Prepared for the 4th Biomass Conference of the Americas, Oakland, California, Aug 29 - Sept. 2, 1999

Production of methanol and hydrogen from biomass via advanced conversion concepts - preliminary results -

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ABSTRACT

A limited set of promising conversion concepts for the production of methanol and hydrogen from biomass has been identified and their technical and economic performance has been evaluated. Technology reviews and preliminary performance and cost estimates suggest that investment costs could be reduced compared to concepts that are based on commercially available conversion technology. Overall energy efficiencies remain in the range of 50-60% (HHV basis). 'Once through' concepts (for example with Liquid Phase Methanol synthesis) do not result in systems with higher net energy efficiencies, although not all possible improvement options have been incorporated in this, preliminary, work.
Hydrogen production making use of ceramic membrane technology that allows for separating H_2 at high temperatures from a mixed gas stream, is particularly interesting technology, although ceramic membranes are not commercially available at present.
Economies of scale do have a considerable effect on the (economic) performance. When 1000-2000 MW_{th} production facilities are considered, the production costs of methanol and hydrogen drop to impressive cost levels. Assuming biomass is available at 2-3 U$/GJ, the methanol production costs could lay between 7-10 U$/GJ and below 6 up to 8 U$ for hydrogen. Considering the high efficiency with which methanol and hydrogen transportation fuels can be used (in fuel cell vehicles) and the fact that gasoline and diesel prices may fluctuate between 5-10 U$/GJ, the estimated costs for bio-methanol and bio-hydrogen could make these relatively competitive fuels in the foreseeable future.

1. INTRODUCTION AND RATIONALE

Methanol and hydrogen produced from lignocellulosic biomass have been identified as promising sustainable fuels. Both fuels are well suited for use in fuel cell vehicles (FCV). Besides high efficiencies, about a factor 2-3 better than current internal combustion engine vehicles (ICEV), the emission levels of such vehicles are zero or near zero. When MeOH and H_2 are biomass derived, the overall energy chain is almost GHG neutral as well. Fuel cell vehicles may become commercially available within 5-10 years from now.

Starting from available technologies, bio-methanol/hydrogen production facilities typically consist of the following steps: Gasification → gas cleaning → reforming of hydrocarbons → shift to obtain appropriate $CO:H_2$ ratio's → gas separation (H_2 or CO_2, e.g. with PSA technology) or (gas phase) methanol synthesis.

Previous analysis has shown that 400 MW_{th} (0.6 Mtonne dry biomass/yr; biomass cost 2 U$/GJ delivered) input conversion facilities for the production of MeOH and H_2 could obtain the following performance [14]: 12 - 15 US$/GJ for MeOH (net energy efficiency: between 54-58% HHV basis) and 9 - 12 US$/GJ for H_2 (net energy efficiency: 56-64% HHV-basis). (All costs in this paper are given in 1995 US$.) When cultivated biomass is used as a

feedstock, MeOH and H_2 are not competitive with their production from natural gas (about 9 and 7 U$/GJ respectively) and current gasoline and diesel prices (about 5-7 U$/GJ).

Logically, with higher biomass costs, the fuel costs increase. Feedstock costs account for about 30 % of the final fuel costs for the mentioned technologies. Decreasing biomass production costs may be difficult and therefore increasing the efficiency and decreasing conversion costs are extremely relevant to obtain competitive cost levels. High energetic efficiencies are also important to limit the amount of land needed to produce the biomass feedstock needed.

This work focuses on identifying conversion concepts that may lead to higher overall efficiencies and lower costs. Improved performance may be obtained by:

1. Applying improved or new (non commercial) technologies. Examples are the use of Catalytic 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.
2. Combined fuel and power production by so-called 'once through' concepts. Combined fuel and power production may lead to lower cost and possibly higher overall thermal efficiencies because of cheaper reactor capacity and generally reduction of internal energy consumption of the total plant.
3. 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 from biomass as well.

This paper gives preliminary results of analyses carried out so far.

2. APPROACH

The work is carried out in 5 steps:

1. Technology assessment and selection of various concepts. The review includes technologies that are not applied commercially (examples are Catalytic Autothermal Reforming, new shift reaction catalysts, Liquid Phase methanol production, HT gas cleaning, high temperature gas separation techniques, and improved O_2 production).
2. Consulting of manufacturers and experts for performance and cost data of various components.
3. Creation of ASPEN-plus models to evaluate performance and carry out sensitivity analyses. Particular attention is paid to the heat integration of the concepts. The ASPEN models are created including heat exchanger networks. Such an approach does not lead to ideal heat utilization (as would follow from pinch analyses), but is considered to give more detailed insight in the number of heat exchangers needed, as well as a more realistic performance estimate. Both aspects are relevant for the investment costs of the concepts considered.
4. Cost analyses based on component costs (including scale factors and capacity ranges).
5. Chain analyses for calculating costs of energy services delivered, energy balance and GHG emissions.

3. RESULTS

3.1 System selection

Technology assessment and screening of technologies and concepts in more qualitative terms led to a selection of 11 conversion concepts. The eleven concepts selected are considered for their potential low cost and/or potentially higher energy efficiency compared to the conventional technologies mentioned. The concepts are briefly summarized in tables 1 and 2. Two biomass gasifiers are considered for syngas production: the IGT (Renugas) pressurized gasification using oxygen as oxidant and the indirect gasification concept developed at the Batelle Columbus Laboratories. Both concepts produce medium calorific gas, undiluted by atmospheric nitrogen. The pressurized concept is considered in two modes: one standard and one aimed to produce hydrogen rich syngas. Main parameters are given in table 3.
The concepts considered include 'once through' liquid phase methanol production (LPMeOH), as developed by Air Products [13] and advanced gas separation technology for production of hydrogen. LPMeOH synthesis could be about 40-50% cheaper per unit of methanol production capacity compared to more conventional gas phase methanol synthesis. 'Once through' conversion efficiencies of the fractions of $CO+H_2$ in the fuel gas could go up to 70% at higher pressure (about 90 bar) and with some steam addition. Unconverted gas (as well as CH_4) can subsequently be used for power generation in a combined cycle and for raising steam (needed for the gasifier and drying wet biomass). [2,13]
Ceramic membranes are under development for hydrogen separation from hot gases. A unique feature of such membranes (generally Al_2O_3 with very small pore diameters) is that in the presence of steam during hydrogen removal a shift reaction can occur on the surface of the membrane, thereby combining shift and hydrogen removal in one device. Preliminary cost estimates for ceramic membranes suggest that costs may amount about 30 U$/kW H_2 throughput capacity, once they become commercially available [12].

Other 'advanced' technology considered includes hot gas cleaning (instead of quenching and scrubbing) and Catalytic Autothermal Reforming (CAR) instead of Steam Reforming (SMR). CAR is particularly interesting when oxygen is available (IGT-cases only); investment costs are about half of SMR on throughput basis.

Table 1: Methanol production concepts.

Concept	Gas cleaning	Reforming	Shift	MeOH	Power generation
1. IGT - max H_2 ☐ MeOH + power, once through	quench	no	no	LPMeOH	combined cycle
2. IGT ☐ MeOH + power, once through	hot (450 °C)	CAR	no	LPMeOH (with steam addition)	combined cycle
3. IGT ☐ MeOH + power, once through	quench	no	no	LPMeOH (with steam addition	combined cycle
4. BCL ☐ MeOH + power	quench	SMR	no	LPMeOH (steam addition and CO recycle)	steam cycle
5. IGT	hot (450 °C)	CAR	partial	gas phase	steam cycle

☐ MeOH only					
6. BCL ☐ MeOH only	quench	SMR	partial	gas phase	steam cycle

Table 2: Hydrogen production concepts.

Concept	Gas cleaning	Reforming	Shift	H_2 separation	power generation
1. IGT ☐ H_2 + power	hot (450 °C)	no	yes	PSA	combined cycle
2. IGT - max H_2 ☐ H_2 + power, once through	hot (800 °C)	no	no	ceramic membrane + internal shift	expansion of purge gas
3. IGT ☐ H_2 + power, once through	hot (450 °C)	no	no	ceramic membrane + internal shift	combined cycle
4. BCL ☐ H_2 only	quench	SMR	yes	PSA	steam cycle
5. BCL ☐ H_2 + power	quench	no	yes	PSA	combined cycle

Table 3: Main characteristics of biomass gasifiers considered [14].

	IGT		IGT - max H_2 [7]		BCL	
Bed type	Bubbling fluidized bed				ACFB indirect	
Steam (kg/kg dry feed)	0.3		0.8		0.02	
Oxygen (kg/kg dry fuel)	0.3		0.38		-	
Exit temperature (°C)	900		920		860	
Pressure (bar)	34.5		25		near atm.	
Composition (mole % [dry])						
H_2O	32	[-]	48	[-]	20	[-]
H_2	21	[31]	24	[45]	17	[21]
CO	15	[22]	11.5	[22]	37	[46]
CO_2	24	[35]	16	[32]	9	[11]
CH_4	8	[12]	0.5	[1]	13	[16]
C2+	0.3	[0.5]	-	[-]	5	[6]

3.2 System calculations [5]

Table 4 summarizes the, preliminary, main outcomes of the flowsheet models. The 'once through' concepts do not result in higher overall energy efficiencies than the concepts aimed for fuel only production. This is a logical result since the efficiency by which fuel gas from the gasifier is converted to methanol is generally comparable to the efficiency of a combined cycle (overall between 40-50% depending on the gas composition). Advantages of those concepts should therefore particularly come from lower investment costs due to a simpler set-up. Similar reasoning holds for the concepts that combine hydrogen production with power generation.

Table 4: Preliminary results of the ASPEN-plus performance calculations for 427 MW_{th} input HHV, equivalent to 400 MW_{th} LHV for biomass with a 30% moisture content) systems for the methanol and hydrogen production concepts considered.

Concept	Output (MW)		Net energy efficiency* (%; HHV)
	Fuel (HHV)	Net electricity	
Methanol			
1. IGT - max H_2, quench, LPMeOH, combined cycle	149	36	43.3
2. IGT, hot gas cleaning, CAR, LPMeOH (steam addition), combined cycle	179	41	51.5
3. IGT, quench, LPMeOH (steam addition), combined cycle	114	84	46.4
4. BCL, quench, SMR, LPMeOH (steam addition & recycle), steam cycle	262	-15 (input)	58
5. IGT, hot gas cleaning, CAR, shift, gas phase methanol synthesis, steam cycle	207	12	51.3
6. BCL, quench, SMR, shift, gas phase methanol synthesis, steam cycle	254	-15 (input)	55.8
Hydrogen			
1. IGT, hot gas cleaning, no reforming, shift, PSA, combined cycle	176	62	55.7
2. IGT - max H_2, hot gas cleaning, ceramic membrane, expansion purge gas	212	24	55.2
3. IGT, hot gas cleaning, ceramic membrane with steam addition, combined cycle	176	61	55.5
4. BCL, quench, SMR, shift, PSA, steam cycle	269	-14 (input)	59.7
5. BCL, quench, shift, PSA, combined cycle	149	58	48.5

* Net energy efficiency = Energy output / energy input. Only the 427 MW_{th} biomass is taken as input, electricity inputs are subtracted from the fuel output.

3.3 Economics

A simple and preliminary economic evaluation has been carried out for the concepts considered. Based on literature data for various key components and by using scaling factors investment costs of the concepts are determined. Overall cost factors for e.g. engineering, fees, contingencies are used to obtain turnkey costs.

Costs of methanol and hydrogen produced are calculated for the 427 MW_{th} scale systems, assuming a fixed power price. Main assumptions: interest rate: 10%, economic lifetime: 15 years (for a facility built to operate 25 years), load factor: 90%, electricity price: 4 U$ct/kWh (set equal for purchase from as well as delivery to the grid). Biomass price delivered at plant gate: 2 U$/GJ. The results are given in table 5. Overall cost factors include civil works, control systems, piping, etc (33% of investments) and engineering, building interest, fees/overheads, start-up costs and contingencies (40% of total investment costs). Literature sources used are [3,8,11,14,15].

As can be seen, for the scale considered fuel costs range between 8,5 - 12 U$/GJ for methanol. For hydrogen, the cost range is 7,5 - 9 U$/GJ. Given the fact that the uncertainties in the investment costs estimates are considerable (± 30% should be taken into account as a rule of thumb) the outcomes for the various system types are rather comparable.

For methanol, the somewhat lower overall energy efficiency of most of the once-through processes is partly compensated by the lower investment costs for those systems (mainly

because 'once through' processes have a much smaller throughput and no compressing costs of the recycle stream). When using the same gasifier and cleanup, the LPMeOH process is clearly cheaper than gas phase synthesis, because the setup of the reactor is much simpler and preceding CO_2 removal is not necessary. Still, the differences in investment costs between the various systems are not so large. This can be explained by the relatively large fraction of the costs represented by the pre-treatment, gasification and gas cleaning in all cases (generally over 50% of the total investment costs). The equipment needed to convert the fuel gas to methanol makes a relatively modest contribution to the total investment costs.

For both methanol and hydrogen, our estimates for the cost of biofuels production are 20-30% lower than earlier estimates of Williams *et al.* [14] in cases where the process designs are essentially the same. Our lower results are explained by the larger scale of our facility (427 MW_{th} versus 400 MW_{th}) and our lower capital charge rate (13.1%/year versus 15.1%/year).

3.4 Scaling

As argued, the scale of the conversion system is an important factor in the overall economic performance. This issue has been studied for e.g. BIG/CC systems. Various studies have shown that the economies of scale of such units can offset the increased costs of biomass transport up to capacities of several 100's of MW_{th} [3,10]. The same reasoning holds for the fuel production concepts described here. The last rows of table 5 show potential fuel production costs when a simple scaling factor is used for the investment costs. Such economies of scale would certainly apply for most of the equipment considered (such as reformers, combined cycles and most gasification and gas cleaning equipment). Going to 1000 and 2000 MW_{th} scales the fuel production costs reach impressive cost levels as low as 6-8 U$/GJ for methanol and below 6 U$/GJ for hydrogen.
Diesel and gasoline production costs vary strongly depending on crude oil prices, but for an indication: current gasoline production prices are in the range of 7 U$/GJ. Current diesel prices are around 5 U$/GJ. Longer-term projections give estimates of roughly 25 - 35 U$ct/liter, or 8-11 U$/GJ.

It should however be fully realized that production facilities of 1000-2000 MW_{th} require very large volumes of feedstock; 1.5 up to 3 Mtonne of dry biomass per year (190 - 380 tonne/hour). Biomass availability will be a limitation for most locations for such large-scale production facilities, especially in the shorter term. In the longer term, if biomass production systems become more commonplace, this can change. It should be realized as well that such large-scale systems are not without precedent. In Brazil various large-scale sugar/ethanol plants have a biomass throughput of 1-3 Mtonnes of sugarcane per year (and concentrated in a production season that covers less than half a year). Also large paper and pulp complexes have comparable capacities.
Furthermore, international (intercontinental) transport of biomass could be considered. Studies have indicated that the costs and energy-use of international biomass transport are not necessarily prohibitive. Energy needs for a complete (intercontinental) transport chain can remain below 10% of the energy content of the biomass and costs can increase about 1-1.5 U$/GJ when intercontinental shipment of biomass is considered [1]. In case biomass costs are increased to 3 U$/GJ delivered (e.g. with interregional transport), the fuel production costs may still remain in a competitive range with gasoline and diesel.

Table 5: preliminary economic analyses for the concepts considered. Main assumptions are given in the text. Costs are in 1995 U$.

	SYSTEM		MeOH	1	2	3	4	5	6	H_2	1	2	3	4	5
	Total pre-treatment	(MU$)		18,8	18,8	18,8	18,8	18,8	18,8		18,8	18,8	18,8	18,8	18,8
GASIFICATION SYSTEM	BCL gasifier (including feeding)	.					13,0		13,0					13,0	13,0
	IGT gasifier	.		30,0	30,0	30,0		30,0			30,0	30,0	30,0		
	Oxygen plant	.		22,0	22,0	22,0		22,0			22,0	22,0	22,0		
GAS CLEANING	tar cracker	.					7,7		7,7					7,7	7,7
	baghouse filter	.		3,9		3,9	3,9		3,9					3,9	3,9
	condensing scrubber	.		6,5		6,5	6,5		6,5					6,5	6,5
	Hot gas cleaning	.			9,7			9,7	0,0		9,7	9,7	9,7		
SYNGAS PROCESSING	Fuel gas compressor	.		9,1	7,2	7,1	11,5		8,8				4,5	9,3	10,8
	Steam reformer	.					17,0		15,4					14,8	
	Catalytic Autothermal Reformer	.			10,1			10,1	0,0						
	Shift reactor(s)	.						2,4	1,7		3,8			5,0	3,4
METHANOL PRODUCTION	Reformer feed compressor	.					11,2	7,4	7,0						
	Gas phase methanol production	.						34,8	43,4						
	CO_2 removal	.						26,7	14,3						
	Liquid Phase Methanol Production	.		13,9	15,9	11,6	20,7								
HYDROGEN PRODUCTION	PSA recycle compressor	.									3,7			1,6	1,0
	PSA units (with CO_2 removal)	.									12,1			16,3	10,8
	HT ceramic membrane	.										5,1	5,0		
	Hydrogen compressor	.									1,4	5,1	4,6	3,7	2,3
POWER GENERATION	gas turbine (incl. generator)	.		13,5	11,1	23,0					19,7		26,4		20,6
	HRSG + piping & cooling	.		7,3	9,5	8,7	4,8	9,2	5,5		8,2	6,0	3,7	5,4	7,8
	Steam Turbine + condensor	.		4,5	6,0	6,5	3,2	5,5	3,1		3,1	3,0	1,7	3,2	4,7
	SUBTOTAL INVESTMENT	.		129,5	139,3	137,9	118,2	175,6	149,1		132,5	99,7	126,4	109,1	111,2
	OVERALL COSTS (33% of inv.)	.		42,7	46,0	45,5	39,0	57,9	49,2		43,7	32,9	41,7	36,0	36,7
	OVERALL COSTS (40% of total inv.)	.		68,9	74,1	73,4	62,9	93,4	79,3		70,5	53,0	67,2	58,0	59,1
	TOTAL INVESTMENT	**(MU$)**		**241,1**	**259,4**	**256,8**	**220,1**	**326,9**	**277,7**		**246,6**	**185,6**	**235,4**	**203,1**	**207**
	Fuel output	(MW HHV)		149	179	114	262	207	254		176	212	176	269	149
	Power output	(MW_e)		36	41	84	-15	12	-15		62	24	61	-14	58
	Efficiency fuel (HHV)	(%)		34,9	41,9	26,7	61,4	48,5	59,5		41,2	49,6	41,2	63,0	34,9
	Efficiency power (HHV)	(%)		8,4	9,6	19,7	-3,4	2,8	-3,5		14,5	5,6	14,3	-3,3	13,6
ANNUAL COSTS	capital costs	(MU$/yr)		29	31	31	26	39	33		29	22	28	24	25
	O & M (4% of investment)	(MU$/yr)		10	10	10	9	13	11		10	7	9	8	8
	Fuel costs	(MU$/yr)		25	25	25	25	25	25		25	25	25	25	25
	Income/costs power	(MU$/yr)		12	13	27	-5	4	-5		20	8	20	-5	19
FUEL COSTS	**Costs of fuel produced**	**(U$/GJ HHV)**		**11,97**	**10,22**	**11,73**	**8,51**	**12,19**	**10,05**		**8,67**	**7,60**	**8,37**	**7,92**	**9,06**
	Costs of fuel for 80 MWth	.		*17,80*	*15,44*	*19,84*	*11,54*	*17,88*	*13,99*		*13,71*	*10,75*	*13,19*	*10,64*	*14,07*
	Costs of fuel for 1000 MWth	.		*9,96*	*8,42*	*8,93*	*7,47*	*10,23*	*8,69*		*6,92*	*6,51*	*6,71*	*6,99*	*7,34*
	Costs of fuel for 2000 MWth	.		*8,66*	*7,25*	*7,12*	*6,79*	*8,96*	*7,82*		*5,80*	*5,81*	*5,64*	*6,38*	*6,22*

4. DISCUSSION AND CONCLUSIONS

4.1 Further work

Various issues are to be evaluated in further detail:

- Liquid Phase methanol production may be realized with even higher once through efficiencies than considered here. Research suggests that other catalysts and carrier liquids can improve the single pass syngas conversion efficiency up to 95%[4]. Also reforming prior to methanol synthesis could be considered.
- Improvement of the conversion systems thermal efficiencies at larger scales is not taken into account so far. Such effects will certainly occur for the power generation part [3]. This effect will be in favor of larger systems and the 'once-through' concepts in particular.
- So far, scale effects are only taken into account by using scale factors by component. An effect that has been excluded is that at 1000 - 2000 MW_{th} capacities, various components will have to be realized as multiple units. This will for example be the case for the atmospheric gasification and gas cleaning equipment. Pressurized equipment will become relatively cheaper at those larger scales.
- More detailed data on the impact of larger transport distances on the biomass costs need to be incorporated.

Finally, this analysis did not include the use of methanol and hydrogen in fuel cell vehicles. Hydrogen is the optimal fuel for PEM fuel cells (generally considered for automotive applications), since no on board reforming is needed. Hydrogen, however, requires new infrastructure, e.g. with high pressure or cooled storage or storage in carbon or hydrides which are both not commercially available yet. Hydrocarbons produced from biomass via Fischer Tropsch synthesis (see e.g. [9]) can also be reformed and applied in fuel cells. This comes with some efficiency penalty compared to hydrogen, so the performance of hydrogen FCV's is better. However, for liquid hydrocarbons no adaptations of the distribution infrastructure are needed. A full system analysis is required to compare the pro's and con's of both quite different options.

4.2 Conclusions

A limited set of promising conversion concepts for the production of methanol and hydrogen from biomass has been identified and their technical and economic performance has been evaluated. Technology reviews and preliminary performance and cost estimates suggest that investment costs could be reduced compared to concepts that are based on commercially available conversion technology. Overall energy efficiencies remain in the range of 50-60% (HHV basis). Once through concepts do not result in systems with higher net energy efficiencies, although not all improvement options have been incorporated in this, preliminary, work.

Hydrogen production making use of ceramic membrane technology that allows for separating H_2 at high temperatures from a mixed gas stream is particularly interesting technology, although ceramic membranes are not commercially available at present. Scale has a considerable effect on the (economic) performance of the production systems considered. When 1000-2000 MW_{th} production facilities are considered, the production costs of methanol and hydrogen drop to impressive cost levels. Assuming biomass is available at 2-3 U$/GJ, the methanol production costs could lay between 7-10 U$/GJ and below 6-8 U$ for hydrogen. Considering the high efficiency with which methanol and hydrogen transportation fuels can be used (in fuel cell vehicles) and the fact that gasoline and diesel prices may fluctuate between 5-10 U$/GJ, the estimated costs for bio-methanol and bio-hydrogen could make these

relatively competitive fuels in the foreseeable future. In case biomass supplies are limited production of those fuels in combination with fossil fuels as feedstock could be considered as well [11].

Acknowledgements
Part of this work was carried out by André Faaij during a working period at the Center for Energy and Environmental Studies of the Princeton University. CEES is thanked for it's hospitality and stimulating working environment. Air Products, ORNL, IGT and many other companies and institutes are thanked for providing information.
The participation of André Faaij and Carlo Hamelinck in this work was funded through the National Research Programme on Global Air Pollution and Climate Change (NOP/MLKII) of the Netherlands, the Foundation of Technical Sciences (STW) of the Netherlands, and NOVEM (Dutch Organisation for Energy and Environment) who is responsible for the new research programme for introduction of sustainable liquid and gaseous energy carriers in the Dutch energy system. Eric Larson and Tom Kreutz thank the W. Alton Jones Foundation, the Energy Foundation, and the Geraldine R. Dodge Foundation for financial support.

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