

Complementing existing CHP plants with pyrolysis and gasification to produce liquid biofuels

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Abstract

In Northern Europe there are many CHP plants operating with biomass and waste as fuel. As more wind and solar power is introduced the operating hours of these plants is reduced and thereby also the capital burden is distributed on fewer annual hours. At the same time there is a strong request to replace fossil oil by renewable alternatives for many different purposes. Here biomass and waste are the major resources available to produce liquid or gaseous bio-based products at existing CHP plants. In this study we have simulated system solutions to identify energy and material balances as well as rough economic figures. The products assumed are primarily fuels like diesel, hydrogen and methane, but also other organic compounds can be considered. Today PREEM and St1 are planning large scale production of primarily bio-diesel, or HVO, where liquid products from both pulp and paper industry and CHP plants will be suitable feedstock. The study includes a comparison between hydrogen production in gasifiers to electrolysis, and even a combination of these as oxygen from the electrolyser can be used for the gasification, to avoid ballast of nitrogen in the product gas. The study aims to identify optimal solution under different conditions with respect to both electricity and raw material costs, as well as capital cost and operating hours.

Keywords: CHP, pyrolysis, gasification, HVO, liquid bio-fuel, hydrogen.

1 Introduction

Pyrolysis has been studied as a method to convert solid biomass into liquid bio-oil. A certain percentage of the biomass is converted to bio-oil and gaseous compounds, while a residue of mainly carbon is also produced. At Brista (Stockholm Exergi) an interest has been to produce bio-char primarily (Jonsson, 2016) while the bio-oil is the primary product in e.g. Joensuu pyrolyser plant (Joensuu, 2013). This is considered one of the first large scale pyrolysis for converting biomass to bio-oil. The production is around 50 000 ton per year. At Setra in Gävle, Sweden, saw dust will be converted to bio-oil as well, with a capacity of approximately 30 000 ton per

year (Setra, 2019). The goal is to have the plant up and running at the end of 2021. Here PREEM is one of the part owners. PREEM has a plan to produce 3 million m³ bio-diesel and other bio-fuels per year by 2030 being this is a driving force for pyrolysis of biomass in Sweden. Already today PREEM is producing all their diesel with 30% HVO (hydrogenated vegetable oils) including one third tall oil and two thirds vegetable oil, mostly palm oil. At Chalmers University tests have been performed with pyrolysis (2 MW) in the G-valve of an 8 MW CFB (Circulating Fluidized Bed) boiler. Approximately 70 % of the biomass has been converted to gases, while the remaining solid fraction is passing down into the CFB bed, where the solids are combusted (Larsson et al, 2013). Most of the gases are condensed into liquids, which is an energy rich mixture of hydrocarbons. At Gobi gas a demonstration plant is operated to convert solid biomass into hydrocarbons using the FT (Fischer-Tropsch) process (Larsson et al, 2018). The product then can be used to replace fossil hydrocarbons. In Gussingen a steam-based gasification has been demonstrated in a CHP plant to produce a nitrogen free gas with high heating value (12 MJ/Nm³). This was reported in Rauch et al (2004). An alternative route can also be to utilize black liquors from pulp and paper industry, and especially the tall oil. Tall oil has been refined at Sunpine in Piteå followed by hydrogenation and distilled at PREEM oil refinery in Gothenburg. Here the oil is reacted with hydrogen to get a product equal to the fossil oil used to produce diesel and kerosene. From the previous work it can be concluded that CHP plants can be used also for production of liquid bio-fuels. In this system study we are looking for the possibility to convert solid biomass to bio-oil by integration to existing CFB-boilers at Malarenergi AB's CHP, but principally any FB-boiler could be converted in a similar way.

2 System Study

The study investigates the integration of a pyrolysis reactor and a combination with a gasifier, alternative with an electrolyser to produce H₂, hydrogen, and also O₂, oxygen, which can be used as complement to air in the combustion and gasification in a CFB boiler with the

capacity of 150 - 180 MW_{th} operating on biomass or organic waste. We then study complementing this with only a pyrolyser or a combination with also a gasifier.

The sensitivity analysis includes the fluctuations in electricity price for purchased electric power, the price for biomass and waste as well as the annuity for the capital cost. Different operational modes are then evaluated assuming different demand of heat and electricity over a year in order to investigate reasonable operating hours for production of the liquid or gaseous fuel. Here also different products are assumed like pyrolysis liquid for HVO production (hydrogenated vegetable oil), methane and hydrogen. Hydrogen can be utilized either for refinement of the pyrolysis liquid directly or extracted and sold as a product for different applications like fuel to fuel cells or similar.

The system alternatives then become:

1. CFB with pyrolyser (pyrolysis liquid as it is to a refinery).
2. CFB with gasifier (for hydrogen production primarily).
3. CFB with pyrolyser combined with gasifier and gas upgrading with separation system.
4. CFB with pyrolyser combined with electrolyser.
5. CFB with pyrolyser combined with gasifier and gas upgrading with separation system plus electrolyser.

The system design with dimensioning of different equipment to balance the 180 MW thermal capacity of the CFB boiler is carried out. Thereafter a rough cost estimate is made for the investment cost and maintenance cost for the different solutions. The annual costs for each system design is estimated. Concerning dual bed gasifiers Lundberg et al (2018) have presented conversion measured in a pilot plant and discussed scale up of this.

The assumptions for some different possible scenarios are based on heat demand for previous years. Year 2016 when the temperature dropped to -20°C for several months, 2010 which was a harsh winter generally with much snow and 2019 when temperature has been quite high most of the time. On the other end we have summer 2018 when the temperature was 30°C and higher for two months. For “extreme periods” with very high or low temperature we look at hourly values, while normally monthly averages. From this we calculate energy demand with respect to heat, cooling and electricity and make a diagram showing the demand as a function of hours for each year.

We also look for the price of electricity, heat and cooling over the year for these years but also simulate other prices that might be expected in the future during different time periods. Here we look for how much of the total electricity production that comes from wind and solar power today and from this try to predict future

probable price. When there is a lot of wind power we can expect low electricity price and then electrolyzes may be the best alternative for hydrogen production, and summertime solar power will be in surplus

The calculations are plotted in a diagram where we can compare cost of product as a function of hours per year, fuel cost and electricity cost. The products assumed are H₂ and HVO.

3 Simulations

The model is primarily energy and mass balances for the different equipment complemented with chemical reactions for combustion, gasification and pyrolysis as well as electrolysis.

$$\frac{\partial m_i}{\partial t} = (\dot{m}_{in,i} - \dot{m}_{out,i}) \tag{1}$$

$$\frac{\partial m_i}{\partial t} = k(T) * m_{0,i} \tag{2}$$

$$\sum_i^n m_{i,in} x_{i,in} = \sum_i^k m_{i,out} x_{i,out} \tag{3}$$

$$\frac{\partial Q_i}{\partial t} = (\dot{m}_{in,i} - \dot{m}_{out,i}) * C_{p,i} * \Delta T + \Delta H * \frac{\partial m_i}{\partial t} \tag{4}$$

Here $\dot{m}_{in,i}$ is flow in (kg/s) of component i and $\dot{m}_{out,i}$ is flow out. $k(T)$ is a reactivity constant for a given reaction, Q_i is the energy in kWh or kJ. $C_{p,i}$ is the heat capacity and ΔT is the temperature difference. ΔH is the heat released (or taken up) during the reaction.

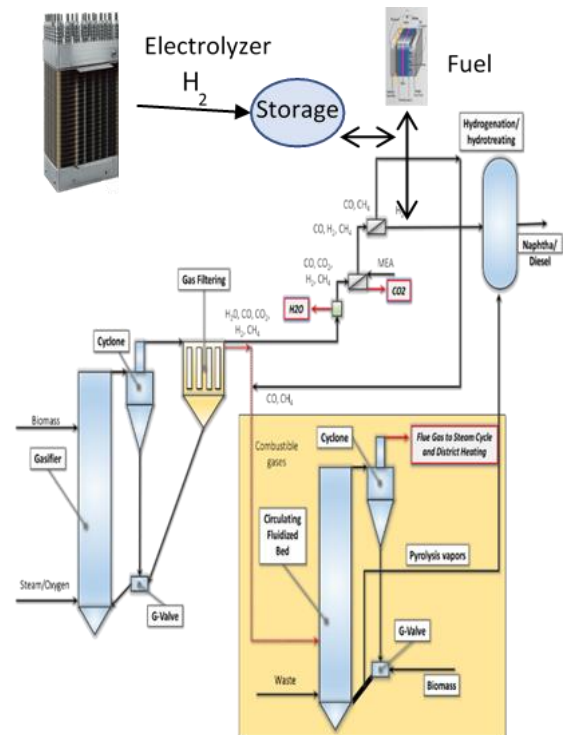


Figure 1. System with CFB boiler, pyrolyser, gasifier and electrolyser.

The fluidization of sand was determined from the balance between gas flow and buoyancy forces versus gravity forces. For full combustion this forms only CO₂

and H₂O. For the gasification we get different composition depending on the relative oxidation (how much O₂ is added in comparison to what is needed for 100% combustion), capacity (ton DS/m²h) and temperature. The gas composition for two different cases are given in Table 4 (Dahlquist et al, 2018). The energy balance in the CFB boiler is given in Table 1 and for the pyrolysis in the G-valve in Table 2. The boiler considered is recycled wood CFB boiler at Malarenergi with approximately 150 MW fuel feed. Part of this is used to heat sand which is passing over to the G-valve to heat fuel injected also there. This heat together with partial combustion is used as input to pyrolyse bio-mass and waste.

Table 1. Energy balance for the CFB boiler

Alternative	1
Fuel to CFB boiler	
kgTS/s	7.7
MW HHV (21MJ/kg)	161.7
MW LHV (19 MJ/kg)	146.3
Fuel to G-valve- pyrolyzer	
kgTS/s	7.3
MW HHV	152.5
MW LHV	138
Air boiler	
m ³ /s	27.3
kg/s (1 m ³ = 1,293 kg)	35.3
MW to heat air to 850 oC	29.3
Air pyrolyzer	
m ³ /s	3
kg/s	3.9
MW to heat air to 450 oC	1.7
Sand to cyclone	
kg/s	54.6
temp boiler	850
temp after pyro	450
MW to pyrolyzer from sand	17.9

Table 2. Energy balance for the pyrolyser.

Pyrolyzer	
MW from combustion in pyrolyzer	
MW to heat fuel to 450 oC	2.6
Losses	
%	11.5
MW (incl drive pyrolysis)	17.5
MW to drive pyrolysis 1123 kJ/kg	8.2
Pyrolyzate liq	
%	64.6
MW	98.5
kg/s	4.7
kg/h	16884
Gas from pyrolysis	
%	7.1
MW	10.8
Solid residue to CFB boiler from G-valve	
%	16.8
MW	25.6
Sum MW from pyrolyzer to boiler	36.4

3.1 Pyrolyser

Pyrolysis demands 8.2 MW while the heat from sand from the cyclone is 17.9 MW. Total losses are estimated to be 17.5 MW, assuming same proportion as in the small pilot plant. In reality losses can be reduced in a larger plant. Heat in gas and solids that are fed back to the CFB boiler are 36.4 MW solid organic residues and 10.8 MW gaseous compounds. The heating value in the pyrolysis liquid is 98.5 MW which correspond to 16.9 ton/h. Pyrolysis balances have been given in e.g. Neves et al (2015), where different conditions have been investigated. In our example we have used the balance presented by Atsonios et al. (2015) which is quite typical.

1 kg bio-oil may contain 46.7 mol C, 62 mol H₂ and 23.6 mol O. To remove 23.6 mol O we then need 23.6 mol H₂. 4.69 kg liquid/s means 23.6 mol O/kg*4.69 kg/s = 110.7 mol/s.

3.2 Gasifier

We have used experimental data from our pilot gasifier using wood pellets with 1.2 ton DS/m², h at 42 % relative oxidation (EOR) and steam addition. The gasification temperature was 670°C. The gas composition was 8.4% H₂, 10.9 % CO, 2.4% CH₄, 17.8 %CO₂, 14.9 % H₂O and 45.7% N₂. In reality the water content is quite uncertain as an unknown amount was condensing in the sampling system. We have made two assumptions. In the first case we just have used air as oxidation media and not treated the gas before the membrane separation. In the second case we have used oxygen instead of air and first condensed out water to 90% and then removed CO₂ to 90% using MEA. In the first case we can assume some H₂O and some CO₂ may be passed over with the H₂ through the membrane, while

in the second case the gas will be almost 100% H₂. In Table 3 we can see how the gas composition is changing over the different steps. In column 1 we have the raw gas after the gasifier. Then we have membrane filtrate where we have assumed 97% of the H₂ as clean gas in column 2 and residual gas going back to the CFB boiler in column 3. In column 4 we have the “raw gas” after condensing out water to 90% and removed CO₂ to 90% in a MEA scrubber or membrane filter with MEA at the back side. In column 5 we then have 100% H₂ and in column 6 the composition of the residual reject gas.

Table 3. The gas composition from raw gas to clean H₂ respectively residual gas for the two alternatives – gas separation in membrane filter directly respectively after separation of H₂O, CO₂ and not using air but oxygen from hydrolyzer.

	Membr separation		No N2 and remove H2O + CO2 to 90%				
	Gas from direct 97% gasifier	Filtrate	Reject	Feed gas to membr	Membr separatic Raw gas		
H2%	8.4	100	0.27	33.5	100	1.5	20.5
CO%	10.9		11.8	43.6		64.5	26.6
CH4%	2.4		2.6	9.6		14.3	5.9
CO2%	17.8		19.3	6.9		10.3	43.4
H2O%	14.9		16.3	6.4		9.4	3.7
N2%	45.6		49.7	0		0	0
	100		100	100		100	100

Calculations are now made for two different cases. In the first case it is actual gas composition from our pilot gasifier where we operated at 670 °C, 42 % relative oxidation and a capacity of 1.2 ton DS/m².h. The moisture content was 30% by addition of steam to wood pellets (Table 4 case 1). The second case is using a regression model from many gasification experiments in the pilot plant and assuming 35% relative oxidation, 2 ton DS/m².h, 800 °C, 30 % moist (Table 4 case 2).

Table 4. Gas composition and mol/kg DS for the different components for two different raw gas compositions.

	Case 1		Case 2			
	Raw gas without N2 ; water condensed out		Raw gas		Raw gas without N2 ; water condensed out	
	%	mol/kg DS	%	%	%	mol/kg DS
H2%	20.5	17.7	20.2	40.7	40.7	41.6
CO%	26.6	14.9	12	24.2	24.2	18.2
CH4%	5.9	3.3	4.3	8.7	8.7	6.5
CO2%	43.3	24.4	11.8	23.8	23.8	17.8
H2O%	3.7	6.4	13.6	2.7	2.7	2.8
N2%	0	0	38.1	0	0	0
tot	100	66.7	100	100	100	86.9
H2+CO+CH4		39.2				72.8

The balances for the pyrolyser is taken from Atsonios et al (2015). We have noticed that for 4.67 kg pyrolyzate/s we need to remove 110.7 mol O/s. For the first gas composition we produce 17.7 mol H₂/kgDS. (110.7 mol H₂/s)/ (17.7 mol H₂/kgDS) = 6.25 kg DS/s. If we can convert CO and CH₄ to H₂ as well we get 17.7 + 14.9 + 2*3, 3 = 39.2 mol H₂/kg DS. This would mean that 2.8 kg DS/s would be needed to the gasifier to cover the

reduction of Oxygen in the pyrolyzate. For the second gas composition (case 2) we get 41.6 mol H₂/kg DS which gives a demand of 110.7/41.6 = 2.7 kg DS/s. If we convert also CO and CH₄ to H₂ through reaction with steam, we would get 72.8 mol H₂/kg DS and a demand of 110.7/72.8 = 1.52 kg DS/s. From this we can see that how the system is designed and how the gasifier is operated will have a significant effect on the capacity of the gasifier. With the second case the size would be 43 % compared to the first case for only H₂ utilized and 54 % if also CO and CH₄ is converted to H₂.

If we have a HHV of the fuel at 21 MJ/kg we would need a gasifier with 1.52*21= 31.9 MW while in the case 6.25 kg DS/s*21 MJ/kg the capacity would be 131 MW. Cost estimates for the gasification and enrichment of H₂ has been presented in Naqvi et al (2017). We use the figure 70 M€ for a 180 MW gasifier plant and use the scaling factor 0.8, which gives 23.5 M€ for case 1 and 17.6 M€ for case 2. If we look at alternative 5 where we combine gasifier with electrolyser we get 13.5 respectively 10.1 M€ for the gasifiers but have to add the cost for electrolyser as well.

3.3 Electrolyser

Electrolysers use electricity to split water into H₂ and O₂. If we have a demand of 110.7 mol H₂/s it means that we also produce 55.4 mol O₂ (110.7 mol O). This can be used in the combustion, but especially in the pyrolyser and the gasifier if we combine with also this.

For the electrolyser we can assume the price to be some 500 €/kW at large scale up to the double for small scale electrolysers. Typically, one kg H₂ demand 58 kWhel for production, which means 0.017 kg H₂/h with one kW input electricity. For the demand 0.22 kg H₂/s then means a capacity of 0.221 kg H₂/s/(0.0000047 kg H₂/s)= 46 800 kW. For alternative 5 a 23 400 kW electrolyser is needed. We assume 0.12 as annuity (4%, 10y) and cost for electricity in the range 1-10 €cent/kWh_{el}. The prize of electricity will be dominating. When there is a surplus of electricity production we will be in the lower range of the span, while when there is a deficiency the prize will be in the upper range. This makes it more complicated to decide best technology. It would be good to have both gasifier and electrolyser and use the bio-gasifier when electricity prize is high, and electrolyser when it is low. If we assume a cost of 500 \$/kW it means 446 €/kW in investment cost. This would give an investment cost of 20.9 M€ for the 46 800 kW unit and 10.5 M€ for the 23 400 kW unit.

4 Results and discussion

In Tables 5-7 we see the capacities for the different alternatives. We have assumed a biomass fuel price of 14 €/MWh, which is average today. For electricity we have assumed 10 €/MWh and 35 €/MWh, as we expect quite strong variation over the year, depending on the

balance between production and consumption. We also have looked at 5000 hours and 7000 hours of operations per year. All equipment is assumed having 20 years life time and 5% interest rate, giving annuity 0.081. Concerning the gasification and electrolyzes the figures are reasonable, while for the pyrolysis it is more of a “guestimate”. The cost figures are taken from Naqvi et al (2017). There already are vessels for pyrolysis at the CFB boilers, the G-valves, but addition is needed with fuel feeder, gas outtake and gas condensation. As the reactor for H₂ with pyrolysis liquid is principally same for all alternatives except for alternative 1, it has not been included. We have assumed the value of the pyrolysis liquid to be 70% of the refined (reacted with H₂) for case 1. For case 2 we have just assumed same value for H₂ as for pyrolysis liquid. For case 2 and 3 we have added a cost for membrane separation for H₂ enrichment of 10 M€, and for case 5 half of this. These figures are taken from Naqvi et al (2017).

Table 5. 5000 hours per year, biofuel cost 14 €/MWh, electricity 10 €/MWh, annuity 0.081.

5000 h/year	Case				
Total cost incl capital/y	1	2	3	4	5
Capital cost(0.081) M€	3.2	2.7	6	4.9	5.5
Fuel 5000 h/y 14 €/MWh	13.9	12.3	19.4	17.2	12.3
Cost el 10€/MWh				2.3	1.2
Total cost per year	17.1	15	25.4	24.4	18.9
Income pyroliq	17.2	24.6	24.6	24.6	24.6
Gross benefit	0.12	9.7	-0.7	0.2	5.7

Table 6. 7000 hours per year, biofuel cost 14 €/MWh, electricity 10 €/MWh, annuity 0.081.

7000 h/y el 10 €/MWh	Case				
Total cost incl capital/y	1	2	3	4	5
Capital cost(0.081) M€	3.2	2.7	6	4.9	5.5
Fuel 7000 h/y 14 €/MWh	19.4	19.4	19.4	19.4	17.2
Cost el 10€/MWh				3.3	1.6
Total cost per year	22.6	22.1	25.4	27.6	24.3
Income pyroliq	24.1	34.5	34.5	34.5	34.5
Gross benefit	1.5	12.4	9.1	6.9	10.2

Table 7. 7000 hours per year, biofuel cost 14 €/MWh, electricity 35 €/MWh, annuity 0.081

7000 h/y, el 35€/MWh	Case				
Total cost incl capital/y	1	2	3	4	5
Capital cost(0.081) M€	3.2	2.7	6	4.9	5.5
Fuel 7000 h/y 14 €/MWh	19.4	19.4	19.4	19.4	17.2
Cost el 35€/MWh				11.6	5.8
Total cost per year	22.6	22.1	25.4	35.9	28.4
Income pyroliq	24.1	34.5	34.5	34.5	34.5
Gross benefit	1.5	12.4	9.1	-1.4	6.1

Table 8. 7000 hours per year, biofuel cost 14 €/MWh, electricity 100 €/MWh, annuity 0.081

7000 h/y, el 100 €/MWh	Case				
Total cost incl capital/y	1	2	3	4	5
Capital cost(0.081) M€	3.2	2.7	6	4.9	5.5
Fuel 7000 h/y 14 €/MWh	19.4	19.4	19.4	19.4	17.2
Cost el 35€/MWh				33	16.5
Total cost per year	22.6	22.1	25.4	57.3	39.1
Income pyroliq	24.1	34.5	34.5	34.5	34.5
Gross benefit	1.5	12.4	9.1	-22.9	-4.7

From the results we can see that for 5000 hours operations the alternative with a smaller gasifier and a

smaller electrolyser looks like a good alternative. The electrolyser is then used to produce both H₂ and O₂. The O₂ is used in the gasifier, and thereby giving a better gas from the gasifier, without N₂ from the air. For 7000 hours most cases are economic at low electricity price, but especially alternative 4 becomes very unprofitable with the higher electricity price. As we don't have very accurate figures for the value of pyrolysis liquid of different qualities, and neither for large amounts of H₂, the results are more giving relative impact of different alternatives, but clearly show the importance of cost for both biomass and electricity. If we could use waste instead of biomass, we would have an income of 12 €/MWh instead of a cost for the better biomass of 14 €/MWh, but instead there would be an issue of what quality the pyrolysis liquid would have, if there is e.g. halogens from PVC in the plastic fraction.

From the analysis we can see that the span with respect to economy is high for the different alternatives. This is due to high difference in prize for different fuels both right now, but certainly also in the future. The capital cost also has gone down a lot last years for electrolysers and probably will proceed going down if the technology will be used much more frequently than in the past. We also can see that the sizing of especially the gasification plant will depend a lot on how much oxygen there will be in the liquid product after the pyrolysis. From literature we can see that the amount of oxygen varies between some 20 to 50 wt. %. If we can utilize both H₂ and CO for reduction of O in the liquid product it will also make a large difference compared to if only H₂ is used. Still, we have reliable figures on many process parts verified in both own experiments on gasification of different biomass and black liquors as well as reports from literature from experiment in pilot and demonstration plants, showing that each part of the processes are possible to get to work, although the complete systems have not been implemented yet.

5 Conclusions

The study shows that different system solutions can be the most economic depending on the conditions. If the price for pyrolysis liquid is good even without hydrogenating it, case 1 may be interesting. If there is a strong demand for H₂ as such, case 2 is feasible. If the value of the pyrolysis liquid is much higher after hydrogenation the addition of the gasifier make sense as in case 3. If the electricity price is low case 4 is good. If the electricity price is varying case 5 make sense, and also it is positive that the O₂ produced in the electrolyser can be utilized in the gasification. Thereby we get a N₂ free gas which is much easier to handle than gas with high amount of N₂. The cost figures are of varying quality and thus next step will be to do more detailed cost estimates and more detailed design. Here we will

utilize the simulation model we have developed to give the possibility to do more sensitivity analysis.

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