400°C to ensure complete carbon conversion in the short residence time. In this study an oxygen-blown and pressurised (40 bar) EF gasifier is considered, using CO_2 as inert gas for the feed pressurisation.

This results in six different combinations that are assessed:
- Atmospheric air-blown CFB gasification [A-CFB];
- Atmospheric oxygen-blown CFB gasification [A-CFB(O_2)];
- Pressurised (with nitrogen) oxygen-blown CFB gasification [P_N-CFB(O_2)];
- Pressurised (with carbon dioxide) oxygen-blown CFB gasification [P_C-CFB(O_2)];
- Indirect gasification (atmospheric) [indirect];

- Entrained-flow gasification (pressurised and oxygen-blown) [EF].

Brief descriptions of the gasification technologies are presented in appendix B and the product gas compositions are shown in table 1. Calculations are based on untreated wood as biomass feed (average composition obtained from the ECN Phyllis database [10]).

Table 1 *Product gas compositions and properties for the six gasification concepts*

		A-CFB	A-CFB(O_2)	P_N-CFB(O_2)	P_C-CFB(O_2)	Indirect	EF
CO	[vol.%$_{dry}$]	19.3	26.9	16.1	16.1	42.5	46.1
H_2	[vol.%$_{dry}$]	15.6	33.1	18.3	18.3	23.1	26.6
CO_2	[vol.%$_{dry}$]	15.0	29.9	35.4	46.9	12.3	26.9
H_2O	[vol.%$_{ar}$]	13.4	32.2	34.0	34.0	37.1	16.7
CH_4	[vol.%$_{dry}$]	4.2	7.0	13.5	13.5	16.6	0.0
N_2/Ar	[vol.%$_{dry}$]	44.5	0.7	12.3	0.8	0	0.4
C_2	[vol.%$_{dry}$]	1.4	2.4	4.4	4.4	5.5	0.0
CGE*	[%]	79.1	80.6	79.1	79.1	80.4	77.9
LHV	[MJ/m_n^3]	5.76	8.85	8.44	8.05	13.64	7.43
HHV	[MJ/m_n^3]	6.39	9.11	9.18	9.19	12.59	8.03
ρ	[kg/m_n^3]	1.11	0.97	1.09	1.14	0.92	1.08

* Cold Gas Efficiency. Defined as the chemical output of product gas divided by the thermal biomass input of the gasifier

2.3.2 Fischer-Tropsch synthesis

Detailed description and background information on the Fischer-Tropsch synthesis is given in appendix C. In Fischer-Tropsch synthesis one mole of CO reacts with approximately two moles of H_2. In the modelling, an exact consumption ratio of $H_2/CO = 2$ is assumed, see equation (1) and, therefore, the product gas is shifted to match this ratio before entering the Fischer-Tropsch reactor.

$$CO + 2H_2 \Rightarrow -(CH_2)- + H_2O \tag{1}$$

In order to reduce the operating pressure of the FT-synthesis, all CO_2 is removed from the gasification product gas upstream of the reactor. Within the model, removal of CO_2 is assumed to be possible with a selectivity and purity of 100%. The actual selectivity and purity of CO_2 removal will depend on the CO_2 removal technique used within the process (see appendix D).

In Fischer-Tropsch synthesis hydrocarbons are formed in the whole range of C_1 to $>C_{100}$. In the modelling the formation of products is assumed to follow the ideal Anderson-Schulz-Flory (ASF) equation (2):

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

where W_n is the mass fraction of a product consisting of n carbon atoms and α the chain growth probability factor. To incorporate the deviation from the ideal ASF distribution with regard to the yields of the C_1-C_4 hydrocarbon, these values are calculated according to equations (3) and (4).

$$W_1 = \tfrac{1}{2}(1 - \sum_{i=5}^{\infty} W_i) \qquad (3)$$

$$W_{2,3,4} = \tfrac{1}{6}(1 - \sum_{i=5}^{\infty} W_i) \qquad (4)$$

The Fischer-Tropsch synthesis is evaluated for α values varying from 0.5 to 0.95 and a CO-conversion to Fischer-Tropsch products (η_c) of 0 to 0.95, where $\eta_c = 0$ is used to model Stand-Alone SNG production, *i.e.* no FT production

The gaseous C_1-C_4 hydrocarbons leave the reactor in the off-gas, which is fed to the methanation. All the liquid C_5+ hydrocarbons are assumed to be products. Water formed in Fischer-Tropsch synthesis is separated and removed.

2.3.3 Methanation process

Detailed description and background information on the methanation is given in Section C.2 in the Appendices. In the methanation step [11], carbon monoxide is converted to methane according to equation (5). In contrast to Fischer-Tropsch synthesis, methanation requires an H_2/CO-ratio of three. The FT off-gas is shifted to the exact ratio of H_2/CO = 3. In the methanation, a 100% conversion of CO is assumed and, resultantly due to the set H_2/CO ratio, the hydrogen conversion will be also 100%. Hydrocarbons already present in the syngas are assumed to behave as inert during methanation.

$$CO + 3H_2 \Rightarrow CH_4 + H_2O \qquad (5)$$

CO_2 removal takes place after methanation. The amount of CO_2 removed is defined by the condition that the product SNG must have a quality, expressed by the Wobbe-index, comparable to that of standard Dutch natural (Groningen) gas so it can be delivered to the Dutch natural gas grid. More detailed description and background information on the natural gas infrastructure and SNG gas requirements is given in appendix E.

In the model, a value of 43.7 MJ/m_n^3 is used (the Wobbe-index of Groningen gas has a value between 43.5 and 44.4 MJ/m_n^3 [12]). The Wobbe-index *W* is defined, according equation (6), as the ratio of the gross calorific value to the square root of the relative density, with HHV the High Heating Value (MJ/m_n^3), and ρ_g and ρ_{air} the gas and air densities (in kg/m_n^3) [13].

$$W = \frac{HHV}{\sqrt{\dfrac{\rho_g}{\rho_{air}}}} \qquad (6)$$

In some cases in the modelling CO_2 had to be *added* to the raw SNG, as after shift the Wobbe-index was too high. In this case recompressed CO_2 previously removed from the gas upstream of the FT reactor was used.

2.3.4 Aspen modelling

For all evaluated systems an Aspen$^+$ model is constructed in order to determine the mass, energy, and work balances of the process. In order to clarify the model a simplified flow sheet of the simulated system is presented in figure 7. The product gas from the gasifier |1| is cooled and cleaned from tars and inorganic impurities. Tar removal is established with the ECN OLGA technology [14] (the tars are returned to the gasifier and destructed) and inorganics (*i.e.* water soluble impurities) are subsequently removed by wet scrubbing at 20°C (|2|). The heat duty resulting from the product gas cooling is used for steam and electricity generation. In case of CFB and indirect gasification, the amount of usable heat is limited by a temperature region of 900 to 400°C due to the application of an OLGA unit, with some additional amount of heat resulting from OLGA at a temperature level of at most 200°C. In case of entrained flow gasification usable heat can be obtained at temperature levels from 1300 to 110°C (no OLGA is applied as the product gas is tar free due to the high gasification temperature). After cleaning, the product gas is led to either the Fischer-Tropsch synthesis section |3-11| or the methanation section |12-16|.

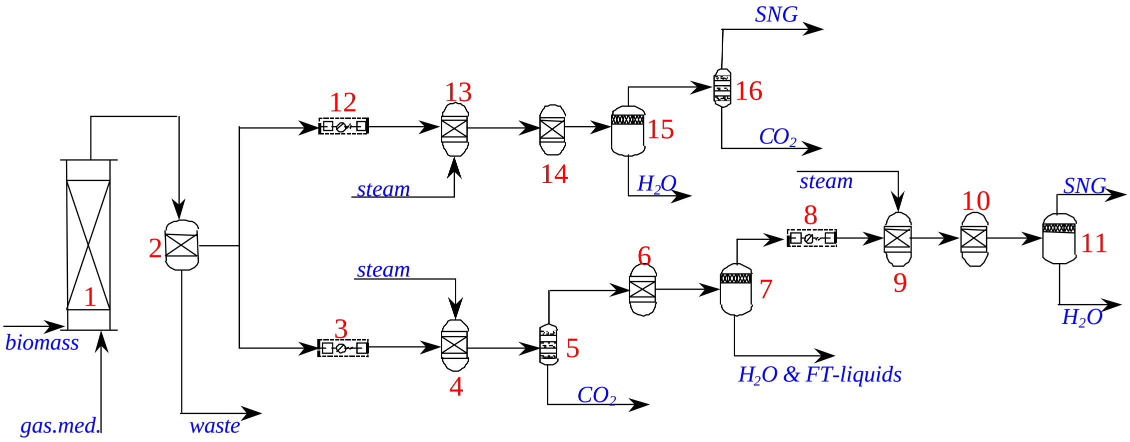

Figure 7 *Simplified flow sheet of the within Aspen$^+$ simulated system. The red numbers refer to explanations in the text*

The Fischer-Tropsch synthesis is operated at a temperature of 220°C and a partial pressure of the syngas components (H_2 and CO) of 40 bar. As gasification is carried out at 20 bar, the product gases need to be compressed |3| and up to total pressures well above 40 bar due to the presence of inert N_2. For oxygen-blown CFB gasification this pressure is 53.5 bar and for indirect gasification 61.8 bar, whereas in case of entrained flow gasification no additional compression is required as the gasification is carried out at 40 bar and the conditioned product gas only consists of H_2 and CO.

Before entering the Fischer-Tropsch synthesis |6|, the synthesis gas is shifted |4| in order to obtain a H_2/CO ratio of two and all CO_2 is removed |5|. At a temperature of 5°C the water and FT-liquids are separated from the gas |7|. The FT-off-gas is then compressed to 66 bar |8|, before being shifted |9| to obtain a H_2/CO ratio of three, necessary for the methanation |10|. During methanation, operated at 66 bar to match the maximum pressure of the high-pressure transmission lines of the Dutch natural gas grid (*cf.* appendix E) and 270°C, all CO is converted. The SNG produced is free of water |11| and has a Wobbe-index of 43.7 MJ/m_n^3 (regulated by adding a part of the recompressed CO_2 removed before the FT-synthesis).

As methanation takes place at 66 bar, gas fed directly to the methanation section has to be compressed |12| in all cases. As with the off-gas of the FT-synthesis, the synthesis gas is shifted in order to obtain the H_2/CO ratio of 3 |13| before being methanated |14|. After methanation the SNG is dried |15|. The Wobbe-index is regulated to a value of 43.7 MJ/m_n^3 by removing part of the CO_2 in the SNG.

The heat duties from the cooling of the synthesis gas, as well as the sensible heat from the methanation and FT-synthesis (at a temperature level of 270 and 220°C, respectively) are used for the generation of high-pressure superheated steam. This steam is used for heating purposes (within the system) and electricity generation, by expanding in a steam turbine. Part of the produced steam is also required within the gasifier. The amount of CO_2 removed from the gases might be used for CO_2 sequestration in deep coal layers. This will, however, require compression of the CO_2, as the CO_2 is obtained at atmospheric pressure.

2.4 Economic system modelling

The economic assessment is based on the Net Present Value and the Internal Rate of Return. The Net Present Value (NPV) represents an approach used in capital budgeting where the present value of cash inflow is subtracted from the present value of cash outflows. This relationship is expressed by the following formula (7):

$$\mathrm{NPV} = \sum_{t=0}^{T} \frac{CF_t}{(1+r)^t} \qquad (7)$$

with CF_t the cash flow at time t and r the cost of capital (*i.e.* interest or discount rate). When the NPV is greater than zero the benefits of the project outweigh the costs at the chosen cost of capital, and the project represents a worthwhile investment. Providing the discount rate has been chosen appropriately, all projects with NPV greater than zero should be considered worthwhile investments [15]. The Internal Rate of Return (IRR) of a project is the discount rate at which the NPV of that project equals zero, hence:

$$\sum_{t=0}^{T} \frac{CF_t}{(1+\mathrm{IRR})^t} = 0 \qquad (8)$$

The IRR decision rule specifies that all (independent) projects with an IRR greater than the cost of capital should be accepted. When choosing among mutually exclusive projects, the project with the highest IRR should be selected (as long as the IRR is greater than the cost of capital). The IRR, unlike the NPV, has the advantage that it does not relate to the size of the original investment [15]. However, in this approach the ‘risk’ related to capital investment in a project is neglected. If the ‘risk’ of a project is included in the investment consideration, a project with a lower IRR and a low risk might be more attractive than a project with a high IRR but also a higher risk.

In this study, the determination of the economic and ecological feasibility will be based on the CO_2 emission reduction costs. By producing either transportation fuels or SNG from biomass a certain amount of CO_2 emission is avoided due to the replacement of fossil-based fuels. The CO_2 emission reduction costs or specific environmental costs involved can be determined by relating the additional production costs (or negative NPV) to the avoided CO_2 emission.

All the input data concerning biomass properties, storage and transport, investment costs, utility consumption, waste production and costs, O&M costs and economic parameters are summarised in appendix A.

3. FEASIBILITY OF CO-PRODUCTION

In this chapter, the technical feasibility of the co-production concept is studied. The amount of Fischer-Tropsch liquids to be produced is taken equal to 10% of the current transportation fuels consumption (500 PJ/yr), hence 50 PJ/yr. As SNG is a “spin-off” product from FT synthesis, the amount produced will depend on both the chosen gasification concept and the FT operating conditions.

3.1 Gasification concepts

In the system definition six combinations of gasifier type, gasification pressure and medium, and pressurisation medium were initially selected in this study (section 2.3.1). However, from the first system modelling it was apparent that in the case of two gasifier product gases it was impossible to meet SNG specifications. Due to the presence of large concentrations of nitrogen in the product gases from the air-blown and the nitrogen-pressurised CFB, the Wobbbe-index of 43.7 MJ/m_n^3 could not be reached. As nitrogen, unlike CO_2, cannot (economic competitive) be removed from the gas, it is not possible to adjust the quality of the gas to the standard quality of the Groningen natural gas. In the further study, these two gasification concepts are therefore excluded and the assessment is concentrated on the remaining four combinations:

- ~~Atmospheric air-blown CFB gasification [A-CFB];~~
- Atmospheric oxygen-blown CFB gasification [A-CFB(O_2)];
- ~~Pressurised (with nitrogen) oxygen-blown CFB gasification [P_N-CFB(O_2)];~~
- Pressurised (with carbon dioxide) oxygen-blown CFB gasification [P_C-CFB(O_2)];
- Indirect gasification (atmospheric) [indirect];
- Entrained-flow gasification (pressurised and oxygen-blown) [EF].

3.2 Production efficiencies

For the four different gasification options the production efficiency from biomass to FT-liquids (C_5+) and SNG as well as the overall efficiencies have been determined as a function of the CO-conversion in Fischer-Tropsch synthesis (η_c) and the chain growth probability factor α. In table 2 the results are presented for three extreme cases:

- Maximum FT production (η_c= 0.95), with maximum liquids C_5+ yield (α= 0.95);
- Maximum FT production (η_c= 0.95), with significant gaseous C_1-C_4 yield (α= 0.5);
- No Fischer-Tropsch synthesis (η_c= 0), only SNG production.

More detailed results are presented in appendix F. It is interesting to note that overall efficiencies for each gasification option are effectively independent of the energy carrier produced (*i.e.* FT-liquids or SNG). The efficiency for sole SNG production is *slightly* lower, however, this effect can be solely contributed to the larger extent of water-gas shift required for SNG production (*i.e.* to H_2/CO= 3) compared to FT synthesis (H_2/CO= 2). The equal production efficiencies means that there is no incentive to produce either energy carrier over the other, with respect to optimising the energy efficiency of a process.

On the other hand, the efficiencies for the independent energy carriers SNG (η_{SNG}) and FT-liquids (η_{FT}) depend strongly on the FT CO-conversion (η_c) and the chain growth factor α. In case sole production of FT-liquids is preferred, entrained flow gasification is the preferred concept as this has the largest efficiency towards FT-liquids (55.4%). When sole SNG production is preferred, the concept of pressurised oxygen blown CFB gasification (efficiency towards SNG of 72.8%) is preferred. However, the differences with the atmospheric and indirect gasification concepts are small.

The *overall* yield of the EF concept is in all cases much lower than for the other three concepts. This is a result from the high concentrations of CH_4 and C_2 compounds that are already present in the product gases of these three gasifiers. In all cases the co-generation of SNG with FT-liquids at conditions for maximum FT production results in a significant increase of the overall yield to energy carriers.

Table 2 *Specific and total conversion efficiencies from biomass to FT liquids and SNG*

		η_{SNG}	η_{FT}	$\eta_{overall}$
Indirect gasification [indirect]				
η_c= 0.95	α= 0.95	45.5	25.9	71.4
η_c= 0.95	α= 0.5	66.0	5.5	71.5
η_c= 0	-	71.2	0	71.2
Atmospheric O_2-blown gasification [A-CFB(O_2)]				
η_c= 0.95	α= 0.95	31.7	37.4	69.1
η_c= 0.95	α= 0.5	61.4	7.9	69.3
η_c= 0	-	68.9	0	68.9
Pressurised O_2-blown gasification [P_C-CFB(O_2)]				
η_c= 0.95	α= 0.95	53.2	19.8	73.0
η_c= 0.95	α= 0.5	68.8	4.1	73.0
η_c= 0	-	72.8	0	72.8
Entrained flow gasification [EF]				
η_c= 0.95	α= 0.95	3.3	55.4	58.7
η_c= 0.95	α= 0.5	47.2	11.7	58.9
η_c= 0	-	58.3	0	58.3

3.3 “Spin-off” SNG production

The amount of FT-product to be produced is taken equal to 10% of the current transportation fuels consumption (500 PJ/yr), hence 50 PJ/yr (*cf.* section 2.1). In case of SNG being a “spin-off” product of the FT-synthesis, the amount of SNG produced will depend on both the chosen gasification concept and the operating conditions of the FT-synthesis. If the Fischer-Tropsch synthesis is operated at maximum FT-production conditions (CO-conversion as well as α equal to 0.95), the product ratio of SNG to transportation fuels varies from 0.06 (for entrained flow) to 2.7 (for pressurised O_2-blown CFB), as shown in table 3.

Table 3 *Product distribution by co-generation with maximum FT-liquids production (η_c= 0.95 and α= 0.95) and SNG as "spin-off" product*

	Transportation fuels [PJ/yr]	SNG [PJ/yr]	Product ratio [-]
Entrained flow	50	3	0.06
Atmospheric O_2-blown CFB	50	42	0.85
Indirect	50	88	1.8
Pressurised O_2-blown CFB	50	134	2.7

In this study, however, the targeted amount of SNG to be produced is 150 PJ/year, equal to 10% of the Dutch natural gas consumption. Therefore, the desired product ratio of SNG and FT-liquids is 3. The product ratio of SNG and FT-liquids can easily be controlled by adjustment of the operating conditions of the FT-synthesis (*i.e.* lowering • and/or η_c). As both α and η_c have a significant influence on the amount of SNG produced there are innumerable combinations for which a product ratio of SNG to transportation fuels equal to 3 can be obtained. These combinations of α and η_c are presented in figure 8. Decreasing either • or η_c results in a decreased production of FT transportation fuels and a higher SNG production (*cf.* data in table 2). The correlation between the synthesis conditions and the product formation (*e.g.* the amount of methane and a estimated value of α) was also shown by experiments in the ECN micro-flow Fischer-Tropsch synthesis reactor [16].

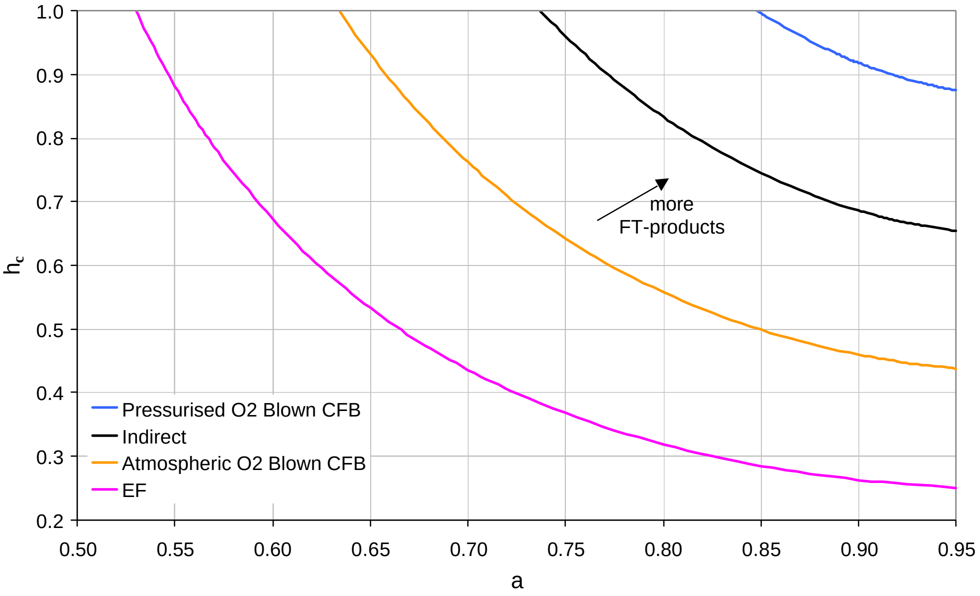

Figure 8 *Operating conditions of the FT-synthesis (• and h_c) of that result in a co-production product ratio of SNG to FT transportation fuels equal to three*

3.4 Choice of co-production approach

Alternatively to varying the operating conditions (*i.e.* decreasing α and/or η_c) of the Fischer-Tropsch synthesis, also part of the product gas can be directly fed to the SNG synthesis section, bypassing the FT-synthesis. This is preferable based on economic considerations, as the overall investment costs for the FT section will be lower due to the smaller volume flow through the FT-synthesis section, whereas its has (almost) no influence on the total flow through the methanation section. More important, however, is that C_5+ liquids produced at high values of the chain growth probability factor α in the FT-synthesis afford better “quality” transportation fuels.[1] Therefore, the FT-synthesis is preferably operated at α= 0.95. As at these conditions the targeted amount of SNG cannot be produced as “spin-off” product, additional methanation of part of the product gas would be necessary. Considering both the economic and product quality arguments, it was decided to assume in further assessment in all cases operational conditions for maximum FT-synthesis (*i.e.* maximum conversion and chain growth probability: $\alpha = \bullet_c = 0.95$). Additional SNG is co-produced by methanation in a dedicated SNG section.

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[1]. Detailed assessment of the quality of FT products and upgrading of the liquids to transportations fuels is not within the scope of this study. However, is short: the main application of FT-derived transportation fuels is foreseen as diesel substitute (being approx. the C_{12}-C_{22} hydrocarbons range). FT diesel can be directly synthesises, as well as, produce by mild and selective hydrocracking of the higher (*i.e.* >C_{22}) hydrocarbons. The shorter FT liquids (*e.g.* C_5-C_{11}) have no diesel application. Lower α values afford more short hydrocarbons - also explaining the larger yield of SNG compounds - and resultantly a lower yield of FT-liquids of the right “quality”.

4. INTEGRATED AND PARALLEL CO-PRODUCTION

The amounts of FT transportation liquids and SNG to be produced were both targeted at 10% of the current Dutch consumption of transportation fuels and natural gas. This corresponds to 50 and 150 PJ per year for FT-liquids and SNG, respectively. The assessment of the concepts for co-production of FT-liquids and SNG in the previous chapter concluded that at conditions for optimal FT-synthesis not sufficient SNG is produced. Furthermore, sub-optimal operation of the FT-synthesis to increase the SNG yield was not preferred. Therefore, additional SNG production by separate integrated or parallel methanation is required.

4.1 Technical evaluation

4.1.1 Gasification concepts

The production of additional SNG can be carried out via an integrated or parallel co-production concept. In these concepts, respectively, a side-stream of the gasifier is used for methanation or the product gas from a separate gasifier (*cf.* the systems depicted in figure 5). Five co-production concepts are considered:

- Integrated co-production with indirect gasification;
- Integrated co-production with pressurised oxygen-blown CFB gasification;
- Integrated co-production with entrained flow gasification;
- Co-production with entrained flow gasification and parallel indirect gasification;
- Co-production with entrained flow gasification and parallel pressurised oxygen-blown CFB gasification.

To reduce the number of concepts to be assessed, the atmospheric oxygen-blown CFB concept is not further studied. The indirect and pressurised CFB gasification concepts are selected as they give the highest yield to SNG (compared to atmospheric CFB gasification). In the parallel systems, the Fischer-Tropsch liquids production is based on co-generation with entrained flow gasification as then (by far) the highest yield to Fischer-Tropsch liquids is achieved.

4.1.2 Energy balances

The targeted annual production of 50 PJ of FT-liquids and 150 PJ of SNG corresponds to production capacities of 1.75 and 5.25 GW$_{th}$, respectively, with an annual plant operating time of 8000 hours. The Fischer-Tropsch synthesis is operated at maximum FT-production conditions (CO-conversion as well as α equal to 0.95). The simplified energy balances for the five co-production concepts are presented in appendix G and the results are summarised in table 4.

It is striking to see that, although in all concepts the same amounts of FT-liquids and SNG are produced, there is a huge variation in the required biomass input due to the different overall energy efficiencies. The integrated co-production concepts with indirect and pressurised gasification require approximately 10% less biomass compared to both parallel concepts (the last columns in table 4). On the other hand, the integrated entrained flow concept requires even more than 30% biomass due to the relatively low overall efficiency of 58%. Solely based on the overall system efficiency, an integrated system based on pressurised oxygen-blown gasification would be preferred.

Table 4 *Summarised energy balances for five SNG and FT-liquids co-production concepts. See text for explanation of the concepts.*

Co-production System Energy flows in [GW_{th}]	Indirect	P_C-CFB(O_2)	EF	EF & Indirect	EF & P_C-CFB(O_2)
Total biomass input	9.95	9.48	12.20	10.56	10.42
Biomass to co-production	-	-	-	*3.21*	*3.21*
Biomass to SNG	-	-	-	*7.37*	*7.21*
Product gas flow	8.0	7.5	9.5	-	-
Feed gas to co-production	*5.5*	*7.0*	*2.5*	-	-
Feed gas to SNG	*2.5*	*0.5*	*7.0*	-	-
FT-liquids*	1.76	1.75	1.78	1.78	1.78
SNG from FT off-gas	*3.11*	*4.71*	*0.11*	*0.11*	*0.11*
Stand-alone SNG	*2.22*	*0.46*	*5.23*	*5.25*	*5.25*
Total SNG*	5.33	5.17	5.34	5.36	5.36
Net energy efficiency	71%	73%	58%	68%	69%

* The small differences compared to the targeted energy flows of 1.75 and 5.25 GW_{th} for the FT-liquids and SNG, respectively, are due to iterations in the calculations.

4.1.3 SNG composition

The specification for the SNG produced is that the Wobbe-index is equal to 43.7 MJ/m_n^3, which is achieved by removing the right amount of CO_2 from the raw SNG. The molar composition and the lower heating value (LHV) of the produced SNG depend on the production concepts. In table 5 the SNG properties are presented for three gasifiers in the cases where the SNG is produced from the FT off-gas as well as in the separate methanation. Furthermore, the composition of the SNG stream is presented that is obtained when both streams are mixed in the integrated co-production concepts. For illustration, also the composition of the Groningen natural gas is included in table 5.

The major differences in the SNG streams are found between entrained flow gasification and the other two gasification concepts. The SNG from indirect and CFB gasification contains large amounts of C_2 compounds that are already present in the gasification product gas. By entrained flow gasification all hydrocarbons are converted mainly into CO, so the only C_2 present is a product from FT synthesis. Therefore, the SNG from entrained flow gasification consists mainly of methane.

4.2 Economic evaluation

The five co-production systems are all technically feasible for the co-production of 50 PJ of transportation fuels and 150 PJ of SNG per year to replace 10% of the national Dutch consumption of these energy carriers. However, the major criterion for further development or implementation is the economic feasibility. In this study, the CO_2 reduction costs are used as comparing value for the economic and ecological potential.

The assessment of the economic potential is carried out primarily with the same data, and under the same assumptions, as the previous study on the “technical and economic feasibility of large-scale synthesis gas production in the Netherlands from imported biomass feedstock” [5]. The input data for the evaluation are presented in appendix A. Most data are presented in a certain (probability) range with a minimum, maximum, and expected value in order to enable sensitivity and risk analysis of CO_2 emission reduction costs, Net Present Value (NPV), and Internal Rate of Return (IRR) towards the uncertainty in the assumptions made.

Table 5 *SNG compositions for the evaluated integrated co-production concepts. N.B. all presented gases have a Wobbe-index of 43.7 MJ/m_n^3 and are on specification for addition to the Dutch natural gas grid. All H_2 and CO are completely converted to CH_4*

SNG composition	CH_4 [mol%]	CO_2 [mol%]	N_2 [mol%]	C_2 [mol%]	C_3 [mol%]	C_4 [mol%]	LHV [MJ/kg]
Indirect gasification [Indirect]							
SNG from FT off-gas	63.0	16.6	-	20.2	0.1	0.1	33.9
Stand-alone SNG	73.6	14.1	-	12.3	-	-	35.3
Mix SNG	*67.5*	*15.5*	-	*16.8*	*0.1*	*0.1*	*34.4*
Pressurised oxygen-blown CFB gasification [P_c-CFB(O_2)]							
SNG from FT off-gas	62.3	13.8	3.5	20.2	0.1	0.1	34.2
Stand-alone SNG	70.5	12.8	2.4	14.3	-	-	35.1
Mix SNG	*63.1*	*13.7*	*3.4*	*19.6*	*0.1*	*0.1*	*34.3*
Entrained flow gasification [EF]							
SNG from FT off-gas*	-	-	-	-	-	-	-
Stand-alone SNG	88.7	9.5	1.8	-	-	-	37.7
Mix SNG	*88.3*	*9.1*	*2.3*	*0.1*	*0.1*	*0.1*	*37.7*
Groningen natural gas	*81.3*	*0.9*	*14.4*	*2.9*	*0.4*	*0.1*	

* The quality of the product SNG from methanation of the FT off-gas is too good to meet the Wobbe-index, *i.e.* addition of CO_2 would be required. Therefore, this product is mixed with the stand-alone produced SNG and the mix fraction is brought to the Wobbe-index specification by CO_2 removal.

4.2.1 CO_2 emission reduction costs

The expected values for the costs of CO_2 emission reduction, as well as the NPV and IRR are presented in table 6. With the NPV being negative for all concepts, co-production of "green" FT transportation fuels and SNG will require some financial (governmental) incentives for profitable energy investments. Furthermore, the Internal Rate of Return (IRR) of the project has to be considered as a negative discount rate at which the NPV of that project equals zero, which means that providing a loan would *cost* interest instead of yielding it.

Table 6 *CO_2 emission reduction costs, NPV, and IRR for the five co-production concepts (ranking in order of increasing CO_2 reduction costs)*

Co-production concepts	CO_2 reduction costs [€/tonne]	NPV [10^9 €]	IRR [%]
Pressurised CFB(O_2)	99	-3.0	-16.5
Indirect	107	-3.5	-17.8
Entrained flow & Pressurised CFB(O_2)	117	-3.3	-16.6
Entrained flow & Indirect	124	-4.0	-18.6
Entrained flow	175	-6.4	-23.0

The uncertainties in the CO_2 reduction costs, NPV, and IRR are the result of the uncertainties in the assumptions and input data used for the feasibility study. In figure 9 *(left plot)* the uncertainties in the CO_2 costs are presented as cumulative probability distributions. The cumulative probability distributions of the five different co-production concepts show significant overlapping, making it difficult to draw a comparison between the economic and ecological feasibility of the concepts.

The major contributors to these uncertainties, however, are *biomass* related (*e.g*. biomass production capacity and price). These uncertainties apply equally to all concepts. Therefore, cumulative probability distributions of the CO_2 costs after exclusion of the uncertainties in biomass production give a more relevant representation of the differences between the five concepts. From the right plot of figure 9 can be seen that due to the exclusion of these major uncertainties the curves become much steeper and less overlapping.

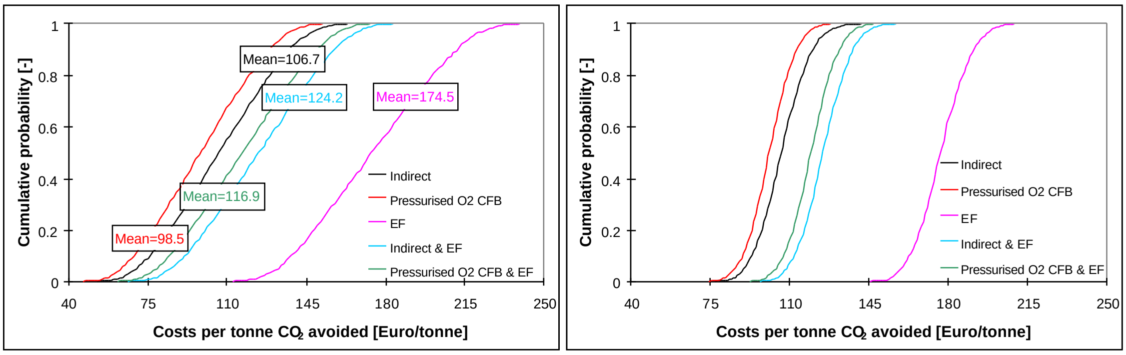

Figure 9 *Cumulative probability distributions of the CO_2 emission reduction costs for the five co-production concepts, with the uncertainties in biomass production included (left) and excluded (right)*

Integrated co-production of FT-liquids and SNG by pressurised oxygen-blown CFB gasification, or indirect gasification, is clearly economically more attractive than by entrained flow gasification. Both concepts with parallel SNG production appear less interesting, however, due to the large overlap of the probability curves the differences are not significant. The ranking of the five concepts based on CO_2 reduction costs correlates to the net energy efficiencies (*cf*. table 4). Due to the (relatively) high biomass price used in this study (2.3 €/GJ), a lower efficiency implies higher costs, as more biomass is required.

4.2.2 Break-even mineral oil price

The economic evaluation and the calculations of the CO_2 emission reduction costs are based on a mineral oil price of 20 $/barrel of crude oil. On the long term, increasing costs of mineral oil should make the co-production of “green” FT transportation liquids and SNG from biomass competitive without financial (governmental) incentives for energy investments. In other words, the CO_2 emission reduction costs will then be zero. In table 7 the break-even mineral oil prices are shown for the five concepts. The break-even mineral oil prices are all well above the current average price of 20 $/barrel.

The CO_2 reduction costs for the integrated co-production options are in range with the energy tax exemption that applies for “green electricity” in the Netherlands (*viz*. the MEP). When for the green fuels a comparable taxation regulation would apply, short-term market implementation of “green” FT transportation liquids and SNG would be made possible.

Table 7 *Break-even mineral oil prices at which CO_2 emission reduction costs are zero (ranking in order of increasing oil price)*

	Break-even mineral oil price [$/barrel]
Pressurised CFB(O_2)	56.0
Indirect	57.9
Entrained flow & Pressurised CFB(O_2)	62.5
Entrained flow & Indirect	64.4
Entrained flow	82.9

4.3 Integrated versus parallel co-production

The motivation to perform the current study was that co-production of FT transportation liquids and SNG from biomass would result in higher biomass-to-fuel efficiencies compared to two separate parallel production process. Resultantly, the economic perspectives of the fuel production would be better.

Actually, the two co-production concepts with parallel SNG production, *i.e.* EF&Indirect and EF&CFB(O_2), can be considered as complete “separate” production concepts as in these cases only 2% of the targeted amount of SNG is co-generated with the 1.75 GW_{th} of FT-liquids. From table 4 it follows that these concepts have a lower net energy efficiency than the integrated co-production concepts based on indirect or pressurised CFB(O_2) gasification. This proves that by co-production the energy efficiency indeed can be increased. Correspondingly, the economic and ecological perspectives, expressed as CO_2 emission reduction costs, of the integrated co-production concepts based on these concepts are better (*cf.* table 6).

In the study the annual amounts of FT transportation fuels and SNG to be produced were fixed (at 50 PJ and 150 PJ, respectively). The conclusions of the study will not change when, for example, equal production quantities of 100 PJ are taken. However, the lower the SNG/FT production ratio, the more SNG can be produced by integrated co-production or by co-generation. In that case the economic perspectives will improve compared to the cases assessed in this study.

5. CONCLUSIONS

This study was directed towards the determination of the technical, economic, and ecological feasibility of systems for the co-production of “green” Fischer-Tropsch liquid transportation fuels and “green” Substitute Natural Gas (SNG) from biomass.

The major conclusions with respect to the technical feasibility of producing SNG as “spin-off” product from Fischer-Tropsch synthesis are:

- Air-blown [A-CFB] or nitrogen-pressurised [P_N-CFB(O_2)] gasification concepts are not suitable for co-production as due to the amount of nitrogen in the gas the Wobbe-index specification for SNG cannot be met.
- There is no incentive to produce either SNG or FT-liquids with respect to optimising the utilisation of biomass, as the conversion efficiencies to both energy carriers are essentially equal for a chosen gasification concept.
- The product ratio of SNG and FT-liquids can be controlled by the CO-conversion (η_c) and chain growth factor (α) in the Fischer-Tropsch synthesis. At decreasing values of η_c and α, increasing amounts of SNG can be co-produced.
- The *overall* efficiencies (of both FT-liquids and SNG) are in the same range for all gasification concepts, except for entrained flow [EF] gasification that has a much lower efficiency (*i.e.* 10-15%-points). This is a result from the high concentrations of CH_4 and C_2 compounds already present in the product gases from CFB and indirect gasifiers.
- The efficiency to FT-liquids is much higher for EF gasification (*e.g.* almost threefold compared to pressurised CFB gasification) resulting from the presence of all the chemical energy in the gas as syngas compounds (CO and H_2), which is available for FT synthesis.
- Additional SNG can be produced either by *“Integrated co-production”*, in which a side-stream of the product gas of the gasifier is used for dedicated methanation or by *“Parallel co-production”*, in which part of the biomass is fed to a second (other type of) gasifier coupled to a dedicated stand-alone methanation.
- Integrated or parallel SNG co-production is preferred over changing the FT synthesis conditions (•, •$_c$), as it is optimal to operate the FT synthesis at maximum α (*i.e.* α= 0.95).

In the economic evaluation the amounts of Fischer-Tropsch transportation fuels and SNG to be produced were fixed at 10% of the current Dutch consumption, *i.e.* 50 and 150 PJ, respectively. The major conclusions with respect to the economic evaluation are:

- The integrated co-production concepts have higher net energy efficiencies compared to the parallel production concepts (*i.e.* 71-73% versus 68-69%) and, resultantly, require the least biomass input. The exception is the integrated entrained flow co-production concept, which has an efficiency of only 58% due to the absence of SNG C_1-C_4 compounds in the product gas.
- None of the co-production concepts is economically feasible at current conditions and with a biomass price of 2.3 €/GJ, as follows from the *negative* net present values (NPV) of 3.0 to 6.4 *billion* euros. The concepts would become economic feasible at mineral oil prices above 56 $/barrel crude oil (in the study a current crude oil price of 20 $/barrel is assumed).
- The corresponding CO_2 emission reduction costs range from 99 to 175 €/tonne. The trend in CO_2 costs corresponds to the trend in efficiencies. The lower value is just in the range of the energy tax exemption for “green power” in the Netherlands of 100 €/tonne.
- In general, pressurised oxygen-blow CFB gasification [P_C-CFB(O_2)] and Indirect gasification are the most suitable technology for co-production (see figure 10). This is due to the presence of already large amounts of methane and C_2 components in the product gases from gasification.

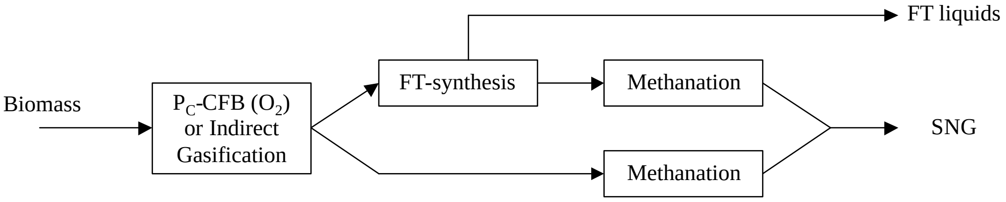

Figure 10 *Optimal system for co-production of 50 PJ/yr "green" Fischer-Tropsch transportation fuels and 150 PJ/yr "green" substitute natural gas (SNG), which corresponds to 10% of the 2001 consumption*

The main conclusion of the study is that the co-production of Fischer-Tropsch transportation fuels and substitute natural gas (SNG) from biomass is economically more feasible than the production of both energy carriers in separate processes. The net biomass-to-fuel energy efficiency is higher, less biomass is required, and the NPVs are less negative (*i.e.* lower CO_2 emission reduction costs).

The co-production of "green" Fischer-Tropsch transportation fuels and "green" SNG will become an economically feasible process in the Netherlands, when both energy carriers receive the same tax exemptions as is currently given to green electricity or at crude mineral oil prices above 56 $ per barrel.

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APPENDICES

APPENDIX A ASSUMPTIONS AND INPUT DATA

In this appendix selected input data concerning biomass properties, investment costs, and economic parameters are summarised. Most data in table 8 are derived from reference [1].

Table 8 *Selected data and assumptions used for the feasibility study*

Parameter	Minimum	Expected value	Maximum	Unit
General				
Dollar exchange rate	0.91	1.13	1.36	€/$
Dutch electricity price	-	40	-	€/MWh
Dutch required IRR	0.09	0.12	0.15	
Interest rate	4	6	8	%
Mineral oil costs	15	20	26	$/barrel
Specific CO_2 emission natural gas	-	56.0	-	kg/GJ
Specific CO_2 emission transportation fuels	-	81.1	-	kg/GJ
Biomass related				
Biomass price	-	2.3	-	€/GJ
Biomass production	2	-	10	tonne_{dry}/acre/yr
Bulk density logs	310	-	375	kg/m^3
Bulk density residues	200	-	300	kg/m^3
Calorific value logs (HHV_{daf})	19	20	21	GJ/tonne
Calorific value residues (HHV_{daf})	19	20	21	GJ/tonne
Moisture content logs (wt.$\%_{wet}$)	0.25	0.30	0.50	-
Moisture content residues (wt.$\%_{wet}$)	0.25	0.30	0.50	-
Volume percentage of residues in biomass	0.30	0.50	0.70	-
Plant related				
Investment air separation	2.355	2.944	3.532	M€/(kg/s)
Investment indirect gasifier	0.120	0.150	0.285	M€/$MW_{th,dry,in}$
Investment CO_2 compression	800	850	900	€/kW
Investment CO_2 removal	0.011	0.014	0.017	M€/$MW_{th,syngas}$
Investment EF gasifier	0.075	0.084	0.092	M€/$MW_{th,dry,in}$
Investment Fischer-Tropsch synthesis	12	14	16	k$/bpd
Investment fuel gas compression	0.022	0.027	0.032	M€/$MW_{th,syngas}$
Investment methanation	30.8	41.1	51.3	M€/GW_{th}
Investment Press.O_2 blown CFB gasifier	0.080	0.100	0.190	M€/$MW_{th,dry,in}$
Investment steam system	428	547	667	k€/(kg/s)
Investment water-gas-shift	0.009	0.011	0.013	M€/$MW_{th,syngas}$
Plant capacity factor	0.85	0.90	0.95	-

APPENDIX B GASIFICATION

Gasification is a technology that converts solid fuel (such as biomass) into a gaseous fuel consisting mainly of hydrogen and carbon monoxide. After cleaning, the produced gas can be used as fuel gas for power generation, or as syngas in the (petro) chemical industry. In the latter case, the gas can also be used to produce liquid fuels or hydrogen (after additional reforming and shifting) or upgraded to substitute natural gas (after additional methanation and gas conditioning) and distributed through the existing gas infrastructure.

B.1 Circulated Fluidised Bed (CFB) gasifier

Three out of five gasifiers are based on Circulated Fluidised Bed gasification, either with air or oxygen as gasifying agent and operated either at atmospheric or elevated pressure (20 bar). The gasifying agent is fed to a riser at the bottom (primary) or at several axial positions (secondary). The biomass fuel is introduced just above the primary inlet of the gasifying medium. Before gasification the fuel has to be dried to 15% moisture and chipped to about 30 mm.

Oxygen versus air blown gasification
The application of oxygen instead of air avoids the dilution of syngas with nitrogen. As a result the heating value of the cooled and purified syngas is higher and the syngas volume is about half that for an air-blown unit for the same amount of biomass gasification energy, thus gas handling and cleanup requires smaller units. Furthermore, the sizes of the gasifier and the heat exchangers, required for the recovery of sensible heat from the syngas, are reduced. The main disadvantage of oxygen-blown gasification is that it has been far less developed than air-blown gasification and that production of oxygen is expensive.

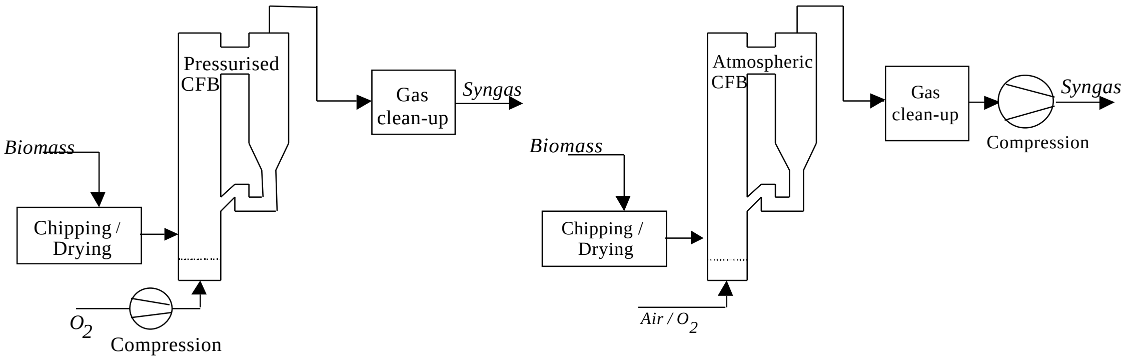

Figure 11 *Schematic presentation of a pressurised (left) and atmospheric CFB gasifier (right)*

Atmospheric versus pressurised gasification
The net cold gas efficiency of a pressurised gasification system decreases with increasing operating pressures. The overall system efficiency, however, will not be determined solely by the cold gas efficiency, but also by the electricity consumption of the system. Considering power generation from or upgrading of the gas from the gasifier (e.g. FT-synthesis or methanation to SNG) the syn- or product gas will be utilised at elevated pressures. By gasifying at elevated pressures, compression of the product gas from the gasifier is not necessary. On the other hand, compression of gasifier inlet gases, *e.g.* CO_2 or N_2 (for the compression of the biomass fed to the gasifier) and air or oxygen (as gasifying medium), will be required. The reduction of the electricity consumption will compensate for the reduction of cold gas

efficiency; hence, pressurised gasification will lead to a higher overall net efficiency of the process. A schematic presentation of a pressurised as well as an atmospheric CFB gasifier is presented in figure 11.

Nitrogen versus carbon dioxide pressurisation
For compression of the biomass fed to a pressurised gasifier both nitrogen and carbon dioxide can be used. The choice for either of the two may depend on the availability on a specific site. Both CO_2 and N_2 will dilute the product gas. However, CO_2 in contrast to N_2 can easily be removed from the gas thus allowing ‘upgrading’ of the gas.

B.2 Indirect gasifier

Within the indirect gasification concept the energy required by for the gasification process is obtained from the combustion of char in a secondary reactor. A schematic presentation of the process with global energy duties is presented in figure 12. Typical gasification temperatures are 800-900°C. The advantage of indirect gasification is that a nitrogen-free gas is produced. As the gasification and combustion are separated the N_2 of the air supplied to the reactor will end up in the flue gas and not in the product gas.

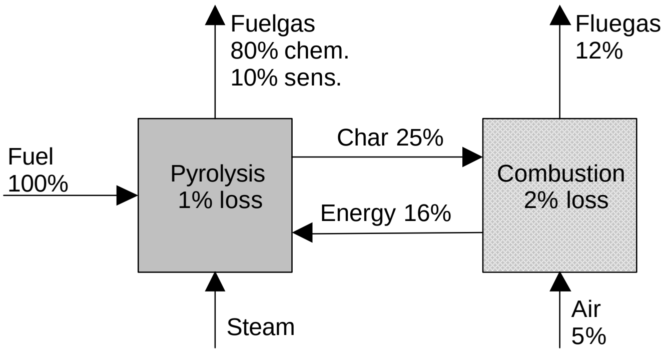

Figure 12 *Schematic diagram of the indirect gasification concept*

B.3 Entrained flow gasifiers

In contrast to the other gasifiers an entrained flow (EF) gasifier is operated without bed material (*e.g*. sand). The (pulverised) feed and the gasifying medium are supplied to the reactor at high velocity and pass the reactor in plug-flow while the biomass is gasified. Typically, oxygen is used instead of air as the gasifying medium, in order to achieve high conversion efficiencies despite the low residence time in the reactor. Similarly, the feed needs to be pulverised to less than 1 mm.

APPENDIX C SYNTHESIS PROCESSES

C.1 Fischer-Tropsch synthesis

C.1.1 Reactions

The Fischer-Tropsch (FT) synthesis was discovered in 1923 by F. Fischer and H. Tropsch at the Kaiser Wilhelm Institute for Coal Research in Mülheim, Germany [2]. 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) and minor amounts of olefins (alkenes) and alcohols; equation (9). The methylene group $–(CH_2)–$ shown in this equation is used to represent the hydrocarbon chain. About 20% of the initial chemical energy is released as heat in this exothermic reaction:

$$\mathrm{CO} + 2\mathrm{H}_2 \Rightarrow -(\mathrm{CH}_2)- + \mathrm{H}_2\mathrm{O} \tag{9}$$

Typical operation conditions for FT-synthesis, when aiming for long-chain products, are temperatures of 200-250°C and pressures of 25-60 bar [3]. Typical FT catalysts are based on iron or cobalt. 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, equation (10):

$$\mathrm{CO} + \mathrm{H}_2\mathrm{O} \Leftrightarrow \mathrm{CO}_2 + \mathrm{H}_2 \tag{10}$$

Iron-based FT catalysts show considerable WGS activity and the H_2/CO ratio is adjusted in the synthesis reactor. In case of cobalt-based catalysts the ratio needs to be adjusted prior to FT-synthesis. The polymerisation-like chain-growth FT reaction results in a range of products, comprising light hydrocarbons (C_1-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. The chain length distribution can be described by means of the Anderson-Schulz-Flory (ASF) equation, which is represented as [3]:

$$\log \frac{\mathrm{W_n}}{\mathrm{n}} = \mathrm{n} \cdot \log \alpha + \log \frac{(1-\alpha)^2}{\alpha} \tag{11}$$

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 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 13 [4].

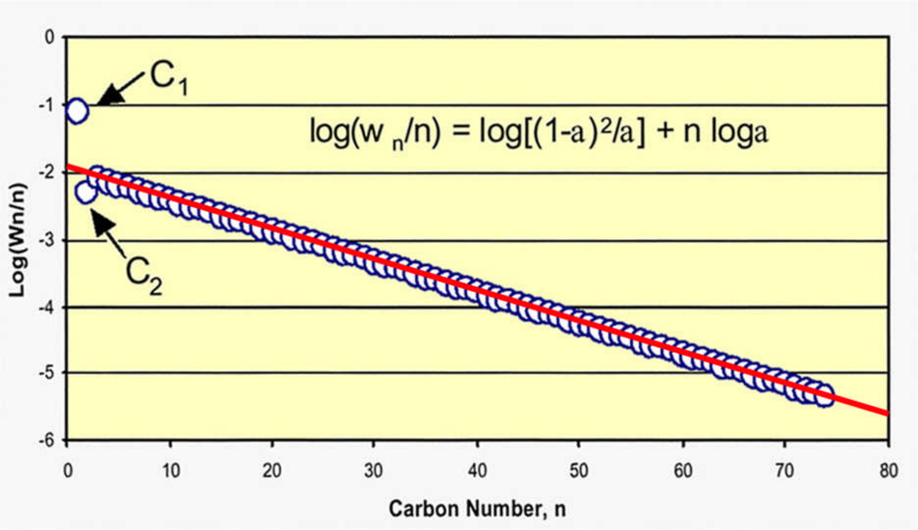

Figure 13 *ASF as well as experimental distribution of FT product yields*

In order to correct for this deviation, the C_1 to C_4 yields are calculated according to equations (12) and (13), whereas the yields of the higher hydrocarbons (C_{5+}) are calculated according to the ASF distribution, *i.e.* equation (11).

$$W_1 = \tfrac{1}{2}(1 - \sum_{i=5}^{\infty} W_i) \tag{12}$$

$$W_{2,3,4} = \tfrac{1}{6}(1 - \sum_{i=5}^{\infty} W_i) \tag{13}$$

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 additional hydrogen is required, which can be produced from a syngas side-stream that is completely shifted to hydrogen via the WGS reaction.

C.1.2 Economics

A detailed consideration of the economics of Fischer-Tropsch synthesis is a complex matter as all commercial implementations have been driven by either political (Germany, South Africa) or strategic (Shell) considerations rather than by straightforward economics [5]. The total investment for the first commercial Shell project is about $660 million for the 12,000 bbl/day Bintulu project [6], whereas a single module of the Sasol slurry phase distillate plant, that converts natural gas into 10,000 bbl/day of liquid transport fuels, costs about $250 million [7]. Several studies, however, mention that there is a large incentive to reduce capital cost of the Fischer-Tropsch process.

With an expected capacity of approximately 25,000 bbl/day (*i.e.* 1.75 GW_{th}[2]) and an assumed location factor of 1.25 [8] the specific costs of a Fischer-Tropsch installation are estimated to be between 30 and 40 k$/bpd with an expected value of 35 k$/bpd. These costs include oxygen plant and gas purification, synthesis gas generation, Fischer-Tropsch synthesis and product upgrade. The relative distribution of capital expenditure for the various phases is presented in

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[2] The lower heating value of Fischer-Tropsch liquids is estimated to be 36.3 MJ/litre or 5.66 GJ/barrel, with 1 barrel (bbl) equals to 159 litres

figure 14 [9]. According to this cost breakdown the actual Fischer-Tropsch synthesis and product upgrading make up for only 40% of the total plant costs, hence the capital expenditure of the Fischer-Tropsch synthesis and product upgrading will be between 12 and 16 k$/bpd with an expected value of 14 k$/bpd.

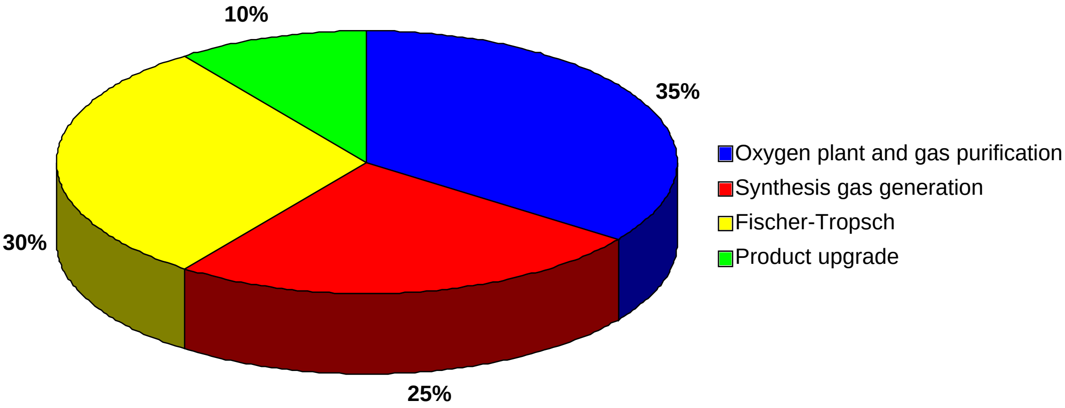

Figure 14 *Typical cost breakdown of the Fischer-Tropsch installation with natural gas feed stock*

The operational expenditure of the SMDS process is approximately $10 per barrel. These costs consist of fixed and variable operating costs as well as feedstock costs. According to Eilers et al. [6], the fixed and variable operating costs are approximately $4 per barrel. This amount corresponds well to the non-feedstock operating costs for gas-to-liquids plants mentioned by Gaffney, Cline & Associates ($4-$4.5 per barrel) [9] and by Sasol ($5 per barrel) [7]. Within this study, therefore, the operational expenditures will be estimated to be between 4 and 5.5 $/bbl with an expected value of $4.5 per barrel.

C.2 Methanation

C.2.1 Reactions

In the methanation step, carbon monoxide is converted to methane according to reaction:

$$CO + 3H_2 \Leftrightarrow CH_4 + H_2O \qquad (14)$$

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

$$CO + H_2O \Leftrightarrow CO_2 + H_2 \qquad (15)$$

The methanation reactions 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. The maximum yield of methane will be at low temperatures. At temperatures to 300°C a 99% conversion or better will be reached [10]. High pressures, up to 100 bar, can improve the conversion at higher temperatures.

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

$$2CO \Leftrightarrow CO_2 + C \qquad (16)$$

$CO + H_2 \leftrightarrow H_2O + C$ (17)

Also cracking reactions of alkenes and aromatic compounds can lead to carbon formation [11]. 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.

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. Nickel catalysts are extremely sensitive to sulphides and chlorides. Sulphur compounds affect the nickel catalyst through the reaction of hydrogen sulphide with nickel. Hydrogen chloride is a permanent irreversible poison to the methanation activity of nickel catalyst; an HCl/HF concentration of less than 25 ppb would be admissible for nickel catalysts [11].

C.2.2 Economics

In 2001 Polman et al. performed a study on the conversion of biomass into a sustainable gas by means of an internally cooled methanation reactor. Synthesis gas from either an atmospheric or pressurised oxygen-blown gasifier at 15 bar is (compressed to 15 bar) cleaned, methanated (at 15 bar), and dried in order to produce SNG suitable for the natural gas grid. The costs of the promotion of synthesis gas to natural gas quality were based on investment costs, maintenance costs (4% of the investment costs), costs of chemicals and of waste disposal [11].

Considering a 100 MW_{th} gasifier the capital costs presented by them are given in table 9. Besides the thermal biomass input, the thermal synthesis gas input of the gas upgrading section is also presented. The operational costs are presented in table 10. The CO_2 removal is based on membrane separation techniques. The thorough gas clean up is required mainly for the removal of sulphuric and halide elements. The consumption of electricity is almost completely linked to compression/expansion of gases. The costs of adding THT (tetra-hydro thiophene), used for odourisation of the SNG, are relatively low and are therefore not mentioned in the breakdown of the operational costs.

Table 9 *Capital costs of upgrading synthesis gas to SNG (Gastec/Kema)*

Capital costs	100 MW_{th} gasifier at 1.2 bar	100 MW_{th} gasifier at 15 bar
[M€]	*(78.88 MW_{th})*	*(79.78 MW_{th})*
Compression	3.6	-
Thorough gas clean-up	0.7	0.8
Methanation	2.6	2.8
Drying	1.1	1.2
CO_2 removal	1.6	1.8
Delivery	0.4	0.5
Total	***10.0***	***7.1***

Table 10 *Operational costs of upgrading synthesis gas to SNG (Gastec/Kema)*

Operational costs [M€]	100 MW_{th} gasifier at 1.2 bar *(78.88 MW_{th})*	100 MW_{th} gasifier at 15 bar *(79.78 MW_{th})*
Operation	0.1	0.1
Maintenance	0.4	0.3
Adsorbents	1.5	1.5
Catalyst	0.3	0.3
Electricity	1.7	-0.4
Waste disposal	0.1	0.1
Total	***4.0***	***1.8***

Within this study capital and operational expenditures (CAPEX and OPEX) of the upgrading will, as far as methanation, drying and gas delivery, be based on the costs mentioned by Polman *et al*. [11].

APPENDIX D UTILITIES

D.1 CO_2 removal

In order to obtain a SNG that matches the standard quality of natural gas, it might be necessary to remove carbon dioxide from the SNG. For the separation of carbon dioxide from methane, several technologies can be used [10], some selected technologies are briefly discussed in this section.

D.1.1 Membrane separation of CO_2

Gas separation by membranes is based on the differences in permeation coefficients of different gas components. Permeation denotes the sequence of absorption (solution) of the component in the membrane material, diffusion through the membrane, and desorption. Membranes may consist of ceramic or metallic materials, organic liquids, or polymers [12]. Since the upgraded gas with reduced CO_2 content is retained at the high-pressure side of the membrane, it leaves the upgrading process at high pressure, which means that the need for further compression prior to its introduction to a gas grid is reduced or eliminated. Typical operating pressures are in the range of 25 to 40 bar [10]. The efficiency of membrane separation varies between 73 and 83%, with an energy efficiency (energy input related to energy output) of approximately 10% [13]. The specific investment costs of membrane systems for syngas to SNG upgrading are approximately 215 to 270 €/($m_n^3{}_{SNG}$/h) [11]. As membrane separation will be a modular based process, cost reduction as a result of large scale processing is limited.

D.1.2 Absorption of CO_2

Numerous proven chemical and physical absorption processes are available for removing, among others, carbon dioxide, with an efficiency of approximately 95% [13]. Physical absorption processes are used predominantly to remove carbon dioxide from gases with high carbon dioxide partial pressure, especially converted gases produced in partial oxidation plants based on coal and residual oil. Here the partial pressure of carbon dioxide is above the level appropriate for economic application of chemical absorption systems. Chemical absorption processes (normal amine-based) are used widely, especially for crude gas containing carbon dioxide at low partial pressure. The selection of amine is influenced by specific process conditions such as the presence of further impurities in the raw gas.

D.1.3 Comparison of CO_2 removal techniques

Within the study on CO_2 capture from power stations the combined use of membranes and scrubbing solvents to remove CO_2 was highlighted [14]. In most cases it has been found extremely viable to operate such units in series, with the membrane removing the bulk of the gas and the chemical solvent cleaning up the trace quantities. Considering SNG production, it however will not be necessary to remove trace quantities of carbon dioxide, as they will not result in problems when adding the SNG to the natural gas grid. Hence, bulk removal of carbon dioxide will be sufficient and, therefore, membrane separation seems to be the most interesting option [11]. In case of FT-synthesis, maximum CO_2 removal is required at elevated pressure. As membrane systems usually cannot achieve high degrees of separation they will require multiple stages or recycling of one of the streams necessary. The multiplicity of required membrane modules will make the membrane system less and physical absorption processes more interesting [15].

D.2 Oxygen production

In the late 1800s, various attempts were made to separate oxygen from air on a commercial scale. These all relied on chemical processes employing thermal cycling. Nowadays mainly three technologies for commercial oxygen production are commercially available, *i.e.* cryogenic separation, pressure-swing adsorption, and membrane air separation. Cryogenic air separation is the O_2 production technique commonly used when large capacities are required as well as the technique that enables production of highly pure oxygen (>99.5%).

Cryogenic separation can be applied technology for oxygen production up to 2250 tonnes of gaseous oxygen per day and production costs of 35-75 $/tonne of oxygen. Cryogenic air separation involves three steps:

1. Purification of the incoming air to remove particles, carbon dioxide, and water;
2. Refrigeration and economisation of refrigeration values contained in the product and waste streams;
3. Separation by distillation.

Filtered air compressed to 6-7 bar is cooled and scrubbed in a direct-contact water wash tower (a) and then directed into a plate-fin reversing heat exchanger (b), where it is cooled further in counter-flow to product oxygen and waste nitrogen (figure 15). Carbon dioxide and water are removed from the air stream by condensation and solidification in this heat exchanger. Every few minutes, the paths of the incoming air and the waste nitrogen streams are reversed so that deposited impurities can be purged from the exchanger. In this way, the process is able to operate continuously on a cyclic basis. State-of-the-art plants are, however, being constructed with molecular sieve adsorption units, in place of the reversing heat exchangers. After carbon dioxide and water removal, the incoming air is cooled by cold waste gas in counter-flow heat exchangers.

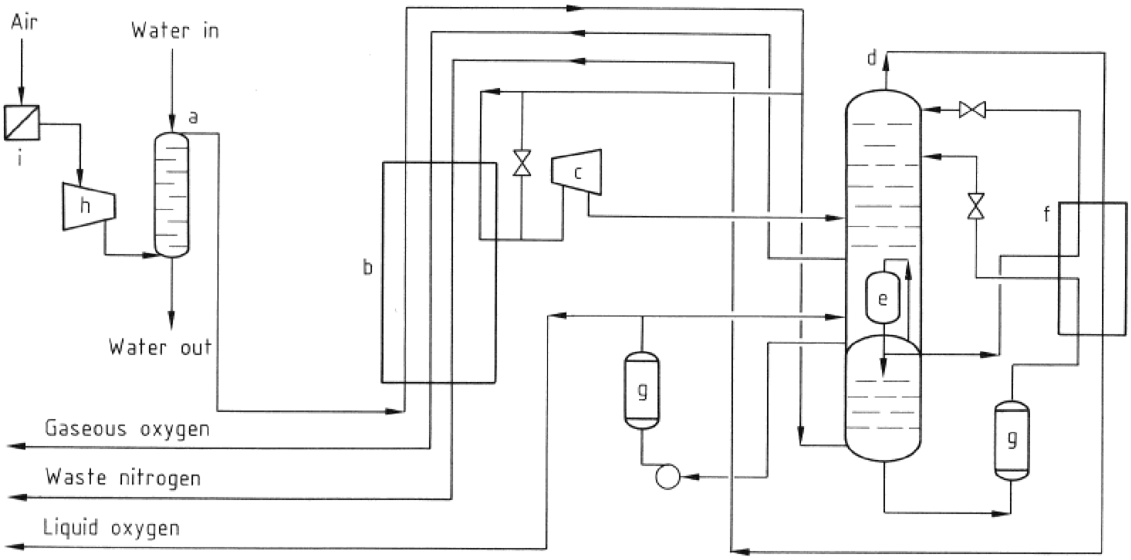

Figure 15 *Cryogenic air separation plant*

Refrigeration for this process is achieved primarily by the Claude principle of conversion of energy into work by the expansion of a compressed gas in a turbine. Some of the cooled air is returned through the cold end of the heat exchanger before expansion to ca. 1.2 bar in a turbine (c), and then introduced at an appropriate point in the upper, low-pressure column of the double column rectifier (d). The remainder of the air passes into the lower, high-pressure column at 5-6 bar where it is separated into nitrogen gas at the top and oxygen-enriched liquid (38% O_2) at the bottom. Gaseous nitrogen is condensed by liquid oxygen (which is vaporised) in the condenser-reboiler (e). Part of this liquid nitrogen is returned as reflux to the lower column, and part is expanded into the top of the upper column as reflux liquid. The oxygen-enriched liquid (rich liquid) from the sump of the lower column is expanded as feed into an appropriate point in the upper column. The reflux liquids used are sub-cooled by returning nitrogen in the heat exchanger (f), to reduce the amount of liquid vaporised on expansion. In the upper column, the feed is separated into pure oxygen (>99.5%) at the bottom and waste nitrogen with 1-2% oxygen at the top. The adsorber (g), packed with silica gel, removes trace hydrocarbon matter from liquid oxygen in the condenser-reboiler and from the rich liquid, ensuring that unsafe hydrocarbon concentrations do not accumulate in the process. In table 11 the most important parameters of cryogenic air separation are presented [16].

Table 11 *General cryogenic air separation parameters*

Cryogenic air separation	118.9 kg/s O_2(g)
Power requirement	135.1 MW_e
Outlet pressure O_2(g)	1 atm.
Outlet pressure N_2(g)	1 atm.
Investment costs [17]	280 ~ 420 M€
Scale O_2-plant (more trains)	9000 tonnes O_2/day

As the gaseous oxygen is obtained at atmospheric pressure and is required at elevated pressure, additional costs concerning the compression of oxygen have to be taken into account. The costs of compression of oxygen from 1 atmosphere to either 20 or 40 bar are presented in table 12.

Table 12 *Costs of compression of oxygen*

Oxygen compression		1.0 kg/s O_2(g)
Compression 1 atm. •	20 bar[3]	0.55 MW
Compression 1 atm. •	40 bar	0.77 MW
Investment costs [18]		850 €/kW

D.3 HRSG section with steam turbine

Within the Heat Recovery Steam Generation section (HRSG) flue gases are cooled as far as possible in order to produce a maximum amount of steam. The superheated steam, produced in the HRSG, is expanded in a partly condensing steam turbine, producing an amount of electricity in a turbine-coupled generator. The expanded steam, mainly vapour, is subsequently condensed completely in a surface water-cooled condenser. After de-aerating of the water, it is compressed to a pressure of 66 bar by water pumps, and fed to the boiler feed water heater in the HRSG. As steam is withdrawn at several pressure levels for shifting purposes, the HRSG will require a certain amount of fresh water. Within a study on long term perspectives of biomass integrated gasification with combined cycle technology the costs of a HRSG section with steam turbine were presented for state of the art, as well as advanced systems at three different scales, as presented within table 13.

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[3] Compression power has been calculated within the Aspen Plus flow sheet simulation package

Table 13 *Investment costs of the HRSG section with steam turbine*

Investment costs [M€]	Amount of steam		
	13.2 kg/s	*26.4 kg/s*	*44.4 kg/s*
HRSG	2.4 ~ 3.3	4.4 ~ 6.0	8.8 ~ 11.9
Steam turbine & condenser	3.2 ~ 4.3	5.4 ~ 7.4	9.9 ~ 13.4
Water & steam system	0.3 ~ 0.4	0.5 ~ 0.7	1.1 ~ 1.5
Cooling	0.7 ~ 0.9	0.9 ~ 1.2	1.1 ~ 1.5
Total	***6.6 ~ 8.8***	***11.3 ~ 15.2***	***21.0 ~ 28.3***

The cost estimations on HRSG and steam turbine within this study are based on these cost figures, with a minimum, maximum and expected value of the total investment costs of 428, 667 and 547 k€ per kg/s steam.

APPENDIX E DUTCH NATURAL GAS INFRASTRUCTURE & REQUIREMENTS

In the Netherlands, Gasunie is responsible for the gas transport and providing the gas to the customers, *viz*. the Dutch gas supply companies, large industrial consumers, and for gas export to some European countries. Gasunie [19] buys natural gas from a large number of producers that supply the gas via feeding stations at a pressure of around 65 bar. The main supplier is NAM (Nederlandse Aardolie Maatschappij), the company that operates the 29 well clusters on the Groningen field. Additionally, a small volume of gas is imported from Norway.

E.1 Transmission infrastructure

The Gasunie transmission system comprises approximately 11,000 km of natural gas pipelines plus plants and equipment for compressing, blending, metering, and regulating the different gas flows (see figure 16 [20]). The pipeline diameters range from 45 up to 120 cm. The pipes are made of various grades of steel and a cathodic protection system against corrosion.

High-pressure Transmission Lines
Gasunie's High-pressure Transmission Lines (HTL) network has a length of 5,000 km. It comprises 48-inch pipes, carrying gas at pressures in the range 43 to 66 bar, and occasionally as high as 80 bar. The gas enters the system at either producers' gas conditioning stations on the gas fields, or at import stations on the border. Nine compressor stations maintain the pipeline pressure at every 80-100 km in the HTL network. HTL end points are the metering and regulating stations (M&R), which form the link between the HTL and the Regional Transmission Lines (RTL) networks, and the export stations.

Regional Transmission Lines
Gasunie's RTL network has a total length of 6,000 km. The operating pressure generally ranges from 16 to 40 bar. The RTL network is supplied from the HTL network via the M&R stations. At M&R stations, the pressure in the HTL system is reduced to an operating pressure of not more than 40 bar. Another function performed at the M&R stations is to give the gas its characteristic smell, *i.e*. odourisation of the odour-free gas by adding tetrahydrothiophene (THT) at a rate of 18 mg/m^3. Odourisation is mainly for the benefit of the domestic consumer. Heavier business and institutional users will often have their own gas detection systems. M&R stations also supply measurement data relating to pressure and flow, which are vital for the control of the gas flows in the network.

The gas networks of the gas supply companies operate at pressures of 8 bar or lower. The pressure drop of more than 30 bar (RTL stations), and as much as 60 bar (HTL stations) is accompanied by a substantial drop in temperature. To prevent exit temperatures from falling too low, the gas is preheated. By the time the gas enters domestic gas pipes, the relative pressure is down to 25 millibar, the standard appliance pressure in the Netherlands.

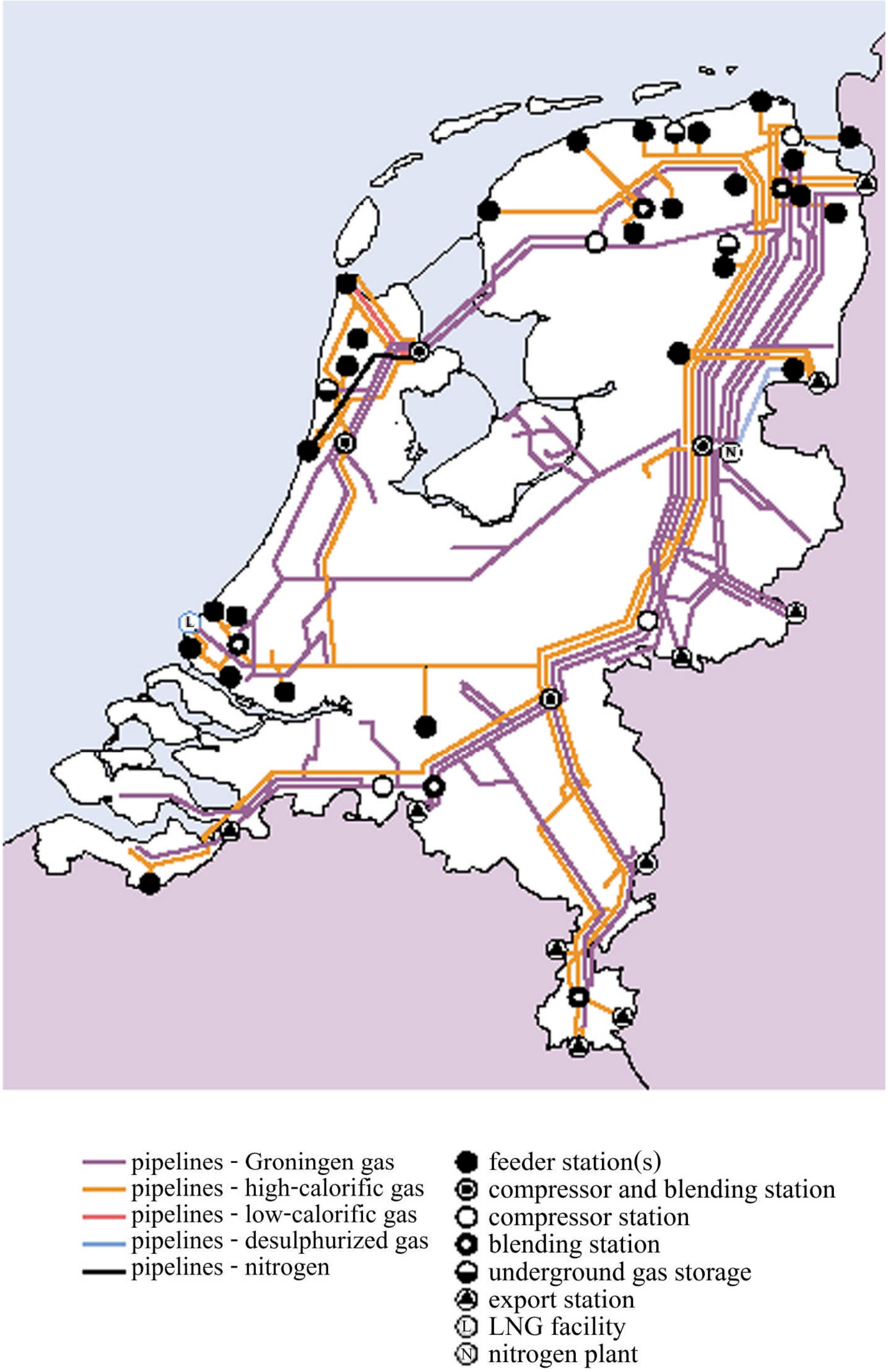

Figure 16 *The main gas transmission system (end 2000)*

E.1.1 Specifications

The supplied gas has to meet strict specifications, regarding composition, Wobbe-index, calorific value, and relative density. Fifteen blending stations mix gas streams from different sources to produce the desired qualities. The specification of natural gas with Groningen quality (G-gas) is presented in table 14.

Table 14 *Composition of natural gas with Groningen quality*

		Natural gas [21]
CH_4	[mol.%]	81.30
H_2	[mol.%]	0.00
CO_2	[mol.%]	0.89
C_2	[mol.%]	2.85
C_3	[mol.%]	0.37
C_4^+	[mol.%]	0.23
N_2	[mol.%]	14.35
O_2	[mol.%]	0.01
H_2O	[mol.%]	Dew-point < -10ºC
CO	[mol.%]	0.00
Wobbe	[MJ/m_n^3]	43.70

Not all natural gas is the same as Groningen gas. In the North Sea large amounts of high-calorific gas are released during the extraction of mineral oil, as associated gas, with high concentration of high hydrocarbons. Also many of the small fields contain gas of high calorific value (H-gas), while others contain gas of low calorific value (L-gas), rich in carbon dioxide [22]. Since the gas appliances of nearly all Dutch consumers are designed for G-gas, various facilities are needed in order to supply the different groups of customers with the required gas quality:

- To serve large industrial users, whose installations could be converted to operation on H-gas, an extra system of main transmission pipelines was built;
- H-gas and L-gas were mixed to produce a gas of Groningen quality, which could be introduced into the main transmission system for G-gas;
- H-gas was diluted with nitrogen to achieve Groningen quality before introduction into the G-gas pipelines.[4]

E.2 Potential & requirements for SNG transport

As the produced SNG would be transported through the natural gas net, it has to satisfy the requirements concerning the Wobbe-index and composition that are set to the G-gas. The specification of the G-gas delivered by Gasunie to energy distribution companies is presented in table 15 [10,11]. Also the requirements concerning the SNG, according to Gastec study [11], are given in this table.

The quality of Groningen natural gas is related to a Wobbe-index of about 43.7 MJ/m_n^3. The SNG should also have a composition resulting in a comparable Wobbe-index. Furthermore, 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 sensitivity to corrosion [11]. 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 [23], and therefore, corrosion problems [19].

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[4] Groningen gas itself contains no less than 14.35% nitrogen.

Table 15 *Specification of the G-gas delivered by Gasunie to energy distribution companies and the requirements concerning SNG, according to Gastec as well as to the Danish and Swedish centre*

Component or quality		G-gas	SNG [11]	SNG [10]
Wobbe-index	[MJ/m_n^3]	43.46< W <44.41	43.46< W <44.41	*
H_2	[mol%]	-	<12	<12
CO	[mol%]	-	<0.8	<1
O_2	[mol%]	<0.5 (wet net)	<2.5 (dry net)	<0.5 (wet) & <4 (dry)
C_2H_4	[mol%]	-	conform Wobbe	-
BTX (incl. naftalene)	[ppm]	<500	<500	-
Aromatic comp.	[g/m_n^3]	-	-	<10
HCl	[ppm]	-	<1	-
NH_3		-	<1 ppm	<3 NH_3
HCN		-	<10 ppm	<150 mg/m_n^3
H_2O		Dew-point < -10ºC	Dew-point < -10ºC	Not any liquid at any P or T in the grid
H_2S	[mg/m_n^3]	<5	<2	<6
Mercaptanes	[mg/m_n^3]	<10.0	<5.0	<5
S total	[mg/m_n^3]	<45	<25	<120
Hg	[mg/m_n^3]	0	<0.01	-
Dust	[mg/m_n^3]	0	<0.5	0
THT	[mg/m_n^3]	>10 and <30	>10 and <30	-

** The Wobbe-index will be dependent on the reference natural gas. Nearly all natural gases in Europe are within high quality limits (between 46 and 57), however Dutch natural gas is within low quality limits (between 38 and 47)*

APPENDIX F BIOMASS CONVERSION EFFICIENCIES

In this appendix the plots (in figure 17 through figure 20) are presented for the conversion efficiencies from biomass to FT liquids and SNG as function of the CO conversion (η_C) in the FT synthesis and the α ASF product distribution parameter. The FT liquids are subdivided into the C_5-C_{10} fraction and the C_{11}+ fraction. Furthermore, a plot over the summarised yield is presented. N.B. the gasification efficiencies are incorporated. The data are summarised in table 16.

Results for co-production with four gasification processes are presented:

- Indirect gasification (atmospheric) [indirect];
- Atmospheric oxygen-blown CFB gasification [A-CFB(O_2)];
- Pressurised (with carbon dioxide) oxygen-blown CFB gasification [P_C-CFB(O_2)];
- Entrained-flow gasification (pressurised and oxygen-blown) [EF].

Table 16 *Specific biomass conversion efficiencies*

	$\eta_{overall}$	η_{SNG}	η_{C5-C10}	η_{C11+}
Indirect gasification				
η_c=0	71.2	71.2	0.0	0.0
α=0.95, η_c=0.95	71.4	45.5	2.1	23.7
α=0.5, η_c=0.95	71.5	66.0	5.3	0.2
Atmospheric O_2-blown gasification				
η_c=0	68.9	68.9	0.0	0.0
α=0.95, η_c=0.95	69.1	31.7	3.1	34.3
α=0.5, η_c=0.95	69.3	61.4	7.7	0.2
Pressurised O_2-blown gasification				
η_c=0	72.8	72.8	0.0	0.0
α=0.95, η_c=0.95	73.0	53.2	1.6	18.2
α=0.5, η_c=0.95	73.0	68.8	4.0	0.1
Entrained Flow gasification				
η_c=0	58.3	58.3	0.0	0.0
α=0.95, η_c=0.95	58.7	3.3	4.6	50.8
α=0.5, η_c=0.95	58.9	47.2	11.3	0.4

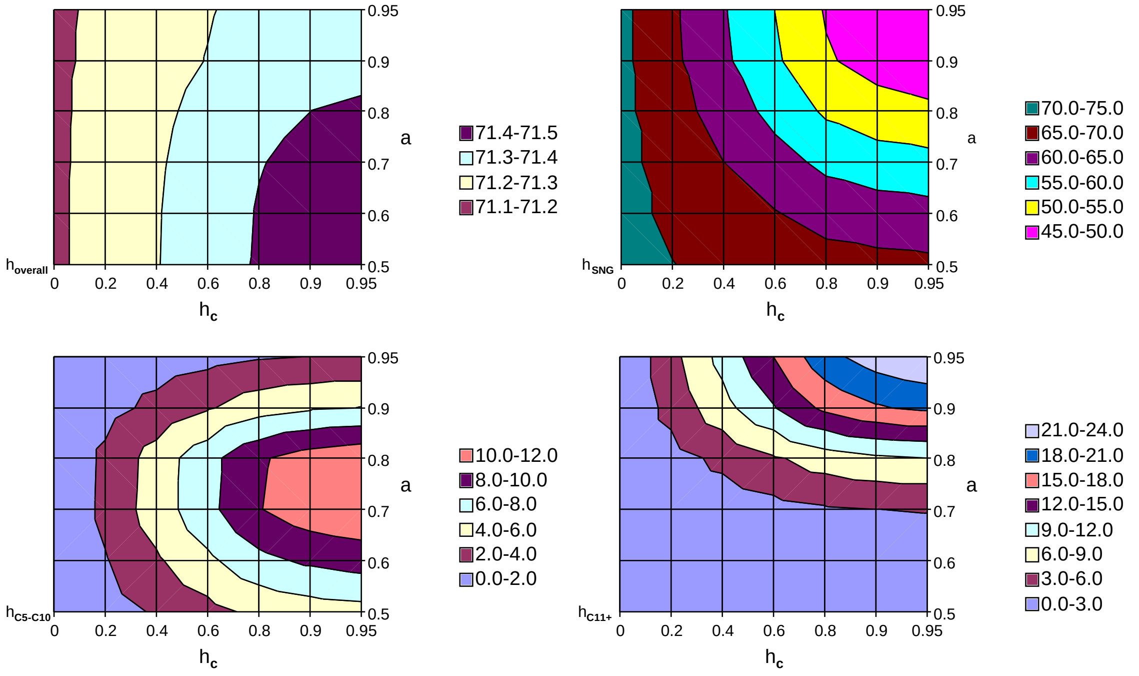

Figure 17 *Efficiencies in case of co-production with indirect gasification*

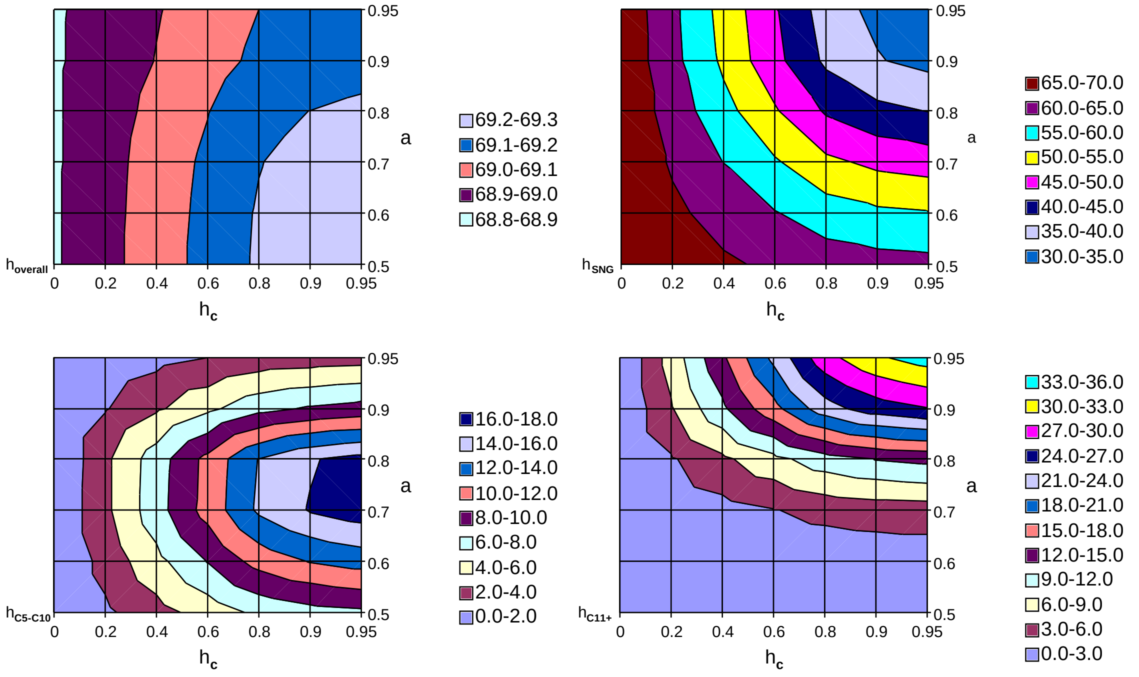

Figure 18 *Efficiencies in case co-production with atmospheric O_2-blown CFB gasification*

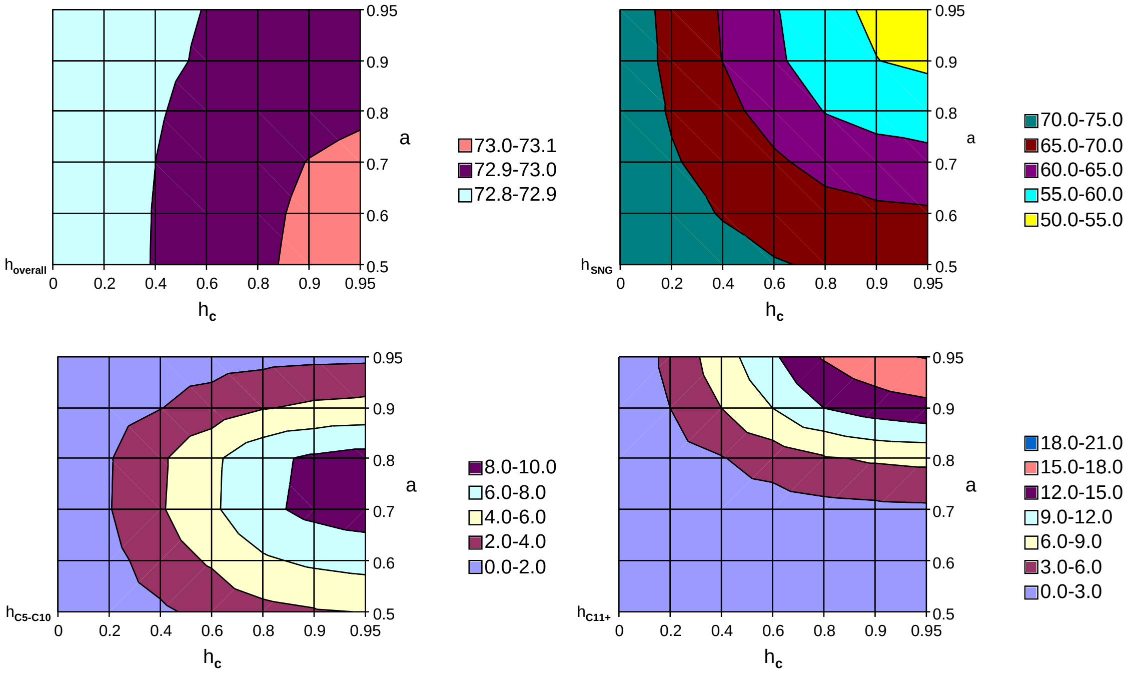

Figure 19 *Efficiencies in case of co-production with pressurised O_2-blown CFB gasification*

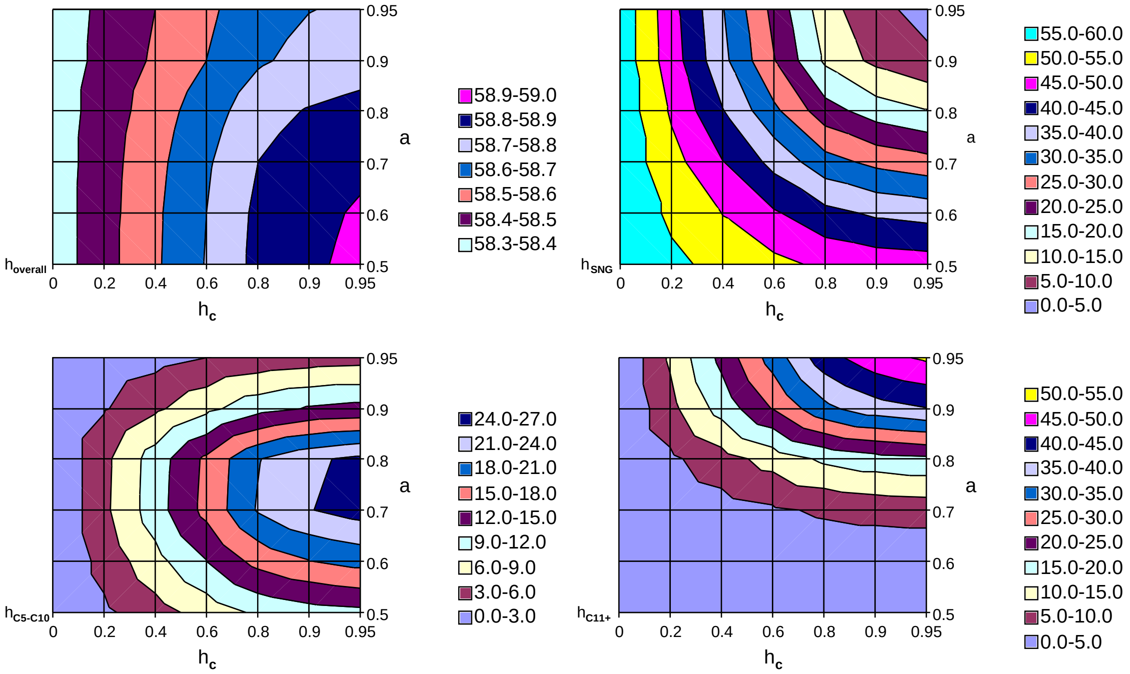

Figure 20 *Efficiencies in case of co-production with Entrained Flow gasification*

APPENDIX G SIMPLIFIED ENERGY BALANCES

The flow sheets for the evaluated integrated and parallel co-production concepts are presented. These concepts are:

- Integrated co-production with indirect gasification;
- Integrated co-production with pressurised oxygen-blown gasification;
- Integrated co-production with entrained flow gasification;
- Co-production with entrained flow gasification and parallel indirect gasification;
- Co-production with entrained flow gasification and parallel pressurised oxygen-blown gasification.

G.1 Indirect gasification

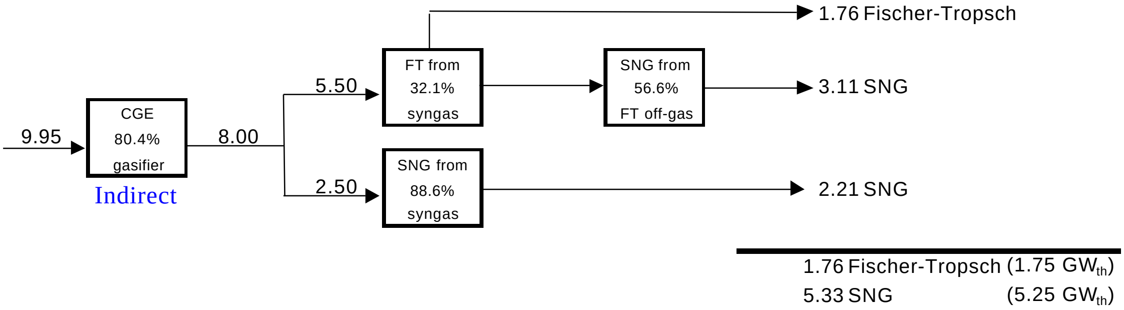

Figure 21 *Co-generation by indirect gasification with additional production of SNG in an integrated methanation step.*

G.2 Pressurised O_2-blown gasification

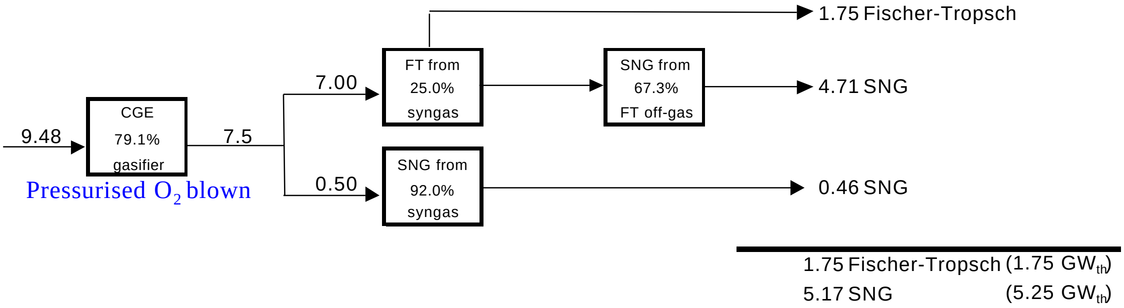

Figure 22 *Co-production by pressurised oxygen-blown gasification with additional production of SNG in an integrated methanation step.*

G.3 Entrained flow gasification

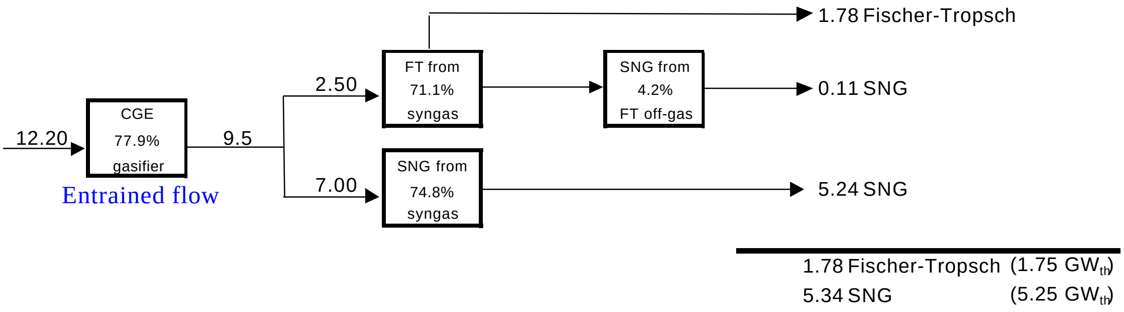

Figure 23 *Co- production by entrained flow gasification with additional production of SNG in an integrated methanation step*

G.4 Entrained flow gasification & Indirect gasification

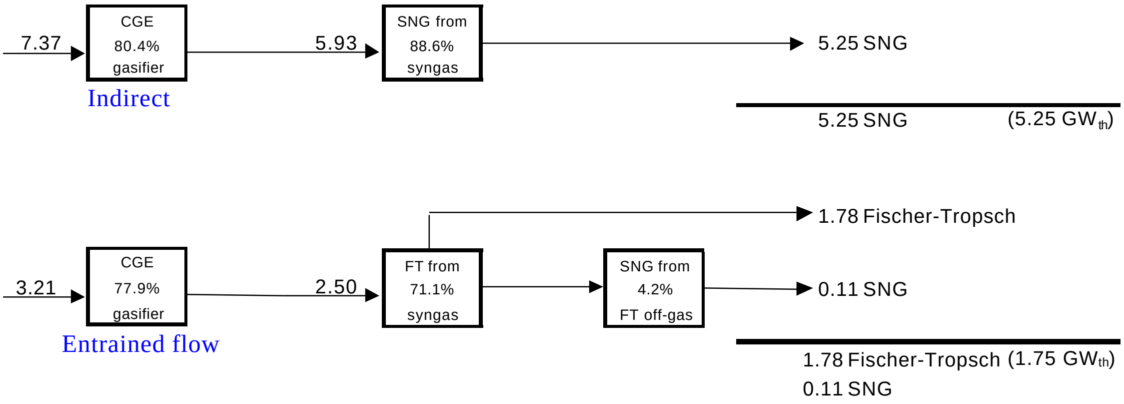

Figure 24 *Co-production by entrained flow gasification combined with parallel production of SNG by indirect gasification*

G.5 Entrained flow gasification & Pressurised O_2-blown gasification

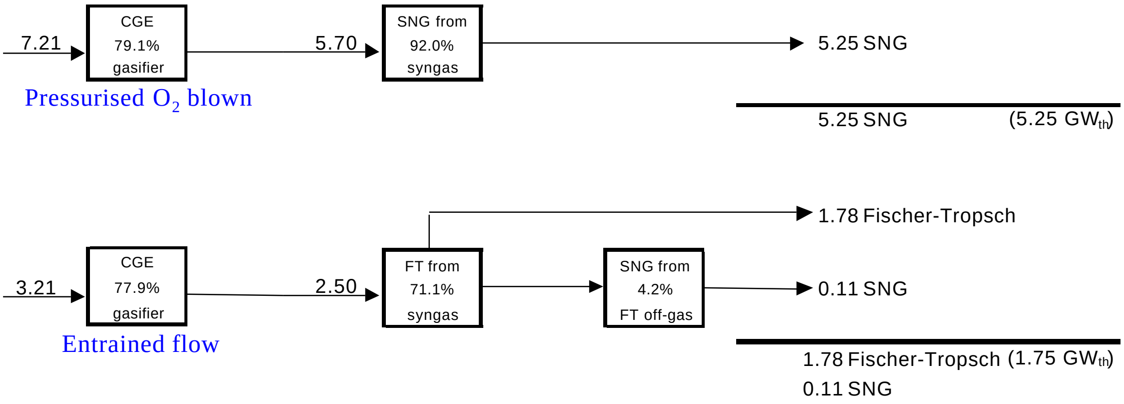

Figure 25 *Co-production by entrained flow gasification combined with parallel production of SNG by pressurised O_2-blown gasification*

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