Towards large, fossil free mills with integrated biorefineries – trends in modern pulp mills

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TOWARDS LARGE, FOSSIL FREE MILLS WITH INTEGRATED BIOREFINERIES – TRENDS IN MODERN PULP MILLS

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ABSTRACT

Global economics require ever larger pulp mills with increased energy efficiency. Paris agreement means pulp and paper mills need to significantly reduce fossil fuel based carbon dioxide emissions. World markets are turning towards biobased products. This creates possibilities to manufacture new sellable products. At modern pulp mills, renewable heat and electricity are produced in excess of the own process requirement. Biogas and solid biofuels can be produced at the mill by processing woody biomass residue. These biofuels can then be combusted e.g. in the lime kiln to replace fossil fuels. In Northern regions, crude tall oil and crude turpentine have been produced and refined for sale. Newest mill integrate biorefineries to further increase returns from smaller revenue streams. In this study, benefits and constraints regarding selectable future mill designs are discussed.

Keywords: Kraft pulp mill, Lime kiln, Biorefinery, Electricity

INTRODUCTION

Kraft pulping is more than 125 years old. Nevertheless, changes are still occurring at breathtaking place efficient (Tran and Vakkilainen 2007; Vakkilainen *et al.* 2014). In the traditional kraft process, about half of the wood is dissolved, and together with the spent pulping chemicals, forms a liquid stream called weak black liquor (Vakkilainen 2005). The weak black liquor is separated from the pulp by washing, and is sent to the kraft recovery system, where the inorganic pulping chemicals are recovered for reuse, while the dissolved organics are used as a fuel to make steam and power. The high strength of kraft pulp, the ability of the process to handle almost all wood species, the favorable economics due to high chemical recovery efficiency (about 97%), the excess electricity and heat production (Vakkilainen *et al.* 2008) give the kraft process an advantage over many other pulping processes.

The environmental pressures force mills to continue to reduce emissions to air and water and look at more sustainable management of biomass resources. In modern kraft pulp mills, most of the energy is biomass-based. Fossil fuels are used in lime kilns and to minor extent during upsets, start-up, and shutdown. Recent Paris 2015 Agreement has forced the industry to search new ways to reduce its already low CO2 footprint. The agreed on targets require the pulp industry to have replaced fossil fuel based carbon dioxide emissions by 2050. This means changes in fuel usage.

The economic trends require pulp mills to be larger and more energy. This is clearly shown by e.g. several just started pulp mills in Brazil that are over 1 000 000 ADt/d. In the recent past chemical pulping capacity first increased in Indonesia and then started growing in Brazil. Lately China has added significant manufacturing capacity. In traditional pulp producing countries, like Finland and Sweden the production has stagnated. In North America, the production has even declined steeply, Figure 1. Of the 130 million tons/year of chemical wood pulp that is produced globally, hardwood pulp starts to be as frequently produced as softwood (FAO 2016).

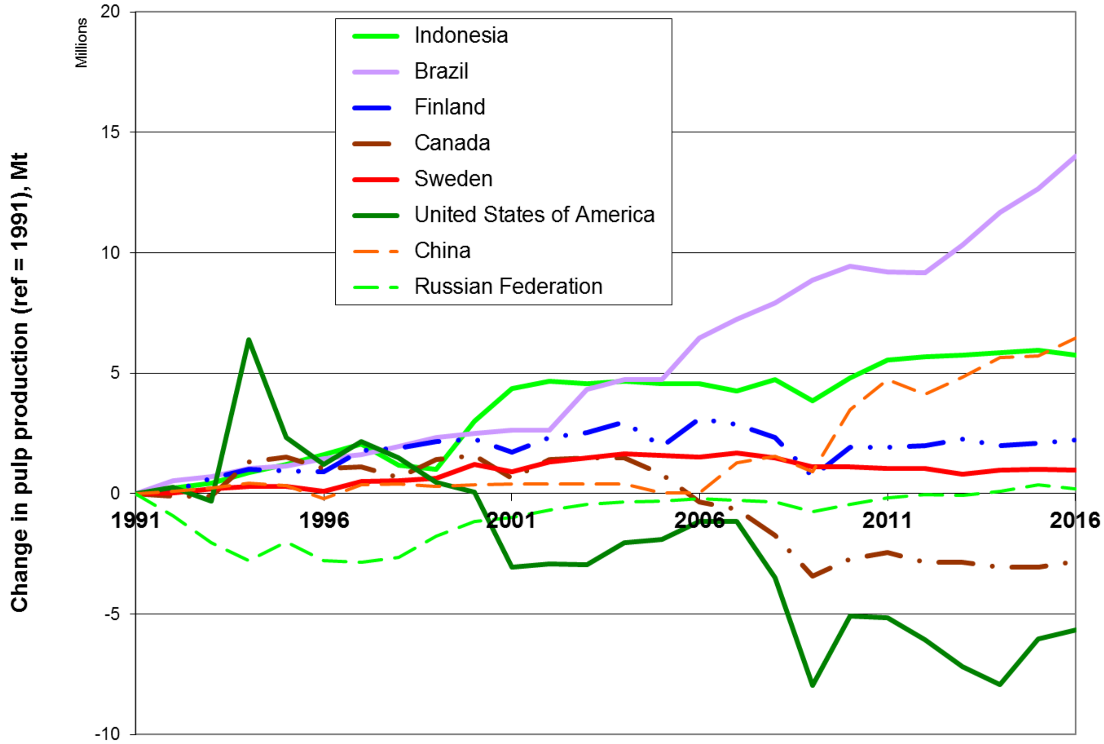

Figure 1: Change in wood chemical pulp production in selected countries, data from FAO 2016.

The competitive pressure is making the industry reinvent itself from production of single commodity to offering multiple renewable bioproducts. The main challenge is to change the way P&P companies are operating. Rather than to try to concentrate on maximizing the production of single product at single line, the companies must reinvent themselves as networked entities co-operating with several added-value products.

The economic attractiveness of additional products comes from several directions; Firstly one can manufacture transportation fuels to replace fossil fuel ones. The success of these depends on the future requirements of biofuels replacing fossil fuels used by cars and especially on political will to change; subsidies. Secondly, one can manufacture biofuels like white of black pellets to be used in traditional production of electricity and heat. This field is opening up and even if several companies are playing with it, the attractiveness depends of the future development of the fuel markets. The most active one is the production of new biomaterials. There is an increasing need to find new sustainable raw materials and products. New packaging material, fibers for clothes, materials to be used in automotive industry and additives for pharmaceuticals are already taking off.

STUDIED REFERENCE MILL

To clarify some of the suggested changes mass and energy balances have been calculated to a reference mill. The studied mill produces 1.5 MADt of hardwood pulp in 350 annual operating days. The wood handling receives 24 370 m^3sob/d of eucalyptus logs. It produces chips to the digester and woody residue like bark, fines from screening and other biobased woody wastes. The reference mill was modeled using the Millflow spreadsheet. Millflow includes detailed mill mass and energy balances and is introduced in more detail in previous work (Kuparinen and Vakkilainen 2017, Hamaguchi *et al.* 2013). The main operating values for the reference mill are shown in Table 1.

Table 1. Main process values for the reference mill.

	Unit	Base case mill
Production		
-Operating hours	h/a	8400
-Bleached pulp production	ADt/d	4560
Wood handling		
-Wood income	m^3sob/d	24 370
-Residue generated	BDt/d	567
-Wood moisture	%	50
Recovery boiler		
-Solids as fired	BDt/d	7266
-Net steam flow	t/h	1021
Power boiler		
-Woody biomass fuel use	BDt/d	567
-Net steam flow	t/h	116
Lime kiln		
-Product	t/d	1128
-Heat requirement	MW	72
-Oil consumption	t/d	143
Energy		
-Steam use in pulp mill	t/h	860
-Power generation	MW	195
-Power consumption in pulp mill	kWh/ADt	609

Steam is generated in the recovery boiler and to lesser extent in a separate power boiler. This power boiler is fired with woody residues from the wood handling process.

Steam is used in steam turbines for electricity generation and for heating purposes in the pulp production processes. There is abundantly low value heat available in the form of steam and water flows. On demand, these flows can be utilized for heating purposes, such as for biomass drying. Electricity generation exceeds the mill power demand. All available biomass residue is combusted in the power boiler for additional steam generation.

The CaO requirement is 918 t/d. This translates to the kiln production of 1128 t/d because of unreacted lime, losses and make-up lime of 27 kg/ADt. The lime kiln heat demand is 72 MW. 143 t/d oil is used to fire the lime kiln, and it represents approximately 90% of the kiln heat demand. The rest is supplied from mill by-products with good calorific value.

In modern pulp mills in South America, the eucalyptus logs are usually debarked at the forest. As a consequence, the amount of biomass waste produced is usually lower than in softwood based Scandinavian pulp mills. The power boiler is thus much smaller and installed only to incinerate the residual bark, sizing residue and biomass side fractions from the woodhandling area.

FOSSIL FUEL FREE OPERATION

Rising oil and natural gas prices have encouraged finding alternative fuels to be used in the lime kiln (Manning and Tran 2015). Methanol, turpentine and hydrogen have been successfully fired in lime kilns. Firing biomass as pulverized or as biogas from gasification has started to gain acceptance (Kuparinen and Vakkilainen 2017). When one changes the fuel in a lime kiln, it affects the whole causticization and might have an effect on pulp quality. Without proper attention fluctuations in fuel flow, heating value and moisture content can create problems as the temperature profile of the lime kiln changes causing ringing. The moisture and oxygen content of biomass-based fuels is typically higher and adiabatic flame temperature is lower than those of the fossil fuels, Table 2.

Table 2.Comparison of lime kiln fuel properties.

	Lower heating value (MJ/kg)	Adiabatic flame temperature (°C)	Contaminants
Heavy fuel oil	40.6	2210	No
Natural gas	50.0	2050	No
Biogas, hot	5.5	1870	Some[1]
Pulverized wood	14.3	1950	Significant[2]

[1] Depending on the local biomass available
[2] Bark as pulverized material should be avoided

Use of the biobased fuels in lime kilns are more thoroughly discussed in (Kuparinen and Vakkilainen 2017, Manning and Tran 2015). The use of biomass-based fuels lowers the firing end temperature in the kiln, which leads to requirement for higher firing rates in order to maintain the production capacity at the same level. This is one reason why the full replacement in kilns designed to operate with oil or natural gas becomes challenging. For these existing lime kilns, some vendors recommend reduction of the replacement rate to e.g. 80% in order to keep the kiln capacity at the same level. Appropriately, sized kiln can be built when a new pulp mill is constructed with a target of burning only bio-based fuel in the kiln. Several new pulp mills have chosen to install biomass gasifiers for their lime kilns, Figure 2.

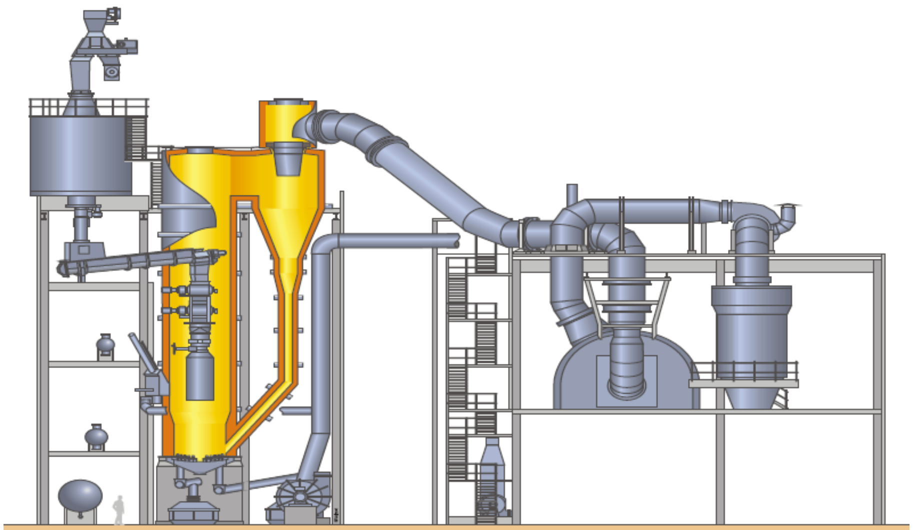

Figure 2: Bark gasifier for Lime Kiln (Courtesy of Valmet)

Biomass gasification is a well-known process, and it has been used before in pulp mills to fire lime kilns, for example in Finland in the beginning of the 1980s' (Isaksson *et al*. 2008). Recently, it has gained interest again due to increasing demand on fossil fuel replacement and utilization of biomass residue. Circulating fluidized bed (CFB) gasifiers are typically suitable for pulp mill integrations due to their size and ability to gasify biomass residue of varying quality. For a CFB gasifier, biomass moisture content should be at or below 15% and average particle size at maximum 6 mm. Electricity use in the pulp mill increases somewhat because of gasifier integration due to electricity consumption in biomass preprocessing and gasifier internal use. The main part of power used by the gasifier is due to air fans. The gasifier needs to be located as close to the kiln burner as possible in order to minimize the cost with refractory ducts, which are needed to *e.g.* reduce the risks related to erosion and corrosion. Producer gas from gasifier needs to be burned hot as cooling it would unnecessarily increase the consumption of said gas.

The costs of the gasifier depends on the planned process and the needed equipment. The costs increase when for instance storage facilities or backup system for unplanned biomass delivery breaks is desired. Three scenarios were estimated. These are neutral, optimistic, and pessimistic scenario. In the scenarios, the values for investment cost, interest rate, fuel oil and biomass prize were varied to estimate the effect of uncertainties in these factors on the feasibility of the concepts. The initial values used in the calculations are presented in Table 3.

Table 3. Initial values for evaluation of economic feasibility.

	Unit	**Gasification**		
Scenario		Neutral	Optimistic	Pessimistic
Biomass needed	BDt/d	283	283	283
Electricity price	USD/MWh	35	25	45
Interest rate	%	10	8	12
Investment	MUSD	42	34	50
Biomass price	USD/t	6.2	3.2	12.5
Make-up lime price	USD/t	180	180	180

The capital and operational costs for the studied cases were estimated based on recent investments in Scandinavia and vendor data. The values were adjusted due to higher construction costs, import taxes and other tributes in Brazil. Investment cost is based on equipment capable of replacing 100% of the fossil fuel during normal operation in the lime kiln. For the studied case, the biogas will cover 74.8% of the kiln heat demand without additional biomass brought in. The main parameters from the mill calculations are presented and compared with the base case in Table 4. Other operational information presented in Table 1 remain unchanged.

Table 4. Effect of integration of lime kiln fuel production on the reference mill process.

	Unit	**Base case**	**Gasification case**
Power boiler			
-Waste from woodhandling	BDt/d	567	283
-Net steam flow	t/h	115.6	58.4
Lime kiln			
-Product	t/d	1128	1128
-Heat requirement	MW	72	72
-Make-up lime use	kg/ADt	27	29
-Oil consumption	t/d	143	-
-Biogas consumption	t/d	-	813
-Biofuel share of kiln energy	-	-	74.8%
Biofuel production			
-Power consumption	MW_e	-	4.8
-Energy for drying	MW_{th}	-	11.5
Energy			
-Power generation	MW	195	180
-Power to the grid	MW	41	23

Gasification of woody material to be burned in lime kiln does not affect the actual pulp production nor the pulp quality as long as we can keep the white liquor quality intact. A major effect is the increased amount of non-process elements in the lime cycle, especially phosphorous, which leads to increase in the use of make-up lime. In a modern, energy-efficient mill, it is possible to dry the major part of lime kiln biomass fuel by utilizing existing side streams and secondary heat.

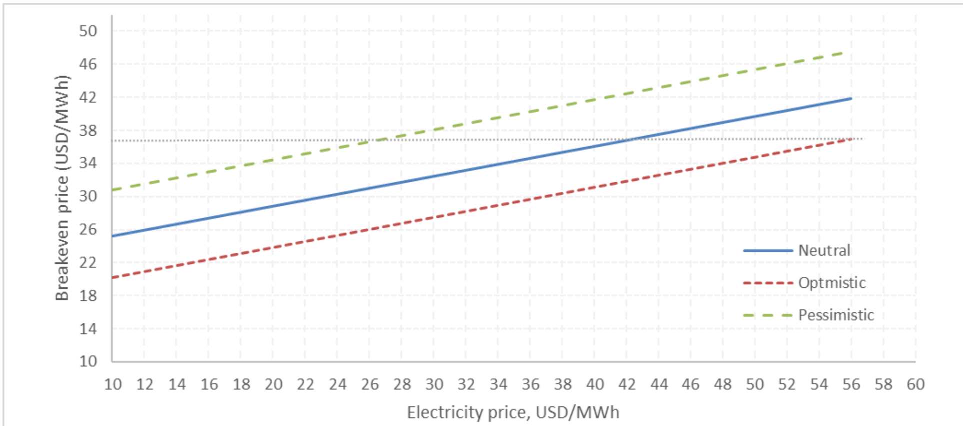

Figure 3: Breakeven lime kiln fuel price for gasification case in neutral, optimistic, and pessimistic scenarios as a function of electricity sale price

The breakeven lime kiln fuel price as a function of electricity sale price for the gasification is represented in Figure 3. A case can be considered profitable when the price of replaced fuel exceeds the calculated breakeven price. Gasification can be profitable, except for the pessimistic scenario (dashed horizontal line in Figure 3). However, the payback time gets more attractive as the price of possible electricity sales goes down. It is also important to point out that the oil price varies much depending on the mill location. In the future, fossil fuel-free solutions are needed in every mill, and then the decisions will be made based on mill-specific details and considering the political situation, namely, which solutions are encouraged and possibly subsidized based on political decisions.

In addition to lime kiln use, fossil fuels are sometimes used in a pulp mill to increase safety of NCG burning as well as during boiler and kiln upsets, start-up, and shutdown. Substituting renewables for fossil lime kiln fuels will remove most of the fossil fuel use and therefore make the mill more than 90% fossil fuel-free. In order to run a pulp mill entirely without fossil fuels, a renewable option for the auxiliary fuels is required. Pyrolysis oil or ethanol, for instance, could be used for this purpose, but the implementation needs further studies.

PLATFORM TO SUSTAINABLE BIOINDUSTRY

The demand and cost of energy is increasing rapidly while climate change appears to be progressing at an unacceptable pace. Increased use of bioenergy and biofuels can reduce greenhouse gas increases. Pulp and paper mills are logical sites for increased biomass use. They have access to biomass feedstock, and a possibility to utilize process residues. Pulp and paper mills also have readily the infrastructure such as steam, chemical handling, effluent treatment, oxygen production and other utilities and logistics needed.

The modern trend is to try to add additional process to kraft process to gain more revenue. Processes installed commercially include e.g. lignin removal, biogas production and biomaterials production (Metsä Fibre 2014; Weymarn 2015). Possible additional processes could include biomass torrefaction and bio-oil production, Figure 4.

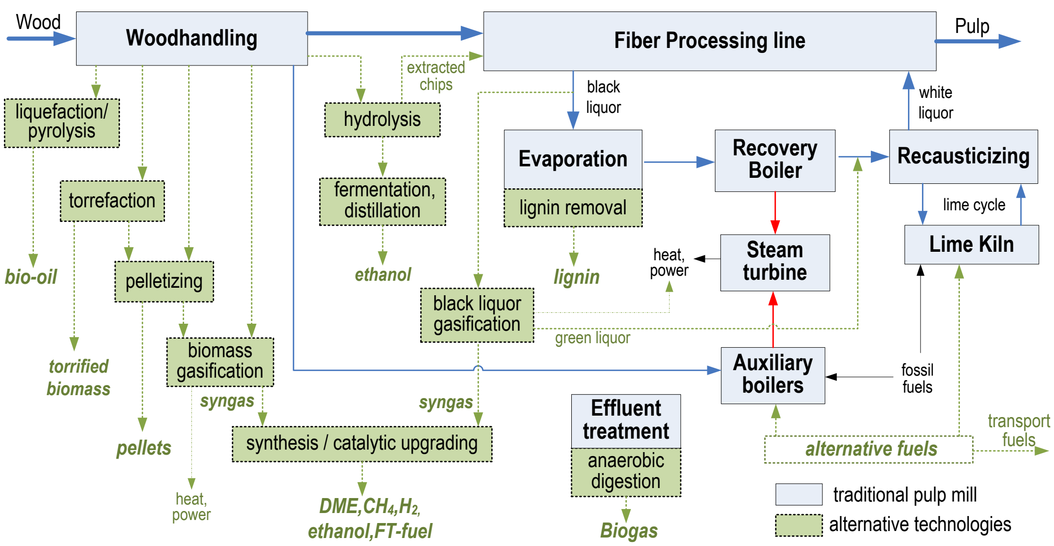

Figure 4: Some possible biorefinery options in a kraft mill (Hamaguchi *et al.* 2012)

Production of additional solid fuels

The pulp mill utilizes large amounts of wood. It therefore generates large amounts of biofuels; black liquor, bark and other side streams. As biofuels like pellets have become tradeable goods (Proskurina *et al.* 2017) it would seem logical that also pellet production would be included in modern mills. However short transport distance of the logs is one of the mainstays of mill economics. If more biomass is required, then the transport distance grows.

Pyrolysis and torrefaction are known, newly commercial processes that can be integrated to pulp mill. Both could be used to produce fuels for sale of for mill internal use. New inroads have been opened also in producing sellable bioproducs from pulp mill sidestreams like biosludge (Alatalo *et al.* 2013).

Integrated lignin separation

The separation of lignin from black liquor is an option in the pulp mills that enables lower recovery boiler capacity (Vakkilainen and Välimäki 2009; Hamaguchi *et al*. 2011; Tomani 2013). Lignin removal from the black liquor decreases the amount of organics in the black liquor burned in the recovery boiler and thus the heat load. If no additional use is found, the separated lignin can be burned in the lime kilns. More and more of the separated lignin is sold to advanced uses, as lignin can be used in e.g. several new corrugated products and as part of insulation materials.

In typical lignin separation, the black liquor, at a dry solids content of 30-40%, from the evaporator is acidified. When acid is mixed into the black liquor to reduce the pH, the precipitation of lignin occurs (Lundberg *et al*. 2012; Culbertson *et al*. 2014; Dieste *et al*. 2016). Before lignin is taken out, it is dewatered and washed using often a press filter. Most plants use a two-step approach where the first acidification is done with CO_2 and the second with sulfuric acid (H_2SO_4). Carbon dioxide is used to reduce the need of additional sodium hydroxide to balance the mills Na-S. The final pH decrease is done with sulfuric acid, as CO_2 cannot acidify the solution enough. It should be remembered that produced raw lignin needs to be purified if it is not used as fuel (Ziesig *et al*. 2014).

A LignoBoost-lignin separation plant operates at Stora Enso's Sunila mill. Sunila plant produces 50,000 tons of dried lignin per year. Another commercial LignoBoost is at the Domtar Plymouth mill North Carolina, USA. It produces 25,000 tons of dried lignin per year.

Figure 5: An industrial lignin separation plant at Domtar, Plymouth (Tomani 2013).

Production of transport fuels

Building a feasible renewable diesel production plant requires a source of low-cost feedstock, industrially proven technology as well as efficient plant and energy integration. Production of second generation biofuels is an attractive option for the forest industry. Already in the early 90s, it was seen that old pulp mills could profitably be converted to produce biofuels. At the same time, combining biomass gasification and Fischer-Tropsch (FT) synthesis plant was seen as a way to produce syncrude. This can be upgraded to renewable diesel (Vakkilainen *et al*. 2009). Recently many forest industry players are actively seeking for new business opportunities via different biomass-to-products processes with integration to mill sites (Hamaguchi *et al*. 2012; Hamaguchi *et al*. 2013). Scandinavian modern mills see opportunities in producing biofuels from tall oil (Weymarn 2015). There is an operating plant at Lappeenranta, Kaukas mill that makes transport biofuel BioVerno from tall oil. The capacity of the plant is 100 000 tons of fuel per year. Another example is SunPine company in Piteå that has also a plant of 100 000 tons of fuel per year.

Basically, it is straightforward to produce BioSNG by gasifying biomass (like in biomass gasifier to lime kiln) and then using purification to make methane (Aleshina and Vakkilainen 2012). Joutseno mill in Lappeenranta went as far as obtaining environmental permit, but due to low price of ETS CO2 the project did not materialize. Biogas can also be produced by anaerobic gas production from e.g. biosludge. This is what EcoEnergy does at Äänekoski mill. The capacity of the plant is about 25 GWh of biogas per year with yield of about 30%.

New operating model

Building a feasible renewable bioproduct plant is not easy and requires specialist know-how. Similarlily handling biomass logistics and strict environmental conditions is not easy for a small startup. Therefore, the pulp industry needs to adopt a new operating mode. Even though up until the 1970s pulp mills produced multiple products they then transferred to one product only mode.

The whole mill operation needs to be looked at. Costs of unavailability during shut-downs and services like electricity, steam and waste disposal need to be considered. Co-existence and dependency from main product are known business models from e.g. car industry. The pulp industry has practiced this with new chemical plants located beside some of the new pulp mills. This mode of operation needs to be extended to other, biobased products.

NEGATIVE CO2 WITH BECCS

In the quest to reduce the effect of global warming, the greenhouse gas reductions need to be high and the decarbonisation of the main economies should be done fast. In most scenarios considerable negative emission technologies (NETs) need to be deployed. The negative emissions range from 5 $GtCO_2$/a to 21$GtCO_2$/a at the end of the 21st century (Fuss *et al.* 2018). Main alternative is capturing carbon dioxide emissions from biobased processes (BECCS).

Sources of CO_2

CO_2 is formed in pulp mills primarily during combustion (Kuparinen *et al.* 2018). The main CO_2 sources are the recovery boiler, the biomass boiler, and the lime kiln. In addition, non-condensable gas (NCG) destruction with several small vent streams are other, but negligible sources. Of these, typically, the lime kiln is the major fossil CO_2 source, Figure 6. The main biobased CO_2 emission sources in a pulp mill is the recovery boiler stack. Biomass boiler stack (when present) is typically much smaller. CO_2 removal is energy intensive and removal cost depends on removal method and process integration possibilities.

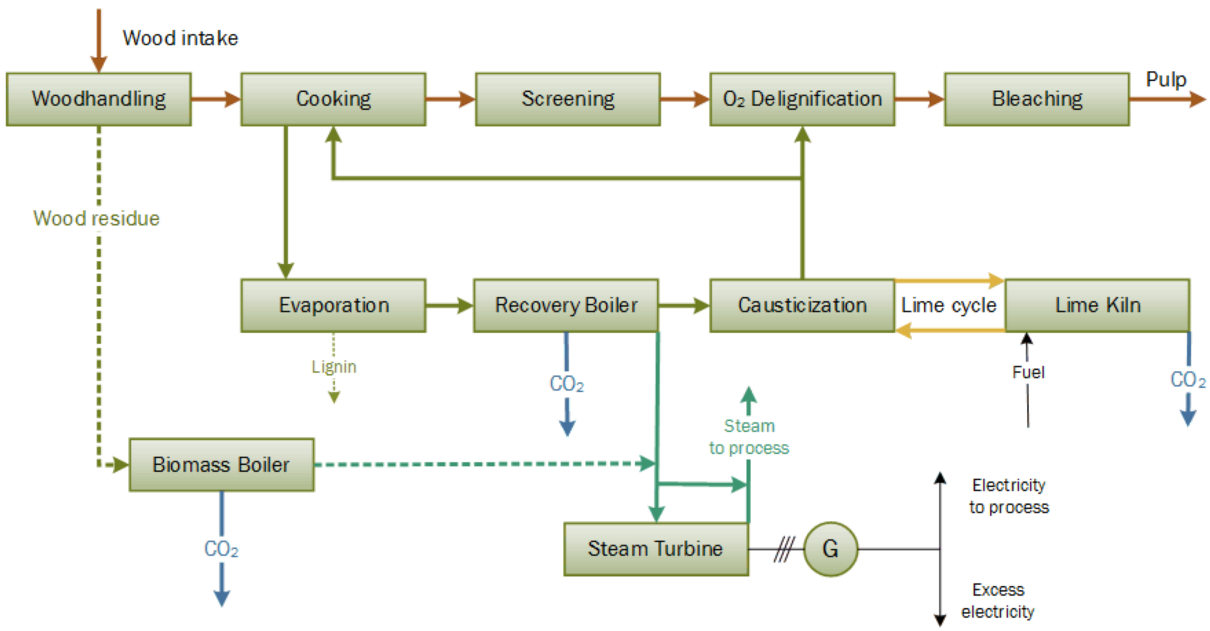

Figure 6: Kraft pulp mill operations and alternative CO_2-removal streams

CO_2 removal

Considering only carbon flows across the plant borders presented (Figure 5), a simplified carbon balance and net CO_2 emissions from the process ($CO_{2,net}$) can be defined for the process as follows (Kuparinen *et al.* 2018)

$$CO_{2,net} = (C_f + C_{CaCO3})\frac{M_{CO2}}{M_C} - \eta_{CCU}\eta_{CC}CO_{2,total}$$

where η_{CC} is the share of CO_2 removed from the total CO_2 emissions ($CO_{2,total}$) and depends on capture method and how it is applied. At first, it is feasible to apply CO_2 removal to the recovery boiler. In the reference mill 87 % of total CO_2 exits with flue gases from recovery boiler stack. If the efficiency of CO_2 capture process is 90 %, it follows that η_{CC} = 78 %. η_{CCU} gives the CO_2 emission effect of selected CO_2

utilization and storage route, where 100 % means that all captured CO_2 is permanently removed from the atmosphere, without additional emissions from the storage/utilization process. C_f and C_{CaCO3} are carbon flows into the plant in fossil fuel and limestone makeup streams. For the studied Mill, where the capture process is applied only on recovery boiler, substantial 2.4 Mt,CO_2/ADt recovery potential for the biobased CO_2 is found. Specific emissions, however, have large variation, depending on capture process efficiency and the CO_2 emissions of the utilization route.

CO_2 removal methods

Different technologies can be utilized for CO_2 capture from pulp mills. Leeson *et al.* (2017) recently presented techno-economic analysis and review of different CO_2 removal methods applied to different industrial CO_2 sources. There is no single winning technology in terms of costs. Amine-based post combustion CO_2 capture systems are a proven technology that is commercially available. CO_2 capture efficiency of monoethanolamine (MEA) process is usually between 80-90 % and as a post combustion method it can be applied easily to existing plants. Aqueous solution (30 w-%) of MEA is used as solvent in post combustion capture process. CO_2 is absorbed at temperature 45-50 °C and flue gases needs to be cooled before amine absorption. Desorption occurs at 100-120 °C. Approximately 3.7 MJ/kg,CO_2 heat is needed for sorbent regeneration. Electricity is needed for the process, which increases own electricity use of the mill (Onarheim *et al.* 2017). Recently, Karjunen *et al.* (2017) studied CO_2 capture, transport and intermediate storage logistics. For Finnish energy system the cost of biogenic CO_2 for utilization was 40 – 44 €/t,CO_2. Low costs were encountered for large industrial (e.g. pulp and paper) sources. Table 5 shows the effect of large, 0.65 MtCO2/a, conventional MEA capture process to reference mill.

Table 5. The main parameters for reference mill, when MEA based post-combustion process is used to capture CO_2 from recovery boiler flue gas.

	Unit	Base	CO_2 capture
CO_2 capture	t/d	-	1881
Mill steam use	t/h	860	1008
Power generation	MW	195	179
Power consumption	MW	116	127

CONCLUSIONS

Modern kraft pulp mills are still going strong and world capacity is increasing at steady pace. The key to future success is the ability to constantly improve and adapt to needed changes. Pulp mills need to find ways to operate without fossil fuels. One practical example is gasification based lime kilns. On the other hand, a new era of biorefinery-focused production is emerging. In addition to pulp, there are various new possibilities to produce additional value. The world is going towards bioproducts and pulp mills that operate in new ways are a big part of the future.

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