

Heat-Integrated Novel Process of Liquid Fuel Production from Bioresources – Process Simulation and Costing Study

Iris Vural Gürsel^a, Qi Wang^a, Timothy Noël^a, Gunther Kolb^a, Volker Hessel^{*a}, Andre C. van Veen^b

^aEindhoven University of Technology, Department of Chemical Engineering and Chemistry, Micro Flow Chemistry & Process Technology, Den Dolech 2, 5600MB Eindhoven, The Netherlands

^bUniversity of Warwick, School of Engineering, Library Road, Coventry CV4 7AL, UK
 v.hessel@tue.nl

Innovative ways of providing novel sustainable solutions to reduce the emissions and consumption of exhaustible resources is called for. Production of liquid fuels from bioresources satisfies this call and it is aimed to be achieved with BIOGO project. Proposed process involves coupled reforming of biogas and bio oil followed by methanol production which is then converted to gasoline. This novel route through methanol eliminates challenging separation steps of Fischer-Tropsch process. Decentralized production in modularized small scale plants with process intensified equipment is considered. Process simulation reveals that carbon efficiency of 56 % and energy efficiency of 42 % can be achieved. Cost calculations show that with the total cost of production of gasoline at € 0.5/L; it is economically competitive with petroleum-based production and more economical than Fischer-Tropsch process. Comparison of 35 decentralized small modular plants with one centralized large scale plant reveals that the reduced construction time and operating cost of decentralized plants can outweigh the higher investment cost.

1. Introduction

Concerns about the depletion of fossil fuel reserves, increased energy demand, society's pressure for clean and sustainable production make use of bioresources as energy source an attractive solution. Although, recently there has been increasing research on renewable energy sources, still around 90 % of the world energy needs are met with non-renewable sources (IEA, 2011).

Bioenergy is the largest source of renewable energy and can provide heat, electricity as well as transport fuels. International Energy Agency foresees that biofuels contribution to total transport fuel could increase from 2 % today to 27 % by 2050 (IEA, 2011). The scenario suggests that this will be possible by advanced biofuel technologies that are not yet commercially available. These advanced (second generation) biofuels manufactured from various types of biomass, which are not competitive to food production, can substitute current fossil derived gasoline and diesel fuels (Iglesias Gonzalez et al., 2011). The two main catalytic processes for biomass to liquid fuel production are Fischer-Tropsch (FT) and Methanol-to-hydrocarbons processes. FT is an established process already applied on large scale worldwide in coal and natural gas based plants (Iglesias Gonzalez et al., 2011). There is one commercial methanol to gasoline plant using methanol produced from natural gas (Tabak and Yurchak, 1990). Methanol to olefins process is also developed. A further development is the Mobil olefins to gasoline/diesel process. They are at demonstration phase (Tabak and Yurchak, 1990).

BIOGO, a new project funded by the European commission, proposes to combine bio oil and biogas sources to produce syngas which is then converted to methanol. The methanol produced as intermediate is finally transformed into liquid fuels with a sharper product distribution than the ones from the FT process. The novelty of this route comes also from coupled syngas generation. Decentralized production in modular plant environment utilizing flow reactors is considered. In this paper, results from the preliminary study done to analyse feasibility of the proposed novel process is presented with process simulation and cost analysis.

2. Methodology

Important problems associated with synthesis from biomass are; it is widely distributed, and its composition and amount differs with location and season. Due to its large water content its transportation is costly. Also large stocks of biomass needs to be stored near the production plant because of its low energy density. Therefore, decentralized fuel production is found preferable in this project. Moreover, production in modular plants is taken to benefit from feed flexibility, scalability and faster time-to-market (Vural Gürsel et al., 2012). They are formed by pre-manufactured modules assembled into highly functional plant environment that facilitates piping, utility, control and safety requirements in its infrastructure (Hessel et al., 2012).

For ease of transportation and storage, biomass is usually densified by torrefaction or pyrolysis (Pедуzzi et al., 2013). With fast pyrolysis biomass is converted to liquid bio oil (Figueiredo and Mendes, 2012). Bio oil is up to 10 times the energy density of biomass and it has diverse chemical composition, typically with low hydrogen (7 %) and high oxygen (45 %) content (Zhang et al., 2007). Another bio source of energy is biogas. Biogas is produced by decomposition of organic matter mainly of plant and animal wastes or landfills. It is primarily methane and carbon dioxide along with other trace gases (Pagliai and Felice, 2012). For liquid fuel production, these bioresources need to be converted to syngas. The two main syngas production processes are partial oxidation and steam reforming. Conventionally steam reforming occurs in catalytic fixed bed reactors placed in direct-fired furnaces (Rostrup-Nielsen, 1984). Long contact times are required due to heat transfer limitations. In this project coupling of exothermic partial oxidation with endothermic steam reforming in a microchannel plate heat exchanger reactor is considered (Kolb et al., 2013). This reactor design provides intensive mixing and enhanced mass and heat transfer that allows very short contact times (Hessel et al., 2013).

Fischer-Tropsch (FT) synthesis from syngas irrespective of operating conditions cannot produce uniquely diesel and gasoline. Therefore, FT product upgrading is required that involves challenging separation steps of many side products. Methanol produced from syngas can either be converted to gasoline or olefins (Tabak and Yurchak, 1990). Product upgrading is simpler. There is significantly less gaseous products and light saturates and also no heavy products that require cracking. BIOGO project is formed accordingly for decentralized liquid fuel production from bioresources of biogas and bio oil through methanol synthesis in modularized small scale plants with process intensified equipment including microreactors. To analyse the feasibility of the proposed project, a preliminary techno-economic analysis is performed. First, the process scheme is developed and a mass and energy balance is done. This is carried out with Aspen PlusTM modelling. Production capacity of 117.6 t/d gasoline is selected according to the production capacity of a small scale plant (Baliban et al., 2013). In this way the viability of the proposed process scheme can be shown. Second, cost analysis is done by first estimating capital and operating costs. Accordingly cost of production of gasoline is calculated for rate of return of 10 % and 20-year plant life. This can assist to compare this route with values from FT synthesis and conventional fossil based production. Also, the economics of decentralized small scale production is assessed in comparison with large scale plant with a discounted cash flow analysis.

3. Results and Discussion

3.1 Process Simulation

The preliminary study to assess the BIOGO project is started with a process simulation done with Aspen PlusTM. Biomass handling and drying, pyrolysis, biogas cleaning, air separation and wastewater treatment are not included in the process simulation but are taken into account in cost calculations. The process flow diagram of generation of liquid fuel from biogas and bio oil is given in Figure 1. Biogas is fed together with bio oil from fast pyrolysis to the reformer in 4:1 mole ratio. High feed temperature is required to achieve a high yield of syngas and to inhibit coke formation. The reformer temperature is set to 900 °C and pressure to 45 bar to achieve high methane conversion. Coupled reforming of the two streams enables high hydrogen content of the biogas to be utilized by bio oil. The rapid and intimate heat transfer eliminates coke formation. Pure oxygen is input to the reformer to provide the heat required for the reforming reactions. Its amount is adjusted to limit formation of CO₂ instead of CO. Steam is manipulated to achieve a hydrogen to carbon monoxide (H₂/CO) ratio of two. Therefore, further reverse water-gas-shift reaction is not required. The reformer outlet gas is separated from water, carbon dioxide and methane. Water can be recycled after treatment to the reformer. The methane left after reforming is sent to a combustion unit with oxygen to recover energy. The syngas composed of H₂ and CO

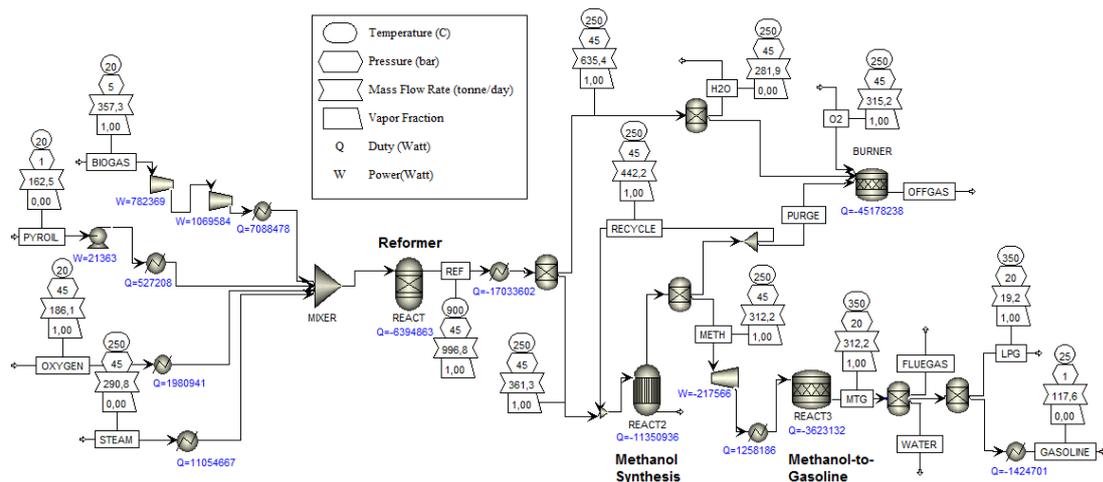


Figure 1: Process flow diagram of liquid fuel production from biogas and bio oil

is fed to methanol synthesis. This is an equilibrium reaction with two moles of H_2 and one mole of CO converted to one mole of methanol. The reactor temperature and pressure are set to $250\text{ }^\circ\text{C}$ and 45 bar respectively. Recycle is employed to maximize yield of methanol. Because of water-gas-shift reaction, some CO is converted to CO_2 which needs to be taken into consideration. The syngas mixture is typically adjusted to contain 4-8 mole % CO_2 for maximum selectivity and a stoichiometric ratio defined as $(H_2 - CO_2)/(CO + CO_2)$ of two is preferred for methanol synthesis (Iglesias Gonzalez et al., 2011). The recycle is adjusted to get this stoichiometric ratio equal to two and the combined syngas with recycle contains 6 mole % CO_2 . 10 % of the recycle syngas stream is purged to prevent accumulation. This purge stream is used as fuel gas and fed to combustion unit. The methanol product has a composition of 2.8 mole % water and 97.2 mole % methanol. It is fed to the methanol to gasoline reactor. The inlet conditions are adjusted to $350\text{ }^\circ\text{C}$ and 20 bar (Iglesias Gonzalez et al., 2011). The methanol is converted to 56 wt. % water and 44 wt. % hydrocarbons. 100 % methanol conversion is assumed. The hydrocarbon product composition is modelled based on typical composition in literature (Tabak and Yurchak, 1990). Hydrocarbon composition is 1.5 wt. % light gas, 5 wt. % C_3 , 11.5 wt. % C_4 , and 82 wt. % C_5+ . The crude hydrocarbons are separated into finished liquid fuel products of which 86 wt. % is gasoline, 14 wt. % is LPG and the balance is fuel gas. Using methanol as intermediate ensures a high conversion to liquid fuels. The light gas formation is much lower than FT synthesis. These light gas streams would need to be recompressed and recycled leading to extra energy requirement and loss of sellable product. A process route via methanol therefore avoids challenging gas separations and recycle which are required for conventional FT route. Thus, a more compact process scheme is achieved.

3.2 Mass, Carbon and Energy Balance

Mass, carbon and energy balance from simulation is presented in Table 1. The inputs to the process simulation are biogas, bio oil and oxygen while the outputs are gasoline, LPG, CO_2 and water. Although there is also water inlet to the system, the net balance of water is written on the table indicating outlet water streams are more than enough to satisfy the inlet water by recycle. 100 % balance is attained. In the

Table 1: Overall mass, carbon and energy balance

	Mass Balance t/d	Carbon Balance kmol/h	Energy Balance MW
Inputs			
Biogas	357.3	484.7	108.1
Bio oil	162.5	230.0	61.6
Oxygen	453.0	0.0	0.0
Outputs			
Gasoline	117.6	343.3	61.0
LPG	19.2	55.3	10.1
CO_2	501.5	316.1	0.0
Water	334.5	0.0	0.0

carbon balance carbon is input to the process by biogas and bio oil and exits the process via gasoline, LPG and CO₂. The carbon conversion efficiency to liquid fuels is calculated as 55.8 % which is slightly higher than reported in previous biomass to liquid studies (Baliban et al., 2013). This is mainly due to reduced separation requirement of the product in this process enhancing carbon utilization. 100 % of carbon is accounted for. Energy balance is given in Table 1 on a LHV basis. The energy efficiency of the process to liquid fuels is calculated by dividing the total energy output of gasoline and diesel by the total energy input to the process from biogas and bio oil. This efficiency is calculated as 41.8 %.

3.3 Cost Analysis

The sections of the plant include biomass pyrolysis, coupled reforming, methanol synthesis, methanol to gasoline (MTG) conversion process. Combustion unit is used to recover heat. Oxygen is produced on site with air separation unit. Water is recycled inside the unit with water treatment. Water treatment unit also includes steam turbine to generate electricity from waste heat in the process. Therefore, system is self-sufficient without external utility requirement. The direct costs of the units are calculated using estimates from several literature sources. Biomass pre-processing, fast pyrolysis and combustion unit costs are estimated using a scaling factor of 0.7 to values by (Wright et al., 2010). The other unit costs are estimated using cost parameters given in Baliban et al. (2011). Chemical engineering plant cost index is used to bring the unit costs to Q4 2013 (567.3). For conversion to euro from dollars 1.33 exchange rate is taken. The total direct cost includes cost of equipment, installation, instrumentation, piping and service facilities. The indirect costs of engineering and construction expenses and contingency are expected to be lower with modular plants since engineering and construction time and expenditure is much reduced with standardization and pre-manufacturing. Accordingly, indirect costs are estimated to be 50 % lower than conventional plants. The fixed capital investment is calculated as sum of total direct cost plus the indirect cost as given in Table 2. This is converted to annual basis to be incorporated into cost of production. Annual capital charge is calculated by multiplying the investment with a factor that depends on rate of return and plant life. For the selected rate of return of 10 % and plant life of 20 years the factor used is 0.117 (Sinnott and Towler, 2008). The other contributions to total cost of production come from raw materials, operating & maintenance (O&M) cost and LPG. LPG is sold as by-product so indicated as negative value. The cost parameters used in calculation are also given in Table 2. The total cost of production is calculated both in MM€/yr and in €/L using gasoline lower heating value of 32 MJ/L. The latter value represents the price of gasoline from which this process becomes economically competitive. The current gasoline price is 0.56 €/L (given regular gasoline spot price of 2.8 \$/gal on www.bloomberg.com/energy) is higher than 0.50 €/L estimated in this study indicating this process can be competitive with petroleum-based process. It should be noted that the estimations are sensitive to the cost parameters selected in the study. Baliban et al. (2013) found a similar value for MTG process (0.51 €/L gasoline) and a higher value for FT process (0.59 €/L gasoline equivalent) at same production capacity. The FT process cost of production is higher mainly due to higher investment cost required for this process with higher separation demand. Baliban et al. (2013) also studied higher capacities up to 50 times the capacity used in this study (5,880 t/d gasoline). The investment and fixed operating cost information of this large scale plant is taken from Baliban et al. (2013) to make comparison of decentralized plants with a centralized large scale plant. Due to seasonality large scale plants can't run at full capacity. 70 % capacity usage is assumed in this study, so one large plant of 5,880 t/d gasoline total capacity is compared with 35

Table 2: Breakdown of investment cost (left) and total cost of production (right) and cost parameters

	MM€		MM€/yr	€/L
Biomass pre-processing	3	Biomass	5	0.10
Biomass fast pyrolysis	5	Biogas	11	0.21
Combined reforming	20	O&M	3	0.07
Syngas separation	7	Capital charge	10	0.20
Methanol synthesis	5	LPG	-4	-0.07
MTG process	7	Total Cost of		
LPG - gasoline separation	3	Production	26	0.50
Combustion	13			
Air separation	9	Item	Cost parameters	
Wastewater treatment	2	Biomass	€ 62.0 per dry ton	
Total Direct Cost	75	Biogas	€ 0.12 per Nm ³	
Indirect Cost	12	LPG	€ 0.32 per litre	
Fixed Capital Investment	87			

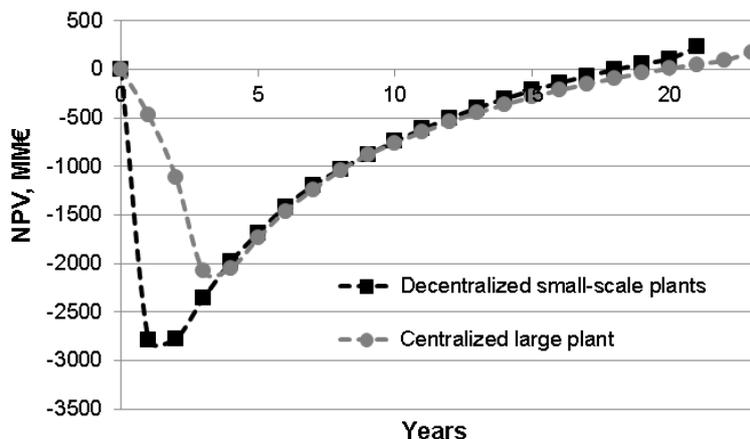


Figure 2: Net Present Value of decentralized small-scale plants and centralized large plant

plants of 117.6 t/d gasoline capacity each. Large scale plants collect biomass typically from 80 km radius so the associated transportation cost is added to the biomass cost giving 75 €/t. One year construction time is taken for small-plants and three year for the large plant. The plant-life is 20 years. The resulting net present value (NPV) calculation is given in Figure 2.

The investment cost is higher for decentralized plants because of economies of scale. However, due to lower construction time, financial gain is earned earlier and due to time value of money this gives higher NPV. Also, operating cost is lower due to no added transportation cost for biomass. It is seen that these positive effects can outweigh the negative effect of higher investment requirement.

4. Conclusion

In the proposed novel production plant process integration (reaction coupling), process simplification (elimination of complex separation steps), modular processing and process intensification (microreactor utilization) is achieved. Syngas can be generated with coupled reforming at a stoichiometric ratio suitable to directly feed to methanol synthesis while simultaneously avoiding coke and minimizing methane formation. Neither challenging gas separation nor extensive light gas recycle loops are required in contrast to Fischer-Tropsch based routes. Therefore a compact process design is attained. Carbon efficiency to liquid fuels of 56 % and energy efficiency to liquid fuels of 42 % is achieved. Cost calculation reveals that with the price parameters used this proposed novel process is economically competitive with petroleum-based process and is more economical than FT process. Capital cost reduction is possible due to lower requirement of product separation and upgrading, also with lowered indirect costs with modular processing. Transportation which has associated costs and emissions is reduced by decentralized production. Discounted cash flow analysis show that decentralized plants can be preferable to large scale plants because they can be built in shorter time with faster time-to-market. Also due to seasonality and presence of variety of bioresources feed flexibility and capacity adaptation is important and can be achieved with these plants. Liquid fuels produced from bioresources cause lower greenhouse gas emissions since carbon dioxide is captured by the feedstock and it offsets carbon dioxide emission from burning fuel. The introduction of environmental regulations such as carbon tax will improve the economics of biomass to liquid processes and can enable the shift from fossil-based processes. This paper is an ex-ante analysis before start of the BIOGO project activities and more coming insight can lead to a differentiated picture.

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