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Future prospects for production of methanol and hydrogen from biomass

Carlo N. Hamelinck^{*}, André P.C. Faaij

Department of Science, Technology and Society, Utrecht University, Centrumgebouw-noord Padualaan 14, de Uithof, 3584 CH 14 Utrecht, The Netherlands

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Abstract

Technical and economic prospects of the future production of methanol and hydrogen from biomass have been evaluated. A technology review, including promising future components, was made, resulting in a set of promising conversion concepts. Flowsheeting models were made to analyse the technical performance. Results were used for economic evaluations. Overall energy efficiencies are around 55% HHV for methanol and around 60% for hydrogen production. Accounting for the lower energy quality of fuel compared to electricity, once-through concepts perform better than the concepts aimed for fuel only production. Hot gas cleaning can contribute to a better performance. Systems of 400 MW_{th} input produce biofuels at US\$ 8–12/GJ, this is above the current gasoline production price of US\$ 4–6/GJ. This cost price is largely dictated by the capital investments. The outcomes for the various system types are rather comparable, although concepts focussing on optimised fuel production with little or no electricity co-production perform somewhat better. Hydrogen concepts using ceramic membranes perform well due to their higher overall efficiency combined with modest investment. Long-term (2020) cost reductions reside in cheaper biomass, technological learning, and application of large scales up to 2000 MW_{th}. This could bring the production costs of biofuels in the US\$ 5–7/GJ range. Biomass-derived methanol and hydrogen are likely to become competitive fuels tomorrow.

Keywords: Biomass; Biofuels production; Fuel cell vehicles

1. Introduction

Methanol and hydrogen produced from biomass are promising carbon neutral fuels. Both are well suited for use in fuel cell vehicles (FCVs) which are expected to reach high efficiencies, about a factor 2–3 better than current internal combustion engine vehicles (ICEVs). In addition, they are quiet and clean, emitting none of the air pollutants SO_x, NO_x, VOS or dust. When methanol and hydrogen are derived from sustainably grown biomass, the overall energy chain can be greenhouse gas neutral. Such a scheme could provide a major alternative for the transport sector world-wide in a greenhouse gas constrained world [1–3].

Methanol and hydrogen can be produced from biomass via gasification. Several routes involving conventional, commercial, or advanced technologies, which are under development, are possible. Methanol or hydrogen production facilities typically consist of the following basic steps (see Fig. 1): pre-treatment, gasification, gas cleaning, reforming of higher hydrocarbons, shift to obtain appropriate H_2 :CO ratios, and gas separation for hydrogen production or methanol synthesis and purification. Optional are

a gas turbine or boiler to employ the unconverted gas, and a steam turbine; resulting in electricity co-production.

However, many process configurations are possible. Gasification can be atmospheric or pressurised, direct or indirect, resulting in very different gas compositions; different options are available for gas cleaning, processing and purification; generation of power is optional. Altogether in theory a very large number of concepts to produce methanol or hydrogen is possible.

Previous analyses by Katofsky [1] and Williams et al. [4] have shown that methanol can be produced from biomass at US\$¹ 14–17/GJ (biomass delivered at US\$ 2.3/GJ), with a net HHV energy efficiency between 54 and 58%. Hydrogen production costs can be US\$ 10–14/GJ, with a net HHV energy efficiency of 56–64%. Those cost levels are not competitive with current gasoline and diesel production costs (about US\$ 4–6/GJ [5]). The evaluations of Katofsky and Williams focused on technologies that are likely to be commercial on the short term. The scale of production was fixed on about 400 MW_{th}. Komiyama et al. [6] calculate hydrogen from biomass to cost US\$ 5.1/GJ and methanol US\$ 5.2/GJ; the biomass input is about 530 MW_{th} HHV, however, a significant amount of energy is added as LPG

^{*} Corresponding author. Tel.: +31-30-253-7686; fax: +31-30-253-7600. *E-mail address:* c.n.hamelinck@chem.uu.nl (C.N. Hamelinck).

¹ All costs are in US\$₂₀₀₁.



Fig. 1. Key components in biomass to methanol/hydrogen production concepts.

and process efficiencies and biomass cost are not given. All this leads to the key question whether advanced, future technologies, larger scales and alternative concepts may enable competitive production of methanol and hydrogen on longer term.

Therefore, the key objective of this work is to identify biomass to methanol and hydrogen conversion concepts that may lead to higher overall energy efficiencies and lower costs on longer term. Improved performance may be obtained by:

- Applying improved or new (non-commercial) technologies. Examples are the use of autothermal reforming (instead of steam reforming), improved shift processes, once through liquid phase MeOH process, high temperature gas cleaning, high temperature hydrogen separation and improved oxygen production processes.
- Combined fuel and power production by so-called 'oncethrough' concepts. Combined fuel and power production may lead to lower cost and possibly higher overall thermal efficiencies because of cheaper reactor capacity and reduction of internal energy consumption of the total plant.
- Economies of scale; various system analyses have shown that the higher conversion efficiencies and lower unit capital costs that accompany increased scale generally outweigh increased energy use and costs for transporting larger quantities of biomass. Furthermore, it should be noted that paper & pulp mills, sugar mills, and other facilities operate around the world with equivalent thermal inputs in the range of 1000–2000 MW_{th}. Such a scale could therefore be considered for production of energy/ fuel from (imported) biomass as well.

These strategies are explicitly taken into account in the present work:

- 1. Technology assessment (Section 2) and selection of various concepts (Section 3). The review includes technologies that are not applied commercially at present.
- Consulting of manufacturers and experts to obtain or verify performance and cost data of various components.
- 3. Creation of Aspen+ models to evaluate performance of the selected process configurations, and carry out sensitivity analyses. Particular attention is paid to the heat integration of the concepts (Section 4).
- Cost analyses based on component costs; including scale factors and capacity ranges (Section 5).
- 5. The work is finalised by an overall discussion and conclusion (Section 6).

2. Production of biofuels

2.1. Production and conditioning of synthesis gas

Syngas, a mixture of CO and H₂, is needed to produce methanol or hydrogen. A train of processes to convert biomass to required gas specifications precedes the methanol reactor or hydrogen separation—as was depicted in Fig. 1.

2.1.1. Gasification

Many gasification methods are available for syngas production. Based on throughput, cost, complexity, and efficiency issues, only circulated fluidised bed gasifiers are suitable for large-scale fuel gas production. Direct gasification with air results in nitrogen dilution, which in turn strongly increases downstream equipment size. This eliminates the TPS and Enviropower gasifiers, which are both direct air blown. The MTCI gasifier is indirectly fired, but produces a very wet gas and the net carbon conversion is low. Two gasifiers are selected for the present analysis: the Institute of Gas Technology (IGT) pressurised direct oxygen fired gasifier, and the BCL (Battelle Columbus) atmospheric indirectly fired gasifier. The IGT gasifier can also be operated in a maximum hydrogen mode, by increasing the steam input. Both gasifiers produce medium calorific gas, undiluted by atmospheric nitrogen, and represent a very broad range for the H₂:CO ratio of the raw fuel gas.

The main performance characteristics of both gasifiers are given in Tables 1 and 2. The IGT gasifier produces a CO₂ rich gas. The CH₄ fraction could be reformed to hydrogen, or be used in a gas turbine. The H₂:CO ratio (1.4:1) is attractive to produce methanol, although the large CO₂ content lowers the overall yield of methanol or hydrogen. For hydrogen production, the H₂:CO ratio should be shifted. The pressurised gasification allows a large throughput per reactor volume and diminishes the need for pressurisation downstream, so less overall power is needed. The maximum hydrogen mode is especially useful for hydrogen production, and also the H₂:CO ratio is still better for methanol production. However, the gasifier efficiency is lower and much more steam is needed. In both modes the IGT uses oxygen to reduce downstream equipment size.

The indirectly heated BCL is fired by air; there is no risk of nitrogen dilution nor need for oxygen production. It produces a gas with a low CO_2 content, but contains more heavier hydrocarbons. Therefore, reforming is a logical subsequent step in order to maximise CO and H₂ production. The tars present need to be cracked and the large CO fraction needs to be shifted to yield hydrogen. The reactor is fast fluidised, allowing throughputs equal to the bubbling fluidised IGT, despite the atmospheric operation. The atmospheric operation decreases cost at smaller scale, and the BCL has some commercial experience (demo in Burlington USA, [7]).

2.1.2. Gas cleaning

The produced gas contains tars, dust, alkali compounds and halogens, which can block or poison the catalysts downstream, or corrode the gas turbine. The gas can be cleaned using available conventional technology, by applying gas cooling, low temperature filtration, and water scrubbing at 100–250 °C. Alternatively, hot gas cleaning can be considered, using ceramic filters and reagents at 350– 800 °C. The considered pressure range is no problem for either of the technologies. Hot gas cleaning is advantageous

Table 1		
Key characteristics	of selected	gasifiers

	IGT ^a bubbling	IGT max H_2^b bubbling	BCL ^c indirectly heated
	Inulaised Dea	Indiased bed	last huidised bed
Initial moisture content (%)	30	30	30
Dry moisture content (%)	15	15	10
Steam (kg/kg dry feed)	0.3	0.8	0.019
Oxygen (kg/kg dry feed)	0.3	0.38	0
Air (kg/kg dry feed)	0	0	2.06
Product temperature (°C)	982	920	863
Exit pressure (bar)	34.5	25	1.2
Gas yield (kmol/dry tonne)	82.0	121	45.8
Composition: mole fraction on wet	basis (on dry basis)		
H ₂ O	0.318	0.48	0.199
H_2	0.208 (0.305)	0.24 (0.462)	0.167 (0.208)
CO	0.15 (0.22)	0.115 (0.221)	0.371 (0.463)
CO ₂	0.239 (0.35)	0.16 (0.308)	0.089 (0.111)
CH ₄	0.0819 (0.12)	0.005 (0.009)	0.126 (0.157)
C_2H_4	0.0031 (0.005)	0	0.042 (0.052)
C_2H_6	0	0	0.006 (0.0074)
O ₂	0	0	0
N_2	0	0	0
LHV _{wet} syngas (MJ/N m ³)	6.70	3.90	12.7
Cold gas efficiency (%)	HHV 82.2/LHV 78.1	HHV 72.1/LHV 60.9	HHV 80.5/LHV 82.5

^a [50] quoted by [4].

^b [51].

^c Compiled from [52] and [53] by [4].

for the overall energy balance when a reformer or a ceramic membrane is applied directly after the cleaning section, because these processes require a high inlet temperature. However, not all elements of hot gas cleaning are yet proven technology, while there is little uncertainty about the cleaning effectiveness of low temperature gas cleaning. Both cleaning concepts are depicted in Fig. 2.

In low temperature wet cleaning [1,8–14], particulates are completely removed by the cyclone, the bag filter and the

scrubbers. Essentially, all alkali and the bulk of sulphuric and nitrogenous compounds are removed by consecutive scrubbers. The ZnO bed or solvent absorption unit brings the sulphur concentration below 0.1 ppm. The effectiveness of cold gas cleaning has been proven for coal gasification combined cycle and Fischer–Tropsch synthesis applications [12]. Hot gas cleaning [1,9,12,13,15–22] removes particles for about 99.8% by granular beds and ceramic candle filters. Simultaneously SO_x and NO_x are removed by injection of

 Table 2

 Selected methanol and hydrogen production concepts

Methanol					
Gasifier	Gas cleaning	Reforming	Shift	MeOH	Power generation
IGT-max H ₂	Wet	_	-	Liquid phase	Combined cycle
IGT	Hot (550 °C)	ATR	-	Liquid phase, with steam addition	Combined cycle
IGT	Wet	-	_	Liquid phase, with steam addition	Combined cycle
BCL	Wet	SMR	_	Liquid phase, with steam addition and recycle	Steam cycle
IGT	Hot (550 °C)	ATR	Partial	Conventional solid bed, with recycle	Steam cycle
BCL	Wet	SMR	Partial	Conventional solid bed, with recycle	Steam cycle
Hydrogen					
Gasifier	Gas cleaning	Reforming	Shift	H ₂ separation	Power generation
IGT	Hot (350 °C)	_	Dual	PSA	Combined cycle
IGT-max H ₂	Hot (800 °C)	_	_	Ceramic membrane + internal shift	Purge gas expansion
IGT	Hot (350 °C)	-	_	Ceramic membrane + internal shift	Combined cycle
BCL	Wet	SMR	Dual	PSA	Steam cycle
BCL	Wet	-	Dual	PSA	Combined cycle



Fig. 2. Conventional low temperature wet cleaning (top) and advanced high temperature dry cleaning. The tar cracker is required after atmospheric gasification. COS hydrolysation becomes redundant when amine technology is applied for CO_2 removal downstream [12].

sorbents. Alkali removal via physical adsorption or chemisorption can be implemented at 750–900 °C, although lead and zinc can not be removed at this temperature. Sulphur is further removed by chemisorption. Thereafter, in absence of H₂S, 99.5% of the NH₃ can be decomposed over a nickel catalyst. Only HCN may be insufficiently removed by hot gas cleaning, leading to shorter catalyst life in downstream reactors.

Tijmensen [12] assumes maximum acceptable values of the contaminants for catalysts and equipment to lie in the 10–20 ppb range. The proposed cleaning technologies are appropriate and sufficient to meet most of these constraints.

2.1.3. Syngas processing

The syngas can contain a considerable amount of methane and other light hydrocarbons, representing a significant part or the heating value of the gas. Steam reforming (SMR) converts these compounds to CO and H₂ driven by steam addition over a nickel catalyst. Autothermal reforming (ATR) combines partial oxidation in the first part of the reactor with steam reforming in the second part, thereby optimally integrating the heat flows. It has been suggested that ATR, due to a simpler concept could become cheaper than SMR [1], although others give much higher prices [23]. There is dispute on whether the SMR can deal with the high CO and C+ content of the biomass syngas. Where Katofsky writes that no additional steam is needed to prevent coking or carbon deposition in SMR, Tijmensen [12] poses that this problem does occur in SMR and that ATR is the only technology able to prevent coking.

The syngas produced by the BCL and IGT gasifiers has a low H₂:CO ratio. The water gas shift (WGS) reaction is a common process operation to shift the energy value of the CO to H₂, which can than be separated using pressure swing adsorption. If the stoichiometric ratio of H₂, CO and CO₂ is unfavourable for methanol production, the water gas shift can be used in combination with a CO₂ removal step. The equilibrium constant for the WGS increases as temperature decreases. Hence, to increase the production to H₂ from CO, it is desirable to conduct the reaction at lower temperatures, which is also preferred in view of steam economy. However, to achieve the necessary reaction kinetics, higher temperatures are required [24,25].

2.2. MeOH production

2.2.1. Fixed bed technology

Methanol is produced by the hydrogenation of carbon oxides over a Cu/Zn/Al catalyst. The synthesis reactions are exothermic and give a net decrease in molar volume. Therefore, the equilibrium is favoured by high pressure and low temperature. During production, heat is released and has to be removed to maintain optimum catalyst life and reaction rate. The catalyst deactivates primarily because of loss of active copper due to physical blockage of the active sites by large by-product molecules, poisoning by halogens or sulphur in the synthesis gas, and sintering of the copper crystallites into larger crystals.

Conventional methanol reactors [26,27] use fixed beds of catalyst pellets and operate in the gas phase. Two reactor types predominate in plants built after 1970. The ICI lowpressure process is an adiabatic reactor with cold unreacted gas injected between the catalyst beds (Fig. 3, left). The subsequent heating and cooling leads to an inherent inefficiency, but the reactor is very reliable and therefore still predominant. The Lurgi system (Fig. 3, right), with the catalyst loaded into tubes and a cooling medium circulating on the outside of the tubes, allows near-isothermal operation. Conversion to methanol is limited by equilibrium considerations and the high temperature sensitivity of the catalyst. Temperature moderation is achieved by recycling large amounts of hydrogen rich gas, utilising the higher heat



Fig. 3. Methanol reactor types: quench (left) and steam raising (right).

capacity of H_2 gas and the higher gas velocities to enhance the heat transfer. Typically, a gas phase reactor is limited to about 16% CO gas in the inlet to the reactor, in order to limit the conversion per pass to avoid excess heating.

2.2.2. Slurry technology

Processes under development at present focus on shifting the equilibrium to the product side to achieve higher conversion per pass. Examples are the gas/solid/solid trickle flow reactor, with a fine adsorbent powder flowing down a catalyst bed and picking up the produced methanol; and liquid phase methanol processes where reactants, product, and catalyst are suspended in a liquid. In liquid phase processes [26,28] heat transfer between the solid catalyst and the liquid phase is highly efficient, thereby allowing high conversions per pass without loss of catalyst activity. Different reactor types are possible for liquid phase methanol production, such as a fluidised beds and monolithic reactors. The slurry bubble column reactor of the LPMEOH process (registered trademark of Air Products and Chemicals, Inc.; Fig. 4) was invented in the late 1970s and further developed and demonstrated in the 1980s. Reactants from the gas bubbles dissolve in the liquid and diffuse to the catalyst surface, where they react. Products then diffuse through the liquid back to the gas phase. Heat is removed by generating steam in an internal tubular heat exchanger.

Conversion per pass depends on reaction conditions, catalyst, solvent and space velocity. Experimental results show 15–40% conversion for CO rich gases and 40–70% CO for balanced and H₂ rich gases. Computation models predict future CO conversions of over 90%, up to 97%, respectively [26]. Researchers at the Brookhaven National Laboratory have developed a low temperature (active as low as 100 °C) catalyst that can convert 90% of the CO in one pass [1]. With steam addition, the reaction mixture becomes balanced through the water gas shift reaction, so that the initial hydrogen to carbon monoxide ratio is allowed to vary from 0.4 to 5.6 without a negative effect on performance [28].

Investment costs for the LP MeOH process are expected to be 5–23% less than for a gas phase process of the same

MeOH capacity. Methanol from a 420 MW electricity and 450–770 tonne per day methanol co-producing plant would cost under US\$ 0.50/gal. Methanol from an all methanol plant would cost about US\$ 0.60–0.70/gal. This compares with new methanol plants which produce methanol at US\$ 0.55–0.60/gal [28].

2.3. H_2 production

2.3.1. Pressure swing adsorption

After reforming and shifting to a hydrogen rich synthesis gas, hydrogen can be separated and compressed. Different process concepts are used in hydrogen plants in operation today. In the conventional design, CO₂ was removed and traces of CO and CO₂ were converted to easily removable methane to give hydrogen with 98% purity. This process is no longer dominating, but many plants using this concept are still operating. New hydrogen plants are almost invariably designed using pressure swing adsorption (PSA) for final hydrogen purification. The quality of the hydrogen produced is a major issue for its eventual automotive application. Specifically, CO is a strong poison to polymer electrolyte membrane (PEM) fuel cells. Studies indicate that levels as low as 1–2 ppb, will deactivate the platinum catalyst of such fuel cells [1]. PSA is based on the difference in adsorption behaviour between different molecules [1,29], it separates components of a gas stream by selective adsorption to a solid at high pressure, and subsequent desorption at low pressure. This adsorption/desorption is in fact a batch process, but by placing two beds in parallel it operates nearly continuous. While adsorption takes place in one bed, the other is desorbed [30].

PSA (see Fig. 5) was thoroughly described by Katofsky [1]. First, activated carbon in the set of beds 'A' selectively adsorbs nearly all CO_2 and all H_2O . The remaining gas then passes to the second set of beds 'B' containing a zeolite molecular sieve, which selectively adsorbs essentially all the remaining compounds and some hydrogen. The overall recovery of hydrogen is increased by recycling some of the desorbed gas from the 'B' beds. There is a





Fig. 4. LPMEOH reactor with detail of reaction [28].



Fig. 5. PSA for hydrogen purification [1].

trade-off in that the recycled gas must be recompressed and cooled to near ambient temperature, adding to capital and operating costs, and a slightly larger PSA unit will also be needed. As with the methanol synthesis loop, some of the recycled gas must be purged to prevent the build-up of methane and other non-hydrogen gases. Recovery rates of 90% and up are achievable, the product purity is extremely high: 99.999%.

2.3.2. Ceramic membranes

Membranes are a promising technology for gas separation. They are attractive because of their simple design and may have the ability of combining shift and separation in one reactor. Membranes for e.g. nitrogen separation are already applied at several small size facilities, where they have better economics than traditional separation technologies [1]. Membranes for hydrogen are evaluated as being an advanced option.

Membrane separation of gas mixtures is based on the difference in mobility of compounds through a surface. The driving force for transport of a component through the membrane is a difference in partial gas density, of this component on the two sides of the membrane. The membranes themselves affect the rates at which different gas molecules are transported through the membrane, depending on the physical and chemical interaction of the gases with the membrane.

Much R&D effort is put in decreasing the pore size to the size of molecules so that membranes can be molecular sieves, allowing only one component through. These membranes with small pore sizes are expected to perform better at high temperature [31], implying an important energy advantage when ceramic membranes are combined with hot gas cleaning, because than between gasification and gas turbine, no temperature drop would have to occur. Furthermore, the membrane surface catalyses the water gas shift reaction, this reaction is than driven to hydrogen as it is removed by the membrane permeable to H₂ but not to other gases. The shift reaction is demonstrated in the hydrogen separation device (HSD) made by Oak Ridge National Laboratory [32]. Since most of the information is confidential, it is not clear whether the catalytic activity stems from a catalyst condensed on the membrane surface or from the surface itself. If the former is the case, than sulphur removal upstream as not to poison this catalyst may be necessary [17]. The energy of the entering gas is shifted to hydrogen of which eventually 95% is separated at a purity of 99.5%.

Ceramic membranes have the advantage of a broad temperature and pressure operating range. Construction of membrane separation devices is potentially very simple and cheap when compared with other separation technologies such as pressure swing adsorption. Moreover, membranes do not suffer the efficiency losses and high capital costs for heat exchangers, associated with the need to cool the synthesis gas [17].

2.4. Electricity co-production

2.4.1. Gas turbines

Unconverted fuel gasses that remain after the methanol or hydrogen production section can still contain a significant amount of chemical energy. These gas streams may be combusted in a gas turbine, although they generally have a much lower heating value (4–10 MJ/N m³) than natural gas or distillate fuel (35–40 MJ/N m³) for which most gas turbine combustors have been designed. When considering commercially available gas turbines for low calorific gas firing, the following items deserve special attention [11,33, 34]: The combustion stability, the pressure loss through the fuel injection system, and the limits to the increasing mass flow through the turbine.

Different industrial and aeroderivative gas turbines have been operated successfully with low LHV gas, but on the condition that the hydrogen concentration in the gas is high enough to stabilise the flame. Up to 20% H₂ is required at 2.9 MJ/N m³. Hydrogen has a high flame propagation speed and thus decreases the risk of extinguishing the flame [33].

Injecting a larger fuel volume into the combustor through a nozzle originally designed for a fuel with much higher energy density can lead to pressure losses, and thus to a decreased overall cycle efficiency. Minor modifications are sufficient for most existing turbines. In the longer term, new turbines optimised for low heating value gas might include a complete nozzle combustor re-design [33].

The larger fuel flow rate also implies an increase in mass flow through the turbine expander, relative to natural gas firing. This can be accommodated partly by increasing the turbine inlet pressure, but this is limited by the compressor power available. At a certain moment the compressor cannot match this increased pressure any more and goes into stall: the compressor blocks. To prevent stall, decreasing the combustion temperature is necessary; this is called derating. This will lower the efficiency of the turbine, though [11,33]. Higher turbine capacity would normally give a higher efficiency, but as the derating penalty is also stronger the efficiency gain is small [34].

Due to the set-up of the engine the compressor delivers a specific amount of air. However, to burn 1 N m^3 of fuel gas less compressed air is needed compared to firing natural gas. The surplus air can be bled from the compressor at different pressures and used elsewhere in the plant, e.g. for oxygen production [11]. If not, efficiency losses occur.

All the possible problems mentioned for the currently available GTs, can be overcome when designing future GTs. Ongoing developments in gas turbine technology increase efficiency and lower the costs per installed kW over time [11]. Cooled interstages at the compressor will lower compressor work and produce heat, which can be used elsewhere in the system. Also, gas turbine and steam turbine could be put on one axis, which saves out one generator and gives a somewhat higher efficiency. And application of large scales will give increased turbine efficiency. The short-term restraints and long term possibilities of turbine efficiency are both dealt with.

Turbines set limits to the gas quality. The gas cleaning system needs to match particles and alkali requirements of the GTs. When these standards are exceeded wearing becomes more severe and lifetime and efficiency will drop [11]. However, the fuel gas that passed various catalysts prior to the gas turbine has to meet stricter demands. Contaminants are therefore not a real problem in the gas turbine.

2.4.2. Heat integration

As was pointed out in Fig. 1 heat is supplied or needed at several points in the biofuel production process. It is of great importance for the process efficiency that supply and demand are carefully matched, so that more high quality heat is left to raise and superheat high-pressure steam for electricity production in a steam turbine.

Usually, there is a supply of heat after the gasifier and reformer, where the gas streams are cooled prior to gas cleaning or compression. Furthermore, heat is recovered from flue gas from the gas turbine or boiler. There generally is a heat demand for the gas stream entering the reformer, and a steam demand for drying, for the gasifier, the reformer and the shift reactor.

3. Selected systems

Following the train of components of Fig. 1 and given the potential options for gasification, gas cleaning and conditioning, fuel synthesis and separation, many routes to produce methanol or hydrogen from biomass can be imagined. As was explained in Section 2, the IGT direct oxygen fired pressurised gasifier, in the normal and maximised H_2 option, and the Battelle indirect atmospheric gasifier are considered for synthesis gas production, because they deliver a medium calorific nitrogen undiluted gas stream and cover a broad range of gas compositions.

Some concepts chosen resemble conventional production of fuels from natural gas, making use of wet gas cleaning, steam reforming, shift, and either solid bed methanol reactor or hydrogen PSA. Similar concepts have previously been analysed by Katofsky [1]. Advanced components could offer direct or indirect energy benefits (liquid phase methanol synthesis, hot gas cleaning), or economic benefits (ceramic membranes, autothermal reforming). Available process units are logically combined so the supplied gas composition of a unit matches the demands of the subsequent unit, and heat leaps are restricted if possible. The following considerations play a role in selecting concepts:

• Hot gas cleaning is only sensible if followed by *hot* process units like reforming or (intermediate temperature) shifting. Hot gas cleaning is not applied after atmospheric

gasification since the subsequent pressurisation of the syngas necessitates cooling anyway.

- For reforming fuel gas produced via an IGT gasifier before methanol synthesis, an autothermal reformer is chosen, because of the higher efficiency, and lower costs. The high hydrogen yield, possible with steam reforming is less important here since the H₂:CO ratio of the gas is already high. The BCL gasifier, however, is followed by steam reforming to yield more hydrogen. For hydrogen production the IGT gas is not reformed, due to the low hydrocarbon content.
- Preceding liquid phase methanol synthesis, shifting the synthesis gas composition is not necessary since the reaction is flexible towards the gas composition. When steam is added, a shift reaction takes place in the reactor itself. Before gas phase methanol production the composition is partially shifted and because the reactor is sensible to CO₂ excess, part of the CO₂ is removed.
- For hydrogen production, the gas is fully shifted to maximise the H₂ yield. Ceramic membranes, however, do not need a preceding shift because the membrane surface is expected to have shifting capabilities.
- After the methanol once through options, the gas still contains a large part of the energy and is expected to suit gas turbine specifications. The same holds for unreformed BCL and IGT gases, which contain energy in the form of CO₂ + fractions. When the heating value of the gas stream does not allow stable combustion in a gas turbine, it is fired in a boiler to raise process steam. The chemical energy of IGT+ gas is entirely in hydrogen and carbon monoxide. After once through methanol production the gas still contains enough chemical energy for combustion in a gas turbine. After a ceramic membrane though, this energy has fully shifted to the produced and separated hydrogen; only expansion is applied to liberate the physical energy.
- Heat supply and demand within plants are to be matched to optimise the overall plant efficiency.

These considerations lead to a selection of 11 conversion concepts. The eleven concepts selected potentially have low cost and/or high energy efficiency. The concepts are composed making use of both existing commercially available technologies, as well as (promising) new technologies.

4. System calculations

4.1. Modelling

The selected systems are modelled in Aspen+, a widely used process simulation program. In this flowsheeting program, chemical reactors, pumps, turbines, heat exchanging apparatus, etc. are virtually connected by pipes. Every component can be specified in detail: reactions taking place, efficiencies, dimensions of heating surfaces and so on. For given inputs, product streams can be calculated, or one can evaluate the influence of apparatus adjustments on electrical output. The plant efficiency can be optimised by integrating the heat supply and demand. The resulting dimensions of streams and units and the energy balances can subsequently be used for economic analyses.

The pre-treatment and gasification sections are not modelled, their energy use and conversion efficiencies are included in the energy balances, though. The models start with the synthesis gas composition from the gasifiers as given in Table 1. Only the base scale of 80 dry tonne/h (430 MW_{th}) biomass is modelled. Modelling assumptions for the process units are given in Table 3.

Oxygen is used as oxidant for the IGT gasifier and the autothermal reformer. The use of air would enlarge downstream equipment size by a factor 4. Alternatively, oxygen enriched air could be used. This would probably give an optimum between small equipment and low air separation investment costs, but it is not considered in this study.

Gas turbines are modelled both as existing and advanced engines. The performance of the low calorific gas in existing gas turbines is calculated using GT Pro, a simulation program with an extensive database on available engines. Results from these calculations, on efficiency, flow dimensions and duct burning, were translated to Aspen+. On the longer term dedicated turbines for low calorific gas have higher efficiencies [11]. It is assumed that increase of scale can barely further improve these efficiencies.

The heat supply and demand within the plant is carefully matched, aimed at maximising the production of superheated steam for the steam turbine. A summation of all heat inputs and outputs in a heat bin is too simple, since it does not take the quality of heat into account. Pinch analysis, as was also done by Katofsky, gives the ultimate optimisation of energy streams within plants, but also leads to too optimistic ideal outcomes and possibly very large number of heat exchangers. Therefore heat integration of heat demand and supply within the considered plants here is done by hand. The intention is to keep the integration simple by placing few heat exchangers per gas/water/steam stream. Of course, concepts with more process units demanding more temperature altering are more complex than concepts consisting of few units. First, an inventory of heat supply and demand is made. Streams matching in temperature range and heat demand/supply are combined: e.g. heating before the reformer by using the cooling after the reformer. When the heat demand is met, steam can be raised for power generation. Depending on the amount and ratio of high and low heat, process steam is raised in heat exchangers, or drawn from the steam turbine: if there is enough energy in the plant to raise steam of 300 °C, but barely superheating capacity, than process steam of 300 °C is raised directly in the plant. If there is more superheating than steam raising capacity, than process steam is drawn from the steam cycle. Steam for gasification and drying is almost always drawn from the steam cycle, unless a perfect match is possible with

a heat-supplying stream. The steam entering the steam turbine is set at 86 bar and 510 $^\circ\mathrm{C}.$

4.2. System calculation results

Table 4 summarises the outcomes of the flowsheet models. The overall energy efficiencies are expressed in different ways. The most direct is the net overall fuel + electricity efficiency, but this definition gives a distorted view, since the quality of energy in fuel and electricity is considered equal, while in reality it is not. The fuel only efficiency assumes that the electricity part could be produced from biomass at 45% HHV in an advanced BIG/CC [35], this definition compensates for the inequality of electricity and fuel in the most justified way, but the referenced electric efficiency is of decisive importance. Expressing the performance in primary energy avoided divides the co-generation benefit over fuel and electricity. Another qualification for the performance of the system could use exergy: the amount of work that could be delivered by the material streams.

In some concepts still significant variations can be made. In methanol concept 4, the reformer needs gas for firing. The reformer can either be entirely fired by purge gas (thus restricting the recycle volume), or by part of the gasifier gas. The first option gives a somewhat higher methanol production and overall plant efficiency. The hydrogen concept 4 offers a similar choice between reformer combustor feeding directly from the gasifier, or from the purge gas. But in this concept combusting part of the gasifier gas gives the higher efficiency. In methanol concept 5 one can choose between a larger recycle, and more steam production in the boiler. A recycle of five times the feed volume, instead of four, gives a much higher methanol production and plant efficiency. Per concept only the most efficient variation is reported in Table 4.

Based on experiences with low calorific combustion elsewhere [11,33] the streams in this study, which were projected to be combusted in a gas turbine, will give stable combustion. Only the performance of the gas turbine in the hydrogen 3 concept is unsure, having a low calorific value combined with little hydrogen. In GTpro gas turbines are chosen with dimensions matching the heat flow of the purged gas, and with high combined cycle efficiencies. Gas turbine only efficiencies are 33–47%; the high value is found for pressurised hot gas after the ceramic membrane. Advanced turbine configurations, with set high compressor and turbine efficiencies of 41-52% and 1-2% point higher overall plant efficiency than conventional configurations. Table 4 only includes the advanced turbines.

Based on the overall plant efficiency the methanol concepts lie in a close range: methanol 50–57% and hydrogen 52–61%. Liquid phase methanol production preceded by reforming (concepts 2 and 4) results in somewhat higher overall efficiencies and primary energy avoided. After the pressurised IGT gasifier hot gas cleaning leads to higher efficiencies than wet gas cleaning, although not better than

Table 3
Unit modelling assumptions used in Aspen+ calculations

General	
Heat exchanger ^{a,b,c}	$\Delta p/p = 2\%$ Minimum $\Delta T = 15$ °C (gas–liquid) or 30 °C (gas–gas) If $T > 300$ °C, then heat losses are 2% of heat transferred
Centrifugal pump ^d	$\eta = 0.65 - 0.9$ $\eta_{ m driver} = 1$
Blower ^d	$\Delta P < 0.5 \text{ bar}$ $\eta_{\text{isentropic}} = 0.72$ $\eta_{\text{mech}} = 1$
Compressor ^b	$\eta_{ m polytropic} = 0.80$ $\eta_{ m mech} = 0.90$
Multistage compressor ^e	$\eta_{\text{Isentropic}} = 0.76-0.78$ (for 1.0e4–1.7e5 m3/h input volume) $\eta_{\text{mech}} = 1$ Compression ratio is same for each stage, maximum is 4, such that outlet temperature does not exceed 250 °C Intercooling to 25–130 °C, last stage no duty
Gas cleaning ^f Quench scrubber ^{b.g}	Modelled as two outlet flash drum $T_{in, gas} = 250-240-120$ °C (for 34.5-24-1.2 bar) $T_{in, water} = 25$ °C T = 200-180-60 °C preferably (for 34.5-24-1.2 bar) by adjusting amount of water; design spec TQUENCH; Minimally 1 m ³ water per 1000 m ³ gas Q = 0 W $\Delta p/p = 3\%$
Hot gas cleaning ^{c,h}	Modelled as value $T_{\rm in} = 350 ^{\circ}\text{C}/550 ^{\circ}\text{C}/800 ^{\circ}\text{C}$ $\Delta p = -5 \text{bar}$
Reformer	
Steam reformer ⁱ	SMR1 provides heat to SMR2 by combusting flue gas. If this is not sufficient then part of gasifier product is combusted as well SMR1: stoichiometric reactor $T = 890 ^{\circ}\text{C}$ $\Delta p/p = 2\%$ Air is stoichiometric SMR2: Gibbs free energy minimisation reactor $T_{\text{in}} = 860 ^{\circ}\text{C}$; $p_{\text{in}} = 15.5$ bar $\Delta p = -0.5$ bar $T = 890 ^{\circ}\text{C}$; $T_{\text{approach}} = -10 ^{\circ}\text{C}$ 3.5 mol steam injected per mole carbon
Autothermal reformer ⁱ	ATR1 provides heat ATR2 requires $T_{in} = 550 \text{ °C}$ Adjust ratio ATR1/ATR2 to $T_{out} = 1000 \text{ °C}$ Overall 2 mol steam injected per mole carbon, some gas streams do not require additional steam for reforming ATR1: stoichiometric reactor T = 1000 °C $\Delta p = -0.5$ bar Complete combustion of CH ₄ , C ₂ H ₄ and C ₂ H ₆ using stoichiometric amount of air ATR2: Gibbs free energy minimisation reactor 2% of CH ₄ is inert Ar and N ₂ are inert; C ₂ H ₄ and C ₂ H ₆ react completely
Shift	
Partial shift reactor ^a	Part of stream splits to SHIFT reactor such that ratio $(H_2 - CO_2)/(CO + CO_2) = 2.05 \pm 0.02$ after downstream Selexol Modelled as Gibbs free energy minimisation reactor $T_{in} = 330 ^{\circ}\text{C}$ $T_{approach} = +10 ^{\circ}\text{C}$ Q = 0 W $\Delta p = -0.5 \text{ bar}$ Inertia: CH ₄ , C ₂ H ₄ , C ₂ H ₆ , Ar, N ₂ Steam injected is three times CO - H ₂ O
Dual shift reactor ^a	HT shift: Gibbs free energy minimisation reactor $T_{in} = 350 ^{\circ}\text{C}$ maximally

	$T_{approach} = +10 \text{ °C}$ $Q = 0 \text{ W}$ $\Delta p = -0.5 \text{ bar}$ Inertia: CH ₄ , C ₂ H ₄ , C ₂ H ₆ , Ar, N ₂ Steam injected is three times CO - H ₂ O LT shift: Gibbs free energy minimisation reactor $T_{in} = 260 \text{ °C}$ $\Delta T = +20 \text{ °C}$ $\Delta p = -0.5 \text{ bar}$ Inertia: CH ₄ , C ₂ H ₄ , C ₂ H ₆ , Ar, N ₂
Chemical reactors Conventional solid bed methanol ^{a,k}	Modelled as Gibbs free energy minimisation reactor $p_{in} = 106$ bar; $\Delta p = -8$ bar Q = 0 W $T_{in} = 50$ and 250 °C Inertia: CH ₄ , C ₂ H ₄ , C ₂ H ₆ MeOH in reactor product = 6 mol% by adjusting $T_{approach}$ $T = 260$ °C ± 2.6 by adjusting cold/hot feed ratio Recycle to feed ratio = 5
Liquid phase methanol ¹	Modelled as Gibbs free energy minimisation reactor $p_{in} = 90$ bar, $T_{in} = 240 \pm 2.4$ °C by adjusting <i>T</i> before compression, design spec TFEED $\Delta p = -2$ bar, $T = 250$ °C Inertia: CH ₄ , C ₂ H ₄ , C ₂ H ₆ Balanced/H ₂ -rich syngas (H ₂ :CO > 2) 75% conversion in CO CO-rich syngas (2 > H ₂ :CO > 1) 60% conversion in CO by adjusting $T_{approach}$ Optional steam addition H ₂ /CO ratio at reactor outlet is adjusted to 2.05 ± 0.02; design spec STMEOH Syngas becomes balanced; real CO level = (CO _{in} + H _{2 in})/3 Optional recycle with recycle to feed ratio = 2 or lower
Purification Methanol separator ^a	Modelled as two outlet flash drum 45 °C > T_{in} > 30 °C Q = 0 W -50 bar < Δp < -5 bar Subsequent separator for 100% pure MeOH
Selexol ^{a,m}	98% of CO ₂ and 100% of H ₂ O separation $T_{in} = 127 \degree C$ $\Delta p = -0.5 \text{ bar}$ CO ₂ released at 1.5 bar
Water separator ⁿ	Modelled as two outlet flash drum $T_{in} = 40 \ ^{\circ}\text{C}$ $Q = 0 \ \text{W}$ Δp as HX or more when desired before PSA
PSA system ^a	System operating at 14–28 bar, 40 °C Recycling 80% PSA-A $\Delta p = -0.35$ bar 100% CO ₂ + H ₂ O adsorption Desorption at 1.3 bar PSA-B $\Delta p = -0.35$ bar Adsorption of all gas but 84% of H ₂ Desorption at 1.3 bar
Ceramic membrane ^{a.o}	System operating at elevated pressure: 20 bar or higher $T_{in} = T_{out} = 800 \text{ °C}$ Catalytic molecular sieve: shift all CO on surface to H ₂ , therefore, H ₂ O:CO \Rightarrow 1 at entrance, transport 95% of H ₂ and 0% of others to product stream Product at 1.2 bar Δp depleted stream = -0.1 bar
Power generation Advanced gas turbine ^p	Compressor $p_{\text{fuel}} = 50 \text{ bar}, p_{\text{air}} = 40 \text{ bar}$

Table 3 (Continued)

Existing gas turbine	$\eta_{\text{isentropic}} = 0.91$ $\eta_{\text{mech}} = 0.99$ Combustor modelled as stoichiometric reactor $\Delta p = 0$ bar Q = 0 W T after turbine expander = 550 ± 2 °C by adjusting air Temperature is set by adjusting air to compressor Expander: $p = 1.2$ bar $\eta_{\text{isentropic}} = 0.89$ $\eta_{\text{mech}} = 0.99$ T after heat exchanger = 100 °C Data on pressures, efficiencies, turbine cooling, etc. from Gtpro
Boiler	2–3 MJ/m ³ is lower limit for normal combustion Stoichiometric reactor $p_{in} = 1.2$ bar $\Delta p = -0.1$ bar T = 1200 °C by superheating steam $O_2 = 5\% \pm 0.05$ by adjusting air
HRSG ^q	Gas $T_{out} = 100 \ ^{\circ}\text{C}$ Water $T_{in} = 15 \ ^{\circ}\text{C}$
Steam turbine ^f	Steam of preferably 86.2 bar, 510 °C is expanded Intermediate steam extraction is possible: $p = p_{\text{gasifier}} (34.5 \text{ bar/}250 °C, 25 \text{ bar/}240 °C, 1.2 \text{ bar/}120 °C)$ p = 12 bar for drier (200°C) p = 0.04 bar $\eta_{\text{isentropic}} = 0.89$ $\eta_{\text{mech}} = 0.99$
Air composition	$O_2 = 0.2075$ $H_2O = 0.0101$ $CO_2 = 0.0003$ $N_2 = 0.7729$ Ar = 0.0092 T = 15 °C, p = 1 atm

^a [1].

^b [33].

° [12].

^d Aspen+ default value.

^e [54].

^f The tar cracker following the atmospheric gasification (BCL) is not modelled. It is assumed to be integrated with the gasifier.

^g [8].

^h Hot gas cleaning modelled as a black box. Operating temperature as defined in Table 2.

ⁱ Steam reformer operates at 1–3.5 MPa, with molar steam:carbon ratios in the range 3–5:1. Typical reformer temperature is between 830 and 1000 °C [1]. The inlet stream is heated by the outlet stream up to 860 °C to match reformer heat demand and supply. The furnace would typically use 10% excess air for C_1 to C_4 firing, correlating to approximately 1.7% oxygen in the flue gas, to ensure that the burners do not limit plant throughput, and for safety reasons [55]. The modelled SMR furnace is sized as to exactly match the heat demand, without an excess air.

^j Autothermal reformer operates at 20–70 bar, 850–1100 °C, steam to carbon ratio ranges from 0.5 to 3.5 [56]. Oxygen is set stoichiometric for oxidation part of ATR.

^k Conventional gas phase methanol reactor modelled as quench type (ICI low pressure methanol process). Typical methanol synthesis temperature is between 230 and 270 °C. Temperature is set 260 °C by adjusting the cold/hot feed ratio. The reactor operates adiabatic. Pressure is typically 50–150 bar, pressure drop is 5–8 bar. Recycle to feed ratio is typically between 2.3 and 6 [1,13,26,27]. Side reactions to dimethyl-ether, formaldehyde or higher alcohols are not modelled.

¹Liquid phase methanol reactor. Experimental results show 15–40% conversion for CO rich gases and 40–70% CO for balanced and H_2 rich gases, but computation models predict future CO conversions of over 90%, up to 97%, respectively [26,28,36]. Side reactions to dimethyl-ether, formaldehyde or higher alcohols are not modelled.

^m Selexol. Actually, half of the CO_2 is released at 1 bar and half at 4 bar. The net energy demand of a 436 tonne of CO_2/h unit amounts 9 MW_e [57]. ⁿ Over 99% of the water is separated, over 99.99% of the combustible gasses passes through.

^o Ceramic membranes modelled as hydrogen separation device or HSD (developed by Oak Ridge National Laboratory). Operation at high temperature, surface has shifting capabilities [32,58–60].

^p [11].

^q HRSG after GT or boiler. The flue gas can be cooled down to 100 °C without corrosion problems, since the gas is expected to contain less than 100 ppb sulphur [11].

Table 4

Results of the Aspen+ performance calculations, for 430 MW_{th} input HHV systems (equivalent to 380 MW_{th} LHV for biomass with 30% moisture) of the methanol and hydrogen production concepts considered

	HHV output (MW)		HHV efficie	Primary energy	
	Fuel	Net electricity ^a (gross – internal)	Fuel + E	Fuel only ^b	avoided ^e (%)
Methanol					
IGT-max H ₂ , scrubber, liquid phase methanol reactor, combined cycle	161	53 (71–18)	50	52	83
IGT, hot gas cleaning, autothermal reformer, liquid phase methanol reactor with steam addition, combined cycle	173	62 (82–20)	55	59	91
IGT, scrubber, liquid phase methanol reactor with steam addition, combined cycle	113	105 (118–14)	51	58	87
BCL, scrubber, steam reformer, liquid phase methanol reactor with steam addition and recycle, steam cycle	246	0 (25–25)	57	57	90
IGT, hot gas cleaning, autothermal reformer, partial shift, conventional methanol reactor with recycle, steam turbine	221	15 (38–23)	55	56	88
BCL, scrubber, steam reforming, partial shift, conventional methanol reactor with recycle, steam turbine	255	-17 (10-27)	55	54	86
Hydrogen					
IGT, hot gas cleaning, dual shift, pressure swing adsorption, combined cycle	176	73 (93–21)	58	66	85
IGT-max H ₂ , high temperature dust filter, ceramic membrane (internal shift), expansion turbine	259	-1 (25-26)	60	60	79
IGT, hot gas cleaning, ceramic membrane (internal shift), combined cycle	177	84 (103–19)	61	74	91
BCL, scrubber, steam reformer, dual shift, pressure swing adsorption	303	-22 (0-22)	65	63	83
BCL, scrubber, dual shift, pressure swing adsorption, combined cycle	149	72 (97–25)	52	56	77

^a Net electrical output is gross output minus internal use. Gross electricity is produced by gas turbine and/or steam turbine. The internal electricity use stems from pumps, compressors, oxygen separator, etc.

^b The electricity part is assumed to be produced from biomass at $\eta_e = 45\%$ HHV efficiency [35]. The fuel only efficiency is calculated by $\eta = \text{fuel}/(\text{MW}_{\text{th,in}} - \text{electricity}/\eta_e)$.

^c The mix fuel + electricity could also be produced from natural gas at $\eta_e = 54\%$ and $\eta_f = 63\%$ for methanol or $\eta_f = 76\%$ for hydrogen [46]. Primary energy avoided is calculated by PA = (electricity/ $\eta_e + \text{fuel}/\eta_f)/(\text{MW}_{\text{th,in}})$.

concepts with wet gas cleaning after a BCL gasifier. The conventional hydrogen concept 4 has the highest overall plant efficiency, but depends heavily on the import of electricity to the plant. If compared on a fuel only basis, its performance is the worst of the hydrogen concepts. The most advanced concept hydrogen 3 than is the most efficient.

Several units may be realised with higher efficiencies than considered here. For example, new catalysts and carrier liquids could improve liquid phase methanol single pass efficiency up to 95% [36]. The electrical efficiency of gas turbines will increase by 2–3% points when going to larger scale [37].

5. Economics

5.1. Method

An economic evaluation has been carried out for the concepts considered. Plant sizes of 80, 400, 1000 and 2000 MW_{th} HHV are evaluated, 400 MW_{th} being the base scale. The scale of the conversion system is expected to be an important factor in the overall economic performance. This issue has been studied for BIG/CC systems [35,38], showing that the economies of scale of such units can offset the

increased costs of biomass transport up to capacities of several hundreds of MW_{th}. The same reasoning holds for the fuel production concepts described here. It should however be realised that production facilities of 1000-2000 MW_{th} require very large volumes of feedstock: 200-400 dry tonne/h, or 1.6-3.2 dry Mtonne per year. Biomass availability will be a limitation for most locations for such large-scale production facilities, especially in the shorter term. In the longer term (2010–2030), if biomass production systems become more commonplace, this can change. Very large scale biomass conversion is not without precedent: various large-scale sugar/ethanol plants in Brazil have a biomass throughput of 1-3 Mtonne of sugarcane per year, while the production season covers less than half a year. Also large paper and pulp complexes have comparable capacities. The base scale chosen is comparable to the size order studied by Williams et al. [4] and Katofsky [1], 370–385 MW_{th}.

The methanol and hydrogen production costs are calculated by dividing the total annual costs of a system by the produced amount of fuel. The total annual costs consist of:

- annual investments;
- operating and maintenance;

- biomass feedstock;
- electricity supply/demand (fixed power price).

The total annual investment is calculated by a factored estimation [39], based on knowledge of major items of equipment as found in literature or given by experts. The uncertainty range of such estimates is up to $\pm 30\%$. The installed investment costs for the separate units are added up. The unit investments depend on the size of the components (which follow from the Aspen+ modelling), by scaling from known scales in literature (see Table 5), using Eq. (1):

$$\frac{\cos t_a}{\cos t_b} = \left(\frac{\operatorname{size}_a}{\operatorname{size}_b}\right)^R \tag{1}$$

with R, the scaling factor.

Various system components have a maximum size, above which multiple units will be placed in parallel. Hence the influence of economies of scale on the total system costs decreases. This aspect is dealt with by assuming that the base investment costs of multiple units are proportional to the cost of the maximum size: the base investment cost per size becomes constant. The maximum size of the IGT gasifier is subject to discussion, as the pressurised gasifier would logically have a larger potential throughput than the atmospheric BCL.

The total investment costs include auxiliary equipment and installation labour, engineering and contingencies. If only equipment costs, excluding installation, are available, those costs are increased by applying an overall installation factor of 1.86. This value is based on 33% added investment to hardware costs (instrumentation and control 5%, buildings 1.5% grid connections 5%, site preparation 0.5%, civil works 10%, electronics 7%, and piping 4%) and 40% added installation costs to investment (engineering 5%, building interest 10%, project contingency 10%, fees/overheads/profits 10%, start-up costs 5%) [35].

The annual investment cost follows from Eq. (2), which takes the technical and economic lifetime of the installation into account. The interest rate is 10%.

$$I_{\text{annual}} = \frac{\text{IR}}{1 - 1/(1 + \text{IR})^{t_{\text{e}}}} I_t \left(1 - \frac{1}{(1 + \text{IR})^{t_{\text{e}}}} \frac{t_t - t_{\text{e}}}{t_t} \right)$$
(2)

Table 5

Costs of system components used for cost analyses of considered concepts, in MUS\$2001

	Base investment cost (fob)	Scale factor	Base scale	Overall installation factor ^b	Maximum size ^c
Pre-treatment ^d					
Conveyers ^e	0.35	0.8	33.5 wet tonne/h	1.86 (v)	110
Grinding ^e	0.41	0.6	33.5 wet tonne/h	1.86 (v)	110
Storage	1.0	0.65	33.5 wet tonne/h	1.86 (v)	110
Dryer ^e	7.6	0.8	33.5 wet tonne/h	1.86 (v)	110
Iron removal ^e	0.37	0.7	33.5 wet tonne/h	1.86 (v)	110
Feeding system ^{e,f}	0.41	1	33.5 wet tonne/h	1.86 (v)	110
Gasification system					
BCL ^g	16.3	0.65	68.8 dry tonne/h	1.69	83
IGT ^h	38.1	0.7	68.8 dry tonne/h	1.69	75
Oxygen plant (installed) ⁱ	44.2	0.85	41.7 tonne O ₂ /h	1	-
Gas cleaning					
Tar cracker ^e	3.1	0.7	$34.2 \text{ m}^3 \text{ gas/s}$	1.86 (v)	52
Cyclones ^e	2.6	0.7	$34.2 \text{ m}^3 \text{ gas/s}$	1.86 (v)	180
High-temperature heat exchanger ^j	6.99	0.6	39.2 kg steam/s	1.84 (v)	-
Baghouse filter ^e	1.6	0.65	$12.1 \text{ m}^3 \text{ gas/s}$	1.86 (v)	64
Condensing scrubber ^e	2.6	0.7	12.1 m ³ gas/s	1.86 (v)	64
Hot gas cleaning ^k	30	1.0	74.1 m ³ gas/s	1.72 (v)	-
Syngas processing					
Compressor	11.1	0.85	13.2 MW _e	1.72 (v)	-
Steam reformer ^m	9.4	0.6	1390 kmol total/h	2.3 (v)	-
Autothermal reformer ⁿ	4.7	0.6	1390 kmol total/h	2.3 (v)	-
Shift reactor (installed) ^o	36.9	0.85	$15.6 \text{ Mmol CO} + H_2/h$	1	-
Selexol CO ₂ removal (installed) ^p	54.1	0.7	9909 kmol CO ₂ /h	1	-
Methanol production					
Gas phase methanol ^q	7	0.6	87.5 tonne MeOH/h	2.1 (v)	-
Liquid phase methanol ^r	3.5	0.72	87.5 tonne MeOH/h	2.1 (v)	-
Refining ^s	15.1	0.7	87.5 tonne MeOH/h	2.1 (v)	
Hydrogen production					
PSA units $A + B^t$	28.0	0.7	9600 kmol feed/h	1.69	-
Ceramic membrane (installed) ^u	21.6	0.8	17 tonne H ₂ /h	1	-
Power isle ^v					
Gas turbine + HRSG ^{e,w}	18.9	0.7	26.3 MW _e	1.86 (v)	-

Table 5 (Continued)

	Base investment cost (fob)	Scale factor	Base scale	Overall installation factor ^b	Maximum size ^c
Steam turbine + steam system ^{e,x}	5.1	0.7	10.3 MW _e	1.86 (v)	_
Expansion turbine ^y	4.3	0.7	10.3 MW _e	1.86 (v)	

^a Annual GDP deflation up to 1994 is determined from OECD [61] numbers. Average annual GDP deflation after 1994 is assumed to be 2.5% for the US, 3.0% for the EU. Cost numbers of Dutch origin are assumed to be dependent on the EU market, therefore EU GDP deflators are used. $1 \in_{2001} = US_{2001}^{*} 0.94 = 2.204 \text{ Dfl}_{2001}$.

^b Overall installation factor. Includes auxiliary equipment and installation labour, engineering and contingencies. Unless other values are given by literature, the overall installation factor is set 1.86 for a 70 MW_{th} scale [35]. This value is based on 33% added investment to hardware costs (instrumentation and control 5%, buildings 1.5% grid connections 5%, site preparation 0.5%, civil works 10%, electronics 7%, and piping 4%) and 40% added installation costs to investment (engineering 5%, building interest 10%, project contingency 10%, fees/overheads/profits 10%, start-up costs 5%). For larger scales, the added investments to hardware decreases slightly.

^c Maximum sizes from Tijmensen [12].

^d Total pre-treatment approximately sums up to a base cost of MUS\$₂₀₀₁ 8.15 at a base scale of 33.5 tonne wet/h with an R factor of 0.79.

^e Based on first generation BIG/CC installations. Faaij et al. [62] evaluated a 29 MW_e BIG/CC installation (input 9.30 kg dry wood/s, produces 10.55 N m³ fuel gas/s) using vendor quotes. When a range is given, the higher values are used [35]. The scale factors stem from Faaij et al. [35].

^f Two double screw feeders with rotary valves [62].

^g MUS $_{1991}$ 12.72 (already includes added investment to hardware) for a 1650 dry tonne per day input BCL gasifier, feeding not included, *R* is 0.7 [4]. Stronger effects of scale for atmospheric gasifiers (0.6) were suggested by Faaij et al. [35]. Technical director Mr. Paisley of Battelle Columbus, quoted by Tijmensen [12] estimates the maximum capacity of a single BCL gasifier train at 2000 dry tonne per day.

^h MUS $_{1991}$ 29.74 (includes already added investment to hardware) for a 1650 dry tonne per day input IGT gasifier, R = 0.7 [4]. Maximum input is 400 MW_{th} HHV [12].

ⁱ Air separation unit: plant investment costs are given by Van Dijk [13]: $I = 0.1069 \text{ C}^{0.8508}$ in MUS\$₁₉₉₅ installed, C = capacity in tonne O₂ per day. The relation is valid for 100–2000 tonne O₂ per day. Williams et al. [4] assume higher costs for small installations, but with a stronger effect of scale: $I = 0.260 \text{ C}^{0.712}$ in MUS\$₁₉₉₁ fob plus an overall installation factor of 1.75 (25 and 40%). Larson et al. [40] assume lower costs than Van Dijk, but with an even stronger scaling factor than Williams: MUS\$₁₉₉₇ 27 installed for an 1100 tonne O₂ per day plant and R = 0.6. The first formula (by Van Dijk) is used in the present study. The production of 99.5% pure O₂ using an air separation unit requires 250–350 kWh per tonne O₂ [13,63].

^j High temperature heat exchangers following the gasifier and (in some concepts) at other locations are modelled as HRSGs, raising steam of 90 bar/ 520 °C. A 39.2 kg steam/s unit costs 6.33 MUS\$₁₉₉₇ fob, overall installation factor is 1.84 [40].

^k Tijmensen [12] assumes the fob price for Hot Gas Cleaning equipment to be MUS_{2000} 30 for a 400 MW_{th} HHV input. This equals 74.1 m³/s from a BCL gasifier (T = 863 °C, 1.2 bar). There is no effect of scaling.

¹ Katofsky [1] assumes compressors to cost US $_{1993}$ 700 per required kW_{mech}, with an installation factor of 2.1. The relation used in the present study stems from the compressor manufacturer Sulzer quoted by [12]. At the indicated base-scale, total installed costs are about 15% higher than assumed by Katofsky. Multiple compressors, for fuel gas, recycle streams, or hydrogen, are considered as separate units. Overall installation factor is taken 1.72 because the base unit matches a 400 MW_{th} plant rather than a 70 MW_{th} plant.

^m Investments for steam reformer vary from MUS\$₁₉₉₃ 16.9, for a throughput of 5800 kmol methane/h with an overall installation factor of 2.1 [1] to k\$₁₉₉₅ 7867 for a 6.2 kg methane/s (1390 kmol/h), overall installation factor is 2.3 [13]. These values suggest a strong effect of scaling R = 0.51, while Katofsky uses a modest R = 0.7. In the present study the values of Van Dijk are used in combination with an R factor of 0.6. The total amount of moles determines the volume and thus the price of the reactor.

ⁿ Autothermal reforming could be 50% cheaper than steam reforming [1], although higher costs are found as well [23].

^o Investment for shift reactors vary from MUS\$₁₉₉₅ 9.02 for an 8819 kmol CO + H₂/h reactor, and an overall installation factor is 1.81 [4] to MUS\$₁₉₉₄ 30 installed for a 350,000 N m³/h CO + H₂/h (15,625 kmol/h) reactor [57]. Williams assumes an R = 0.65, but comparison of the values suggest only a weak influence of scale (R = 0.94), in the present study the values from Hendriks are used, R is set 0.85. A dual shift is costed as a shift of twice the capacity.

^p Costs for CO₂ removal through Selexol amountsMUS\$₁₉₉₃ 14.3 fob (overall installation factor is 1.87) for an 810 kmol CO₂/h unit, R = 0.7 [1] up to MUS\$₁₉₉₄ 44 installed for a 9909 kmol CO₂/h unit [57]. The value from Hendriks is assumed to be right, since his research into CO₂ removal is comprehensive. ^q Van Dijk et al. [13] estimate that a methanol reactor for a 2.1 ktonne methanol per day plant costs kUS\$₁₉₉₅ 4433 (fob) or kUS\$₁₉₉₅ 9526 installed

(overall installation factor is 2.1). The total plant investment in their study is MUS_{1995} 138, or MUS_{2001} 150. Katofsky [1] estimates the costs for a 1056 tonne methanol per day plant to be MUS_{1995} 50 fob, this excludes the generation and altering of syngas, but includes make-up and recycle compression and refining tower. Correspondence with Van Ooijen [64] of Akzo Nobel and De Lathouder [65] of DSM Stamicarbon revealed that a 1000 tonne per day plant costs about 160 MUS $_{2001}$, and a 2000 tonne per day plant MUS $_{2001}$ 200 (this suggests a total plant scale factor of 0.3). These values come near the ones mentioned by Katofsky. This implies that the values given by Van Dijk are too optimistic and should be altered by a factor 1.33. It is therefore assumed that the base investment for the methanol reactor only is MUS_{2001} 7, the installation factor is 2.1. The influence of scale on reactor price is assumed to be not as strong as for the complete plant: 0.6.

^r Installed costs for a 456 tonne per day liquid phase methanol unit, areMUS\$₁₉₉₇ 29, excluding generation and altering of syngas, but including make-up and recycle compression, and refining tower. R = 0.72 [66]. Corrected for scale and inflation this value is about half the cost of the conventional unit by Katofsky and the corrected costs of Van Dijk. It is therefore assumed that the price of a liquid phase methanol reactor is MUS\$₂₀₀₁ 3.5 for a 2.1 ktonne per day plant, installation factor is 2.1.

Cost number for methanol separation and refining is taken from Van Dijk, increased with 33% as described in note 15.

^t PSA units (excluding the recycle compressor) cost MUS $_{1993}$ 23 for a 9600 kmol feed/h throughput, R = 0.7 [1].

^u Membrane costs US $_{1997}$ 68/(kW/bar), but these costs are only 9% of the total installed cost for a hydrogen separation device. Investment costs stem from Parsons I&TG [32]. The economies of scale of the membrane surface are low because the required surface area is proportional to the throughput, this slightly influences the overall *R* factor of the complete HSD.

^v For indication: a complete combined cycle amounts about US\$₁₉₉₇ 830 per installed kWe. Quoted from [67] by [23].

^w Scaled on gas turbine size.

^x Steam system consists of water and steam system, steam turbine, condenser and cooling. Scaled on steam turbine size.

^y Expansion turbine costs are assumed to be the same as steam turbine costs (without steam system).

where I_{annual} is the annual investment costs; IR the interest rate, 10%; I_t the total investment (sum of unit investments); t_e the economical lifetime, 15 years; t_t the technical lifetime, 25 years.

Operational costs (maintenance, labour, consumables, residual streams disposal) are taken as a single overall percentage (4%) of the total installed investment [35,40]. Differences between conversion concepts are not anticipated.

It is assumed in this study that enough biomass will be available at US\$ 2/GJ (HHV), this is a reasonable price for Latin and North American conditions. Costs of cultivated energy crops in the Netherlands amount approximately US\$ 4/GJ and thinnings US\$ 3/GJ [41], and biomass imported from Sweden on a large scale is expected to cost US\$ 7/GJ [42]. On the other hand, biomass grown on Brazilian plantations could be delivered to local conversion facilities at US\$ 1.6–1.7/GJ [4,43].

Electricity supplied to or demanded from the grid costs US\$ 0.03/kWh. The annual load is 8000 h.

5.2. Results

Results of the economic analysis are given in Table 6 and Fig. 6. The 400 MW_{th} conversion facilities deliver methanol at US\$ 8.6–12/GJ, the hydrogen cost range is US\$ 7.7–11/GJ. Considering the 30% uncertainty range one should be careful in ranking the concepts. Some concepts (methanol 4 and 6 and hydrogen 2, 3 and 4) perform somewhat better than the other concepts due to an advantageous combination of lower investment costs and higher efficiency. The lowest

methanol production price is found for concepts using the BCL gasifier, having lower investment costs. In hydrogen production the ceramic membrane concepts perform well due to their higher overall efficiency and modest investments. The combination of an expensive oxygen fired IGT gasifier with an combined cycle is generally unfavourable, since the efficiency gain is small compared to the high investment. Only in hydrogen 3 (using a ceramic membrane) the extra investment seems justified.

Investment redemption accounts for 42-76% of the annual costs and is influenced by the unit investment costs, the interest rate and the plant scale. The build-up of the total investment for all concepts is depicted in Fig. 7. It can be seen that the costs for the gasification system (including oxygen production), syngas processing and power generation generally make up the larger part of the investment. For autothermal reforming higher investment costs [23], would increase the methanol price from considered concepts by about US\$ 1.5/GJ. Uncertainties in the investment costs for ceramic membranes, however, do not have a large influence. Developments in gasification and reforming technology are important to decrease the investments. On the longer term, capital costs may reduce due to technological learning: a combination of lower specific component costs and overall learning. A third plant build may be 15% cheaper leading to a 8-15% fuel cost reduction.

The interest rate has a large influence on the fuel production costs. At a rate of 5% methanol production costs decrease with about 20% to US\$ 7.2–9.0/GJ, hydrogen to US\$ 6.6–8.5/GJ. At a high interest rate (15%) methanol production costs become US\$ 9.9–14/GJ, hydrogen US\$



Fig. 6. Fuel price for 400 MW_{th} input concepts, with 30% uncertainty on investment costs.

Table 6Economic analyses for the concepts considered

	Methanol					Hydrogen					
	1	2	3	4	5	6	1	2	3	4	5
Gasification system											
Total pre-treatment (MUS\$)	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2	38.2
BCL	0	0	0	30.4	0	30.4	0	0	0	30.4	30.4
IGT	73.0	73.0	73.0	0	73.0	0	73.0	73.0	73.0	0	0
Oxvgen	33.8	43.8	27.7	0	43.7	0	27.7	33.8	27.7	0	0
Gas cleaning											
Tar cracker	0	0	0	9.2	0	9.2	0	0	0	9.2	9.2
Cyclones	1.8	1.2	1.2	6.8	1.2	6.8	1.2	1.8	1.2	6.8	6.8
HT heat exchanger (total installed)	10.9	14.6	9.6	11.2	13.6	10.3	8.9	4.4	74	9.0	6.9
Baghouse filter	1.4	0	0.9	4.1	0	3.8	0	0	0	3.8	4.3
Condensing scrubber	2.1	0	13	6.8	Ő	64	Ő	0	Ő	64	7.2
Hot gas cleaning	0	3.0	0	0.0	30	0.1	23	75	3.0	0.1	0
Syngas processing	0	5.0	0	0	5.0	0	2.5	7.0	5.0	0	Ū
Compressor	0	0	0	16.0	0	16.5	0	0	0	14.0	17.0
Steam reformer	0	0	0	10.9	0	10.5	0	0	0	14.9	0
Catalytic autothermal reformer	0	0	24.5		0	24.5	0	0	0	-2.7	0
Shift reactor(a)	0	0	24.5	0	5.0	24.5	12.2	0	0	21.0	11.5
Silint reactor(s)	0	0	0	0	5.0	1.9	15.5	0	0	21.0	11.5
Selexol CO_2 removal	0	0	0	0	17.4	9.5	0	0	0	0	0
Mela and production	12.2	12.4	10.5	174	11.4	175	0	0	0	0	0
Make up compressor	13.3	12.4	10.5	17.4	11.4	17.5	0	0	0	0	0
Gas phase methanol	0	0	0	0	9.1	9.8	0	0	0	0	0
Liquid phase methanol	3.3	3.5	2.6	4.4	0	0	0	0	0	0	0
Recycle compressor	0	0	0	0.4	0.5	1.2	0	0	0	0	0
Refining	14./	15.3	11.8	19.1	17.8	19.5	0	0	0	0	0
Hydrogen production	0	0	0	0	0	0	01.7	0	0	25.1	26.0
PSA units $A + B$	0	0	0	0	0	0	31.7	0	0	35.1	26.9
HT ceramic membrane	0	0	0	0	0	0	0	10.1	7.5	0	0
Recycle compressor	0	0	0	0	0	0	15.3	0	0	6.2	13.1
Product compressor	0	0	0	0	0	0	6.3	23.2	17.1	14.1	7.4
Power generation											
Gas turbine + HRSG	35.4	31.5	54.5	0	0	0	46.6	0	54.3	0	54.9
Steam turbine + steam system	17.1	22.4	21.1	13.9	18.3	7.6	18.2	0	16.6	0	14.0
Expansion turbine	0	0	0	0	0	0	0	14.5	0	0	0
Total installed investment (MUS\$)	245.0	283.3	252.5	224.6	282.8	237.9	282.7	206.5	245.9	237.9	248.6
Total installed investment	221.5	256.2	228.3	203.1	255.7	215.1	255.6	186.7	222.4	215.1	224.8
corrected for lifetime (MUS\$)											
Biomass input (dry tonne/h)	80.0	80.0	80.0	80.0	80.0	80.0	80.0	80.0	80.0	80.0	80.0
Biomass input (MW _{th})	428.4	428.4	428.4	432.4	428.4	432.4	428.4	428.4	428.4	428.4	428.4
Load (h)	8000	8000	8000	8000	8000	8000	8000	8000	8000	8000	8000
Biomass input (PJ/year)	12.3	12.3	12.3	12.5	12.3	12.5	12.3	12.3	12.3	12.3	12.3
Annual costs											
Capital (MUS\$)	29.1	33.7	30.0	26.7	33.6	28.3	33.6	24.5	29.2	28.3	29.6
O&M	9.8	11.3	10.1	9.0	11.3	9.5	11.3	8.3	9.8	9.5	9.9
Biomass	24.7	24.7	24.7	24.9	24.7	24.9	24.7	24.7	24.7	24.7	24.7
Costs/income power	-12.8	-14.8	-25.1	0.0	-3.6	4.2	-17.4	0.2	-20.3	5.4	-17.3
Total annual costs (MUS\$)	50.8	54.9	39.7	60.6	66.0	66.9	52.1	57.7	43.5	67.8	46.8
Production											
Fuel output (MW HHV)	161.1	172.7	113.4	246.3	220.6	254.8	175.5	259.2	177.1	303.0	149.0
Power output (MW _e)	53.3	61.8	104.5	-0.1	14.9	-17.3	72.7	-0.7	84.4	-22.4	72.2
Efficiency fuel (%)	37.6	40.3	26.5	57.0	51.5	58.9	41.0	60.5	41.3	70.7	34.8
Efficiency power (%)	12.4	14.4	24.4	-0.0	3.5	-4.0	17.0	-0.2	19.7	-5.2	16.9
Efficiency total HHV (%)	50.0	54.7	50.9	56.9	55.0	54.9	57.9	60.3	61.0	65.5	51.6
Costs of fuel produced (US\$/GJ)											
80 MW _{th}	16.01	16.78	19.75	12.31	14.80	12.93	15.37	9.89	12.93	10.68	16.65
400 MW _{th}	10.95	11.03	12.16	8.55	10.39	9.11	10.32	7.72	8.53	7.77	10.92
1000 MW _{th}	9.85	9.67	10.45	7.61	9.36	8.14	9.11	7.36	7.57	7.03	7.03
2000 MW.	9.21	8.90	9.46	7.14	8.77	7.65	8.39	7.13	7.01	6.65	6.65
in	2.21	0.70	2.10	/.17	0.77	1.00	0.57	1.15	/.01	0.05	0.05

Costs in US\$2001.



Fig. 7. Breakdown of investment costs for 400 MW_{th} concepts.

8.9-14/GJ. The last rows of Table 6 show potential fuel production costs in smaller or bigger installations. Going to 1000 and 2000 MW_{th} scales the fuel production costs reach cost levels as low as US\$ 7.1–9.5/GJ for methanol and US\$ 7.0–8.4/GJ for hydrogen.

Feedstock costs account for 36-62% of the final fuel costs for the mentioned technologies. If a biomass price of US\$ 1.7/GJ could be realised (a realistic price for e.g. Brazil), methanol production costs would become US\$ 8.0-11/GJ, and hydrogen production costs US\$ 7.2-10/GJfor 400 MW_{th} concepts. On the other hand, when biomass costs increase to US\$ 3/GJ (short term Western Europe) the cost of produced methanol will increase to US\$ 10-16/GJ, and hydrogen to US\$ 9.4-14/GJ.

If the electricity can be sold as green power, including a *carbon neutral* premium, the fuel production costs for power co-producing concepts drops, where the green premium essentially pays a large part of the fuel production costs. A power price of US\$ 0.08/GJ would decrease methanol costs to US\$ -0.6-9.5/GJ and hydrogen costs to US\$ 1.9-4.6/GJ. Of course the decrease is the strongest for concepts producing more electricity. A green electricity scenario, however, may be a typical western European scenario. As such it is unlikely that it can be realised concurrent with biomass available at US\$ 1.7/GJ.

On the long term different cost reductions are possible concurrently [12]. Biomass could be widely available at US\$ 1.7/GJ, capital costs for a third plant built are 15% lower, and the large (2000 MW_{th}) plants profit from economies of scale. These reductions are depicted in Fig. 8: methanol concepts produce against US\$ 6.1-7.4/GJ, and hydrogen concepts

against US\$ 5.4–6.6/GJ. The influence of capital redemption on the annual costs has strongly reduced and the fuel costs of the different concepts lie closer together. Methanol 4 and hydrogen 3 give the lowest fuel costs.

In this long-term scenario, methanol produced from biomass costs considerably less than methanol at the current market, which is priced about US\$ 10/GJ [44]. Hydrogen from biomass resides in the cost range of hydrogen at a large natural gas reformer plant US\$ 5–9/GJ (small amounts of excess hydrogen from large refineries could cost down to US\$ 3/GJ [45]). Diesel and gasoline production costs vary strongly depending on crude oil prices, but for an indication: current gasoline market prices lie in the range US\$ 4–6/GJ [5]. Current diesel prices are around US\$ 5/GJ. Longer-term projections give estimates of roughly US\$ 8–11/GJ [46].

5.3. Biofuel FCV economy

For a well-to-wheel comparison of biofuels with gasoline, the distribution costs and performance in cars has to be considered, next to the biofuel production cost. This has been done in more detail by Faaij [47] and Ogden [3]. Over long distances, hydrogen is preferably distributed in liquefied form by tank trucks or, in future, pipelines. Costs for distribution and for the refuelling station are summarised in Table 7. Compared to gasoline ICEVs, methanol and hydrogen FCVs have similar fuel costs per km driven. However, the FCV will be more expensive: the fuel cell and necessary electricity system cost more than the corresponding elements in an ICEV [3,48]. The resulting total costs per km driven



Fig. 8. Optimistic view scenario. Different cost reductions are foreseeable: (1) biomass costs US1.7/GJ instead of US2/GJ, (2) technological learning reduces capital investment by 15% and (3) application of large scale (2000 MW_{th}) reduces unit investment costs.

depend on annual capital redemption, annual operating and maintenance costs and the annual amount of km driven. Both Williams [4] and Ogden projected these to amount US\$ cent 26 and US\$ cent 27/km for methanol and hydrogen, while gasoline costs US\$ cent 27/km (without tax).

Next to fuel efficiency, also storage capacity will be important, determining the range. Despite lower fuel economy, the methanol reformed H_2 FCV initially has a larger

 Table 7

 Build-up of delivered fuel costs and fuel costs per km driven

	Hydrogen FCV	Methanol FCV	Gasoline FCV
Production (US\$/GJ)	5–7	6–7	5–8 ^a
Distribution (US\$/GJ)	1 ^{b,c}	$2^{\mathbf{b},\mathbf{d}}$	1 ^b
Tank station (US\$/GJ)	5 ^b	1 ^b	0.5 ^b
Delivered (US\$/GJ)	11–13 ^e	9–10 ^e	6.5–9.5 ^f
Fuel economy (MJ HHV/km)	0.77 ^b	1.18 ^b	1.21 ^g
Fuel economy (US\$ cent/km)	1	1	1

^a BP [5]: 5, DOE/EIA: 8.

^b Ogden [3].

 $^{\rm c}$ Pipeline distribution of e.g. 30×10^3 N m 3 per day over 1 km; costs proportional to distance and inverse to transported volume.

^d Tank truck distribution.

^e Faaij [47] projects US\$ 8/GJ for hydrogen and US\$ 10/GJ for methanol. Ogden [3] projects US\$ 12 and 13/GJ. Differences are mainly due to production costs.

^f Or US\$ 36/GJ in the Netherlands when excise duty included.

^g Current gasoline ICEVs on average consume 2.8 MJ/km. Van Walwijk et al. [68] projects a large increase in fuel economy to 1 MJ/km, while DOE/IEA projects only a modest increase to 2.5 MJ/km. Gasoline reformed FCVs may achieve 1.21 MJ/km [3].

range, due to the larger storage capacity in its simple fluid form. On the other hand, onboard reforming is only an option if the reformer is flexible in providing hydrogen to the fuel cell, as fast or slow as it is being consumed by the fuel cell. If additional hydrogen storage would be necessary, the onboard reformer loses its advantage [49].

For on board hydrogen storage, currently two methods receive the most attention: compressed gas in storage tanks at high pressure or liquid hydrogen in insulated storage tanks at low temperature and pressure. Other methods based on metal hydrides, solid absorbents, and glass micro-spheres have potential advantages but are not as well developed. Hydrogen storage systems can be engineered to be as safe as the fuel systems in current automobiles [49].

6. Discussion and conclusions

Promising conversion concepts for the production of methanol and hydrogen from biomass have been evaluated. The concepts incorporate improved or new technologies for gas processing and synthesis and were selected on potential low cost or high energy efficiency. Some concepts explicitly co-produce power to exploit the high efficiencies of once-through conversion. The biofuel production plants were modelled using the Aspen+ flowsheeting program, and optimised towards internal heat demand and supply, surplus heat is converted to electricity. The models directly yielded the plant energy balance and dimensions of streams and units for the economic calculations.

Overall HHV energy efficiencies remain around 55% for methanol and around 60% for hydrogen production. Accounting for the lower energy quality of fuel compared to electricity, once-through concepts perform better than the concepts aiming at fuel only production. Also hot gas cleaning generally shows a better performance. Some of the technologies considered in this study are not yet fully proven/commercially available. Several units may be realised with higher efficiencies than considered here: new catalysts and carrier liquids could improve liquid phase methanol single pass efficiency. At larger scales, conversion and power systems (especially the combined cycle) may have higher efficiencies, but this has not been researched in depth.

The biofuel production costs are calculated by dividing the total annual costs of a system by the produced amount of fuel. Unit sizes, resulting from the plant modelling, are used to calculate the total installed capital of biofuel plants; larger units benefit from cost advantages. Assuming biomass is available at US\$ 2/GJ, a 400 MW_{th} input system can produce methanol at US\$ 9–12/GJ and hydrogen at US\$ 8–11/ GJ, slightly above the current production from natural gas prices. The outcomes for the various system types are rather comparable, although concepts focussing on optimised fuel production with little or no electricity co-production perform somewhat better. Hydrogen concepts using ceramic membranes perform well due to their higher overall efficiency combined with modest investment.

The biofuel production cost consists for about 50% of capital redemption, of which the bulk is in the gasification and oxygen system, syngas processing and power generation units. Further work should give more insight in investment costs for these units and their dependence to scale. The maximum possible scale of particularly the pressurised gasifier gives rise to discussion. The operation and maintenance costs are taken as a percentage of the total investment, but may depend on plant complexity as well. Long-term (2020) cost reductions mainly reside in slightly lower biomass costs, technological learning, and application of large scales (2000 MW_{th}). This could bring the methanol and hydrogen production costs in the range of gasoline/diesel and even lower: to US\$ 7 and 6/GJ, respectively. Availability of liquid phase methanol synthesis and ceramic membrane technologies are crucial to reach this cost level. R&D are necessary to verify the performance of these components.

Large-scale production facilities require very large volumes of feedstock. For a small country like the Netherlands, this would imply massive biomass import. Longdistance biomass transport will influence the biomass price, and subsequently the biofuel production costs as feedstock costs account for about 45% of the biofuel production costs. Advanced transportation technologies and logistic organisation of large-scale biomass import are currently researched within the department.

Hydrogen as the ultimate fuel for fuel cell vehicles, has a high fuel economy and low costs per km driven, and will certainly compete with gasoline ICEVs or FCVs. However, hydrogen requires new distribution infrastructure—which is the main bottleneck—and further development of on-board storage means. A methanol distribution system is probably easier to realise and FCVs fuelled by on-board reformed methanol will initially have a greater range. More research, focussing on biofuel use through well-to-wheel analysis, is needed to allow a clearer comparison of renewable fuels with their fossil competitors. Nevertheless, the present study has shown that biomass-derived methanol and hydrogen are likely to become competitive fuels tomorrow.

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References

- R.E. Katofsky, The Production of Fluid Fuels from Biomass, Center for Energy and Environmental Studies, Princeton University, Princeton, 1993.
- [2] E-lab, Running Buses on Hydrogen Fuel Cells: Barriers and Opportunities, Massachusetts Institute of Technology, July–September 2000.
- [3] J.M. Ogden, M.M. Steinbugler, T.G. Kreutz, A comparison of hydrogen, methanol and gasoline as fuels for fuel cell vehicles: implications for vehicle design and infrastructure development, J. Power Sources 79 (2) (1999) 143–168.
- [4] R.H. Williams, E.D. Larson, R.E. Katofsky, J. Chen, Methanol and hydrogen from biomass for transportation, with comparisons to methanol and hydrogen from natural gas and coal, PU/CEES Report 292, Center for Energy and Environmental Studies, Princeton University, Princeton, NJ, 1995, p. 47.
- [5] BP, BP World Review, www.bp.com, 2001.
- [6] H. Komiyama, T. Mitsumori, K. Yamaji, K. Yamada, Assessment of energy systems by using biomass plantation, Fuel 80 (2001) 707–715.
- [7] M.A. Paisley, M.C. Farris, J. Black, J.M. Irving, R.P. Overend, Commercial demonstration of the Battelle/FERCO Biomass gasification process: startup and initial operating experience, in: R.P. Overend, E. Chornet (Eds.), Proceedings of the 4th Biomass Conference of the Americas, Elsevier, Oxford, UK, 1998, pp. 1061– 1066.
- [8] R.H. Perry, D.W. Green, J.O. Maloney, Perry's Chemical Engineers' Handbook, 6th Edition, McGraw-Hill, Singapore, 1987.

- [9] P.T. Alderliesten, Systeemtstudie hoge-temperatuur gasreinigingdeelstudie 2.3: Alkalimetalen en overige spore-elementen, ECN, Petten, 1990, p. 42 and annexes.
- [10] S. Consonni, E. Larson, Biomass-gasifier/aeroderivative gas turbine combined cycles, part A: technologies and performance modeling, and part B: performance calculations and economic assessment, in: Proceedings of The American Society of Mechanical Engineers' 8th Congress on Gas Turbines in Cogeneration and Utility, Industrial and Independent Power Generation, Portland, 1994.
- [11] R. van Ree, A. Oudhuis, A. Faaij, A. Curvers, Modelling of a biomass integrated gasifier/combined cycle (BIG/CC) system with the flowsheet simulation programme ASPEN+, The Netherlands Energy Research Foundation ECN and Department of Science, Technology and Society, Utrecht University, Petten, 1995.
- [12] M.J.A. Tijmensen, The production of Fischer Tropsch liquids and power through biomass gasification, M.Sc. Thesis, Department of Science, Technology and Society, Utrecht University, Utrecht, The Netherlands, 2000, p. 45 and annexes.
- [13] K.M. van Dijk, R. van Dijk, V.J.L. van Eekhout, H. van Hulst, W. Schipper, J.H. Stam, Methanol from natural gas—conceptual design & comparison of processes, Delft University of Technology, Delft, 1995, p. 286.
- [14] Hydrocarbon Processing, Gas Processes '98, Hydrocarbon Process, 77 (4), 2000.
- [15] S.C. Mitchell, Hot gas cleanup of sulphur, nitrogen, minor and trace elements, IEA Coal Research, London, 1998, p. 84.
- [16] H. Klein Teeselink, P.T. Alderliesten, Systeemtstudie hoge-temperatuur gasreiniging—deelstudie 2.4: stofverwijdering, Stork Ketels, B.V., Hengelo, 1990, p. 108.
- [17] R.H. Williams, Cost-competitive Electricity from Coal with Nearzero Pollutant and CO₂ Emissions—Review Draft, PU/CEES, Princeton, NJ, 1998, p. 17.
- [18] L.R. White, T.L. Tompkins, K.C. Hsieh, D.D. Johnson, in: Proceedings of International Gas Turbine and Aeroengine Congress and Exposition on Ceramic Filters for Hot Gas Cleanup, Cologne, Germany, 1992, p. 8.
- [19] S.Q. Turn, C.M. Kinoshita, D.M. Ishimura, J. Zhou, T.T. Hiraki, S.M. Masutani, A review of sorbent materials for fixed bed alkali getter systems in biomass gasfier combined cycle power generation applications, J. Inst. Energy 71 (1998) 163–177.
- [20] D. Jansen, Systeemtstudie hoge-temperatuur gasreiniging—deelstudie 2.1: H₂S/COS-verwijdering, ECN, Petten, 1990.
- [21] K. Jothimurugesan, A.A. Adeyiga, S.K. Gangwal, in: Proceedings of Advanced Coal-Fired Power Systems Review Meeting on Advanced Hot-Gas Desulfurization Sorbents, Morgantown, West Virginia, 1996.
- [22] M.J.E. Verschoor, A.G. Melman, System Study High Temperature Gas Cleaning at IGCC Systems, NOVEM/TNO Milieu & Energie, 1991, p. 20.
- [23] H. Oonk, J. Vis, E. Worrell, A. Faaij, J.-W., Bode, The MethaHydroprocess—Preliminary Design and Cost Evaluation, TNO, The Hague, 1997.
- [24] P.S. Maiya, T.J. Anderson, R.L. Mieville, J.T. Dusek, J.J. Picciolo, U. Balachandran, Maximizing H₂ production by combined partial oxidation of CH₄ and water gas shift reaction, Appl. Catal. A: Gen. 196 (2000) 65–72.
- [25] J.N. Armor, Applications of catalytic inorganic membrane reactors to refinery products, J. Membr. Sci. 147 (1998) 217–233.
- [26] A. Cybulski, Liquid phase methanol synthesis: catalysts, mechanism, kinetics, chemical equilibria, vapor–liquid equilibria, and modeling—a review, Catal. Rev. Sci. Eng. 36 (4) (1994) 557–615.
- [27] Kirk-Othmer, Encyclopedia of Chemical Technology, 4th Edition, 1995.
- [28] USDOE, Commercial-scale Demonstration of the Liquid Phase Methanol (LPMEOHTM) Process—Clean Coal Technology Topical Report #11, US Department of Energy, 1999.
- [29] Haldor Topsoe, Hydrogen Production by Steam Reforming of Hydrogen Feedstocks, Haldor Topsoe, 1991.

- [30] A.I. LaCava, A.I. Shirley, R. Ramachandran, How to specify pressure-swing adsorption units—key components of PSA units, Chem. Eng. 105 (6) (1998) 110–118.
- [31] D.E. Fain, G.E. Roettger, High temperature inorganic membranes for separating hydrogen, in: Proceedings of the 9th Annual Conference on Fossil Energy Materials, Oak Ridge, 1995, pp. 185–193.
- [32] Parsons I&TG, Decarbonized fuel plants utilizing inorganic membranes for hydrogen separation, in: Proceedings of the Presentation at 12th Annual Conference on Fossil Energy Materials, Parsons Infrastructure and Technology Group, Inc., Knoxville, 12–14 May 1998.
- [33] S. Consonni, E. Larson, Biomass-gasifier/aeroderivative gas turbine combined cycles. Part A: technologies and performance modeling, in: Proceedings of the American Society of Mechanical Engineers' 8th Congress on Gas Turbines in Cogeneration and Utility, Industrial and Independent Power Generation, Portland, 1994.
- [34] M. Rodrigues de Souza, A. Walter, A. Faaij, An analysis of scale effects on co-fired BIG-CC system (biomass + natural gas) in the state of São Paulo/Brazil, in: S. Kyritsis, A.A.C.M. Beenackers, P. Helm, A. Grassi, D. Chiaramonti (Eds.), Proceedings of 1st World Conference on Biomass for Energy and Industry, James & James, London, 2000, pp. 813–816.
- [35] A. Faaij, B. Meuleman, R. van Ree, Long term perspectives of biomass integrated gasification with combined cycle technology, The Netherlands Agency for Energy and the Environment Novem, Utrecht, 1998, p. 93 and annexes.
- [36] K. Hagihara, H. Mabuse, T. Watanabe, M. Kawai, M. Saito, Effective liquid-phase methanol synthesis utilizing liquid–liquid separation, Energy Conv. Manage. 36 (6–9) (1995) 581–584.
- [37] Gas Turbine World, Gas Turbine World 1997 Handbook, Pequot Publishing, Fairfield, Canada, 1997, p. 256.
- [38] E.D. Larson, C.I. Marrison, Economic scales for first-generation biomass-gasifier/gas turbine combined cycles fueled from energy plantations, J. Eng. Gas Turbines Power 119 (1997) 285–290.
- [39] M.S. Peters, K.D. Timmerhaus, Plant Design and Economics for Chemical Engineers, 3rd Edition, McGraw-Hill, New York, USA, 1980, p. 973.
- [40] E. Larson, S. Consonni, T. Kreutz, Preliminary economics of black liquor gasifier/gas turbine cogeneration at pulp and paper mills, in: Proceedings of the 43rd ASME Gas Turbine and Aeroengine Congress, Exposition and Users Symposium, Stockholm, 1998.
- [41] A.P.C. Faaij, Energy from Biomass and Waste, Ph.D. Thesis, Department of Science, Technology and Society, Utrecht University, Utrecht, The Netherlands, 1997, p. 180.
- [42] A. Agterberg, Bio-energy Trade: Possibilities and Constraints on Short and Longer Term, Department of Science, Technology and Society, Utrecht University, Utrecht, 1998, p. 81.
- [43] D.O. Hall, F. Rosillo-Calle, R.H. Williams, J. Woods, Biomass for energy: supply prospects, in: T.B. Johansson et al. (Eds.), Renewable Energy Sources for Fuels and Electricity, Washington, DC, USA, 1992.
- [44] Methanex, www.methanex.com, 2001.
- [45] J.M. Ogden, Developing an infrastructure for hydrogen vehicles: a Southern California case study, Int. J. Hydrogen Energy 24 (1999) 709–730.
- [46] F. Hendriks, Natural gas as a feedstock for automotive fuels—an alternative to crude oil, M.Sc. Thesis, Department of Science, Technology and Society, Utrecht University, Utrecht, The Netherlands, 2000, p. 85 and annexes.
- [47] A.P.C. Faaij, Long term perspectives for production of fuels from biomass integrated assessment and RD&D priorities, 2000.
- [48] AMI, Beyond the Internal Combustion Engine—The Promise of Methanol Fuel Cell Vehicles, American Methanol Institute, Washington, DC, 2000, p. 60.
- [49] J.E. Brydges, A hydrogen fueling station in 2005—will it happen? How do we get from here to there? M.Sc. Thesis, Massachusetts Institute of Technology, MA, USA, 2000, p. 70 and annexes.

- [50] OPPA, Assessment of costs and benefits fo flexible and alternative fuel use in the US transportaion sector, Technical report 5: costs of methanol production from biomass, US DOE, Washington DC, USA, 1990.
- [51] R. Knight, Personal Communications on the Pressurized Renugas Gasifier for Different Conditions, Institute of Gas Technology, Chicago, 1998.
- [52] R. Breault, D. Morgan, Design and Economics of Electricity Production from an Indirectly Heated Biomass Gasifier, Battelle Memorial Institute, Tecogen, Inc., Waltham, USA, 1992.
- [53] Paisley, Personal Communication, Battelle Columbus Laboratory, Columbus, USA, 1994.
- [54] S.M. Walas, Rules of Thumb, Selecting and Designing Equipment, Chemical Engineering, 16 March 1987, pp. 75–81.
- [55] D.L. King, C.E. Bochow Jr., What should an owner/operator know when choosing an SMR/PSA plant?, Hydrocarbon Process., May 2000, Volume 79 (5), pp. 39–48.
- [56] T.S. Christensen, Primdahl II Improve syngas production using autothermal reforming, Hydrocarbon Process. 73 (3) (1994) 39–44.
- [57] C. Hendriks, Carbon dioxide removal from coal-fired power plants, Ph.D. Thesis, Department of Science, Technology and Society, Utrecht University, Utrecht, The Netherlands, 1994, p. 260.
- [58] D.E. Fain, in: Proceedings of 15th Annual Membrane Technology/ Planning Conference on the Amazing Promise of Inorganic Membranes, Membrane Technology, Boston, 1997.
- [59] K.D. Adcock, D.E. Fain, D.L. James, B.B. Marshall, M.R. Phillips, L.E. Powell, T. Raj, Ceramic membrane for high temperature gas separation, in: Proceedings of the 13th Annual Conference on Fossil Fuel Energy Materials, Oak Ridge, 1999.

- [60] M.R. DeLallo, T.L. Buchanan, M.G. Klett, M.D. Rutkowski, J.S. White, Decarbonized Fuel Production Facility Baseline Plant—Draft Letter Report, Parsons Power Group, Inc., Reading, PA, 1998, p. 14 and annexes.
- [61] OECD, National Accounts—Main Aggregates, 1997 Edition, Paris, 1996, p. 170.
- [62] A. Faaij, Rv. Ree, A. Oudhuis, Gasification of biomass wastes and residues for electricity production: technical, economic and environmental aspects of the BIG/CC option for the Netherlands, Department of Science, Technology and Society, Utrecht University, Utrecht, 1995.
- [63] R. van Ree, Air Separation Technologies—An Inventory of Technologies for 'Pure' Oxygen Production for Pulverised Coal Combustion in a CO₂(g)/O₂(g)-atmosphere, ECN, Petten, 1992, p. 94 (in Dutch).
- [64] P. van Ooijen, Personal Communication, Azko Nobel, February 2001.
- [65] H.C. de Lathouder, March, Personal Communication on Economy of Methanol Production, Stamicarbon, DSM Licensing Subsidiary, Geleen, 2001.
- [66] P.J.A. Tijm, W.R. Brown, E.C. Heydorn, R.B. Moore, in: Proceedings of the Presentation at American Chemical Society Meeting on Advances in Liquid Phase Technology, San Francisco, 13–17 April 1997.
- [67] Y. Solantausta, T. Bridgewater, D. Beckman, Electricity Production by Advanced Biomass Power Systems, Technical Research Centre of Finland (VTT), Finland, 1996.
- [68] M. van Walwijk, M. Bückmann, W.P. Troelstra, P.A.J. Achten, Automotive fuels survey. Part 2. Distribution and use, IEA/AFIS Operated by Innas bv, Breda, The Netherlands, 1996, p. 319 and annexes.