Efficient Integration of Biofuel and Chemical Production Processes with Pulp Mills and Energy Production

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Biofuel production processes FT-diesel and ethanol fermentation combined with biogas and pellet production are analysed based on electricity consumption and minimum energy consumption obtained through Pinch analysis. Finally the efficiency of the integration of various processes with pulp mills and energy production is discussed. Ethanol production combined with biogas and pellet production converts a higher percentage of the biomass energy into products than FT-diesel production. Furthermore the integration of the ethanol process needing additional heat is easier that for the FT-diesel process which produces excess heat.

1. Introduction

Biofuels and chemicals made from lignocellulostic biomass can have significantly lower carbon footprint than corresponding fossil alternatives. The efficiencies of biomass refining depend both on the material and energy efficiency of technology and the efficient process integration. Usually part of the energy present in the raw material is lost as heat or converted into byproducts which significantly effects the feasibility of the production processes as well as the electric power need. Material yields and heat efficiencies were previously discussed (Melin and Hurme 2010). Another way to assess the efficiency of biofuel production is to study loss of exergy (maximum amount of work performed when system is brough to equilibrium with the surrounding.

Integration of biofuel production using excess heat in the pulp mill has been discussed previously (McKeough and Kurkela 2005) Pinch analysis gives the minimum utility consumption needed heating or cooling utilities for all temperature intervals. It is employed previously (Parisutkul and Siemanond 2009) for natural gas facility and synthetic natural gas production (Heyne 2010).

2. Methods

Two cases where calculated to illustrate efficiency of biomass utilization and process integration. Integration with a typical Nordic European integrated Kraft pulp mill is
presented. The first case is Fischer-Tropsch -diesel production from spruce through atmospheric gasification using oxygen, low temperature FT-synthesis and hydrocracking of FT-Wax into FT-diesel and gasoline. The second case is: The production of cellulostic ethanol from spruce with biogas production from the distillation stillage and pellets from the water insoluble solids (lignin etc.).

2.1 Case 1: FT-diesel
Spruce with given composition and heating value (Melin and Hurme 2010) and 50 w-% moisture on total basis is dried to 15 w-% moisture on total basis. The biomass and enough oxygen preheated to 700°C is feed to gasifier operated at (800 °C, 1bar(a)). Tars, methane and recycle gas from FT-synthesis with additional pre-heated oxygen is feed to the thermal reformer operated at 1163 °C producing clean synthesis gas. Both gasifier and reformer gas compositions are calculated with equilibrium models. The accuracy of such model is discussed previously (Melin and Hurme 2009). The reformer outlet the gas is cooled to 520 °C and the heat is used to generating superheated high pressure steam. Cooled reformer outlet gas and 1 bar (a) superheated steam at 300 °C obtained through biomass drying is feed to the shift reactor. A ratio of 1.7 between H2/CO is obtained. The shifted synthesis gas is cooled to 40 °C and the pressure is increased to 20 bar by a 4 stage compressor with equal compression ratio and polytrophic efficiency of 80 % for each stage, cooling and liquid separation between each stage. Carbon dioxide is separated by absorption using a physical solvent such as Selexol regenerated by pressure reduction. FT reactor feed is heated to 200 °C. In FT-reactor conversion of 0.85 and chain growth probability of (\( \alpha =0.95 \)) is assumed (Prins and Ptasaki 2004). Outlet product is cooled to 40°C and condensed FT-Wax and Water is separated. The methane containing recycle gas is expanded in a turbine (efficiency 80 %) to 1 bar(a). A small purge stream avoids buildup of inerts in the system. The residual gas is heated to 700 °C and feed to the thermal reforming. The FT-Wax is hydrocracked in an oil refinery assuming reported yield of 85 w-% FT-diesel, 12 wt-% light gasoline range hydrocarbons (C5-C9) and 3 w-% gaseous hydrocarbons (C1-C4) (Calemma and Cambaro 2010). A rough estimate indicates that that enough hydrogen for the hydrocracking step is obtained by steam reforming of the gaseous hydrocarbon stream.

2.2 Case 2: Lignocellulostic ethanol, biogas and pellet production
Product yields and utilities needed were calculated based on the reported data for the described process (Wingren et. al 2007) assuming that no pellets are used to generate the needed energy. Softwood with 50 w-% moisture on total basis is feed together with SO2, LP steam and HP steam to the pre-treatment stage operated at 215 °C. Outlet product is flashed in two stages to recovered steam at 4 bar (a) and 1 bar(a) pressure. After cooling to 37 °C the mixture is feed to (SSF) Simultaneous Saccharification and Fermentation. The Water Insoluble Solid (WIS) content in the feed to SSF stage is 8,4 w-% and the distillation feed contains 3,8 w-% of ethanol. Fermented mixture containing the unhydrolysed material, biomass produced by the yeast, water and ethanol is separated by distillation. The SSF feed is split in half and LP steam is feed to the first feed stripper operated at 3 bar(a). The condenser of first feed stripper also boils the liquid in the second feed stripper operated at atmospheric pressure. The condenser in the
second column also acts as a reboiler for a third vacuum column which concentrates the distillate from the first columns. Finally the ethanol is dried with molecular sieves to 99.5 w-%. WIS containing lignin, some carbohydrates and yeast are separated by centrifugation and dried to 85 w-% dry matter content and converted into pellets. The stillage from distillation is sent to anaerobic digestion for biogas production 50 % of chemical oxygen demand (COD) is removed at the anaerobic stage and the rest of the COD is removed in the final aerobic stage. Based on the data a case was calculated where no heat or electricity is produced. Instead the biogas and solids are obtained as by products. Externally produced steam and electricity are used.

3. Results

The Grand composite curve for the FT-case is shown in Figure 1. It can be seen that there is an excess of heat in all temperature intervals especially below 230 °C and no Pinch point exists. Amount and pressure of steam produced (HP 90 bar(a) 490 °C, MP steam 12 bar(a) sat. steam and LP steam sat. 4 bar (a)) is shown. The flow of energy in biomass and utilities is shown in Figure 2. Hp steam from the process is converted with a back pressure turbine (power output/heat output ratio 0.27 to electricity and low pressure steam. Additional Electricity is produced by combustion of additional biomass using condensing turbine with a power to heat output ratio of 0.45. For the FT case only the synthesis gas compressor electricity consumption is considered. Totally around 44 % of the energy used as raw material for the synthesis and production of needed electricity ends up in the FT-diesel, light hydrocarbon fraction and purge gas.

![Grand Composite Curve for Biomass Oxygen Gasification + FT Synthesis with Reforming Loop](image)

*Figure 1: Grand Composite Curve for FT-synthesis. Temperature equals hot side temp. minimum temperature difference is 30 °C.*
In the ethanol case shown in Figure 3 there is no Pinch Point. There is heat deficit in the whole temperature interval. The Steam utilities temperature levels, and amount needed are visualized in the grand composite curve. The energy flows are illustrated in Figure 4.

**Figure 2: Energy flows of the products and utilities for FT- synthesis case.**

**Figure 3: The Grand Composite Curve for the ethanol synthesis.**
Figure 3: The heat flows illustrated for biomass and utilities.

Approximately 73 MW of additional biomass is needed shown in Figure 4 to supply the power and steam demand of the process. Due to small LP steam demand only HP and MP pressure steam is supplied to the process. Here only one high pressure level is assumed. The high pressure steam is feed through the back pressure turbine generating needed amount of MP steam with a power output/heat output ratio of 0.20 and rest of the is feed to a condensing turbine with a power output to heat output ratio of 0.45. About 65% of the energy content in the biomass and additional biomass 74 MW used for power and energy production can be found in the products ethanol, biogas and pellets.

Table 1: The heat and Power production calculated based on data by (Gullicken and Fogelholm 2000) for a typical Northern European Kraft Pulp Mill producing annually 600000 ton of air dried pulp with an annual operation of 8000 h. The plant is integrated to paper mill.

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<tr>
<td>GJ/Adt</td>
<td>18.0</td>
<td>4.2</td>
<td>9.0</td>
<td>6.5</td>
<td>4.4</td>
<td>2.3</td>
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<tr>
<td>MW</td>
<td>375.0</td>
<td>87.5</td>
<td>187.5</td>
<td>135.4</td>
<td>91.7</td>
<td>47.9</td>
<td>0.0</td>
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<tr>
<td>KWh/Adt</td>
<td>550.0</td>
<td>650.0</td>
<td>1200.0</td>
<td>225.0</td>
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<tr>
<td>MW</td>
<td>41.3</td>
<td>48.8</td>
<td>90.0</td>
<td>16.9</td>
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In Table 1 it can be concluded that normally there is no excess of LP steam in the pulp mill. About 17 MW of electricity is normally sold to outside. The amount of electricity produce could be increased if condensing turbine would be employed or a lower back pressure in the power generation. This could be done if MP and LP pressure steam produced in the biofuel process could replace steam made in the pulp mill process.
4. Conclusions

The Ethanol production combined with biogas and pellet production convert a larger amount of the energy present in the spruce to products (65 %) compared to the FT-diesel case (42 %) when the electricity needed in the process is produced by combustion of biomass. Gas compression of the synthesis gas from atmospheric pressure to syngas consumes about 18 MW of electricity. Integrated combined cycle operation and pressurized gasification would significantly decrease electricity demand. FT-diesel plant integration with a pulp mill is not optimal in the case if when produced LP-steam cannot be used. For the 600 kt/y Pulp mill and 300 MW FT plant LP steam can be used in the pulp mill when the bark boiler is not in use.

For a smaller pulp mill or a non-integrated pulp mill there might be excess of low pressure steam. The ethanol case combined with biogas and pellet production can utilize excess heat available in non-integrated mills. The integration is also easier since regardless of the size of the pulp mill and ethanol process the heat deficit can be replaced by combusting pellets to satisfy the energy balance. Utilizing secondary heat from the pulp and paper mill using and using a steam ejector to raise the pressure of MP-steam to the needed level could further increase the efficiency. In addition the purge gas in FT-case and the biogas from the ethanol process could most likely be used to substitute fossil fuel in the lime kiln.

References

Calamma V. and Gambaro C, 2010, Middle distillates from hydrocracking of FT waxes: Composition, characteristics and emission properties, Catalysis Today 149, 40 – 46