Lappeenranta-Lahti University of Technology LUT
School of Energy Systems
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Lime kiln fuel options and effects on the pulp mill energy balance

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ABSTRACT

Lappeenranta-Lahti University of Technology LUT

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Energy Technology

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Master´s thesis

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Keywords: lime kiln, lime kiln fuels, energy balance, pulp mill

Pulp mills are increasingly striving for fossil fuel-free pulp production. Currently, however, most of the world's pulp mills use heavy fuel oil and natural gas in the lime kiln as the only fossil fuel within the mill site during normal production. The aim of this thesis is to examine the most potential fuel options for fossil fuel replacement in a lime kiln and the effects on the pulp mill energy balance.

In the literature part, the basic principles of the chemical recovery of a pulp mill are presented, as well as the operating principles of the lime cycle and lime kiln in a little more detail way. The traditional and alternative fuels of the lime kiln are also examined based on the literature, from the important perspectives for the use of each fuel in the lime kiln.

In the experimental part of the work, hypothetical case mills were defined, and fiber and chemical balances were calculated, which resulted in the fuel flows and production capacities required for the calculation of energy balances. Using the main fiber and chemical balance results, energy balances were calculated for the predefined case mills. The energy balance results, and the effects of the fuel options were compared to the base case, with the traditional fossil fuel firing mill model.

Case mills were also compared in terms of operating costs (OPEX), and capital expenditures (CAPEX). The differences for OPEX and CAPEX were evaluated and the economic viability of each case was discussed, by comparing to the cases firing fossil fuels.

TIIVISTELMÄ

Lappeenrannan-Lahden teknillinen yliopisto LUT

School of Energy Systems

Energiatekniikan koulutusohjelma

Paavo Tiitta

Meesauunin polttoainevaihtoehdot ja niiden vaikutukset sellutehtaan energiataseeseen

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Hakusanat: meesauuni, meesauunin polttoaineet, energiatase, sellutehdas

Sellutehtaat pyrkivät siirtymään entistä enemmän fossiilisista polttoaineista vapaaseen sellun tuotantoon. Tällä hetkellä kuitenkin suurin osa maailman sellutehtaista polttaa fossiilisia polttoaineita meesauunissa. Sellutehtaissa meesauuni onkin normaalissa tuotantotilanteessa ainoa fossiilisia polttoaineita käyttävä yksikkö. Tämän työn tavoitteena on tarkastellaan potentiaalisia fossiilivapaita polttoainevaihtoehtoja meesauunille ja niiden vaikutusta sellutehtaan energiataseeseen.

Kirjallisuus osuudessa esiteltiin sellutehtaan kemiallisen talteenoton perustoiminta, sekä hieman tarkemmin kalkkikierron ja meesauunin toimintaperiaatteet. Kirjallisuusosuudessa käsiteltiin myös meesauunin perinteisiä sekä vaihtoehtoisia polttoaineita kirjallisuuteen perustuen, meesauunin ja sellutehtaan toiminnan kannalta tärkeistä näkökulmista.

Työn kokeellisessa osuudessa määritetyille tehdasmalleille laskettiin kuitu- ja kemikaalitaseet, joiden avulla selvitettiin energiataseiden laskemiseen vaaditut polttoainevirrat ja osastojen tuotantokapasiteetit. Päätasetulosten avulla määritetyille tehdasmalleille laskettiin energiataseet, joiden tuloksia ja polttoainevaihtoehtojen vaikutuksia tehtaisiin verrattiin perinteiseen fossiilisia polttoaineita käyttävään tehdasmalliin.

Työn lopussa vertailtiin myös, mallitehtaiden käyttö- (OPEX) ja investointikustannusten (CAPEX) eroja sekä niiden vaikutusta mallitehtaiden taloudelliseen kannattavuuteen.

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Roman

h	Enthalpy	kJ/kg
p	Pressure	bar
T	Temperature	˚C

Abbreviations

ADt	Air dry tonne (10 % moisture content)
af	ash-free
BB	Bark boiler
BDt	Bone dry tonne (0 % moisture content)
CAPEX	Capital expenditures
CFB	Circulating fluidized bed
CNG	Non-condensable gas
CNCG	Concentrated non-condensable gas
CTO	Crude tall oil
DNCG	Diluted non-condensable gas
DS	Dry solids
DTO	Distilled tall oil
d	Day
ESP	Electrostatic precipitator
ETS	Emission trading system
FW	Feedwater
LP	Low pressure
NPE	Non-process elements
MP	Medium pressure
OPEX	Operational expenses
OWL	Oxidized white liquor
RB	Recovery boiler
SOG	Stripper off-gas
sub	solids under bark
TG	Turbogenerator
TOFA	Tall oil fatty acids
TOP	Tall oil pitch
TOR	Tall oil rosin
TRS	Total reduced sulphur

1 INTRODUCTION

Modern kraft pulp mills are moving towards carbon neutral and fossil-fuel-free pulp production. Most of the world's pulp mills are still burning heavy fuel oil or natural gas inside the mill site. The biggest and in most cases, only fossil fuel consumer is a lime kiln, which is part of the pulp mill's chemical recovery and lime cycle. For pulp mills to achieve this fossil-fuel-free production, alternative fuel options have been studied and some of the mills, especially in Nordic countries, pulp mills have started to utilize those options. A good example of such a mill is Metsä Fibre Äänekoski bioproduct mill, which was launched in 2017 and, uses no fossil fuels at all (Metsä Fibre 2019).

A lime kiln is a long rotating tube, which converts lime mud (calcium carbonate) to lime (calcium oxide) with the help of high temperature. The heat is brought to the lime kiln via burner, in which different fuels can be used for combustion. Lime from the lime kiln is used in a causticizing reaction, where it is used to convert green liquor to white liquor. In this process, lime reacts to calcium carbonate and it is separated from the white liquor and reburned in the lime kiln.

In the literature part of this study, basic principles of pulp mill chemical recovery and the lime cycle are presented and a more detailed description of the lime kiln process, structure, and operation is done. Also, alternative lime kiln fuel options are introduced, based on literature. These fuel options are biomass-based fuels from mill’s wood handling and by-products of the pulping process. The fuels are considered from process requirement-, fuel properties-, availability-, operation- and economic point of view.

In the experimental part of this thesis, the objective is to calculate energy balances for different cases and compare the effects of the fuel option on the energy balances of the case mills. The formation of energy balances requires calculation of each case mill’s main fiber- and chemical balances, to solve the available alternative lime kiln fuel flows from the mill’s side streams and each department's production capacity for heat and power consumption. The cases are formed for northern bleached softwood kraft pulp mill with an annual production capacity of 1000 000 ADt and southern bleached hardwood kraft pulp mill with an annual production capacity of 1000 000 ADt. For both cases, a base case with heavy fuel oil as lime kiln fuel is calculated for comparison for alternative fuel cases. The cases are

described in a more detailed way in chapter 4. The main fiber- and chemical balances and energy balances are calculated with AFRY's balance tools and the reference data being used for calculation are based on former projects. After examining the energy balance results and the effects, the economic viability of case mills is examined, comparing operating (OPEX) and capital expenditures (CAPEX).

2 CHEMICAL RECOVERY

Kraft pulp mill consists of fiber line and chemical recovery. The chemical recovery of the kraft pulping process can be defined with two different cycles presented in Figure 1, the cooking chemical cycle, and the lime cycle. The main objectives for chemical recovery are a recovery of the expensive cooking chemicals, combustion of dissolved organic material in a recovery boiler to produce heat, high-pressure steam, power, and recovery of the by-products, such as tall oil, and turpentine (Vakkilainen & Kivistö 2017, 67; Hart 2021, 13). Over the years, the concept of closed cycles inside pulping process has changed. The traditional concept of closed cycles to minimize emissions and effluent has changed more towards an open cycle, where all the side streams and hidden potential are utilized in form of converting it into additional revenue from raw materials, fuels, and energy. (Pehu-Lehtonen 2019)

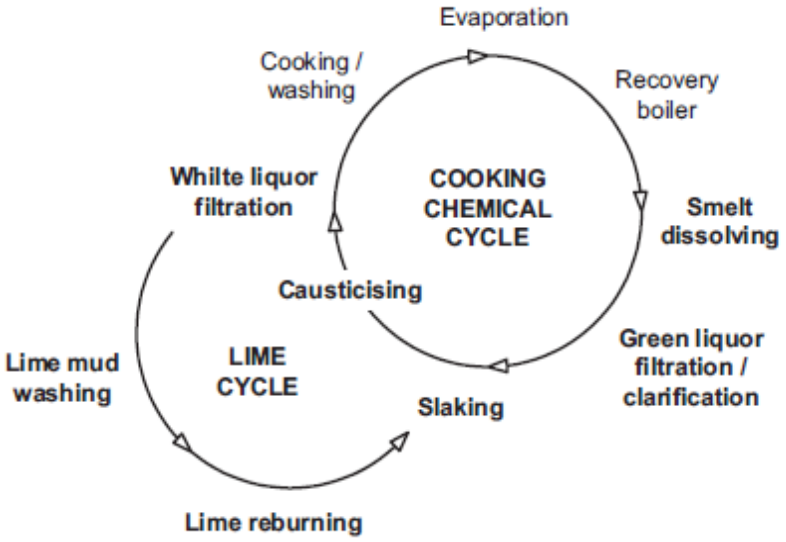

Figure 1. Overall view of chemical recovery (Krotscheck & Sixta, 2006, 986)

In the cooking process, the wood chips are fed to the digester. The wood fibers are extracted to separate pulp fibers with the presence of cooking chemicals (white liquor), elevated heat, and pressure. The main components of the white liquor are alkaline, sodium hydroxide NaOH and sodium sulphide Na_2S. After the cooking process, pulp, and black liquor, which is white liquor consisting of the extracted wood component from the cooked chips are discharged to a blow tank. The pulping process continues with screening and oxygen delignification as the black liquor is washed from the pulp with hot water in a countercurrent washing.

The mixture of black liquor and water is called weak black liquor, which is then pumped to the evaporation plant. (Sixta, et al. 2006, 111-112)

2.1 Evaporation

The evaporation plant aims to increase the dry solid contents of black liquor while losing a minimal amount of chemicals (Vakkilainen 2008, 11). The weak black liquor from pulp washing is typically in 13-18 % dry solids content (Suhr, et al. 2015, 205; Parviainen, et al. 2008, 38 & Krotscheck & Sixta 2006, 974). Before evaporation, the weak black liquor is concentrated to 18-22 % dry solids content, which is then fed to the evaporation plant. This concentrated black liquor is called feed black liquor (KnowPulp 2021). The black liquor arriving at the evaporation plant contains small amounts of by-products depending on the wood species used for pulp raw material. These by-products are, for example, methanol, turpentine, lignin, and soap. However, the soap is only obtained when using softwood. These by-products are generated in the cooking process and separated in connection with the evaporation plant. (Vakkilainen & Kivistö 2017, 68)

A typical modern evaporation plant has five to seven evaporators connected in series. The modern evaporators are rising or falling film-type evaporators. The liquor evaporation usually happens in a counter-current or mixed co-current way, which both are presented in Figure 2. The principle of a multiple effect evaporation plant is that live steam is only used in the first evaporation unit to evaporate water from the black liquor. The vapor from the first unit is then utilized and condensed in the second evaporation unit to evaporate more water. This process continues in each unit until the last one. (Valmet, 2015) The black liquor must be concentrated at the minimum level of 65-70% of dry solids (Smook 2016, 142). In a modern evaporation plant, the dry solids content can be increased up to 85 % (Valmet, 2015; Parviainen, et al. 2008, 38 & Clay 2008, 3.1-1).

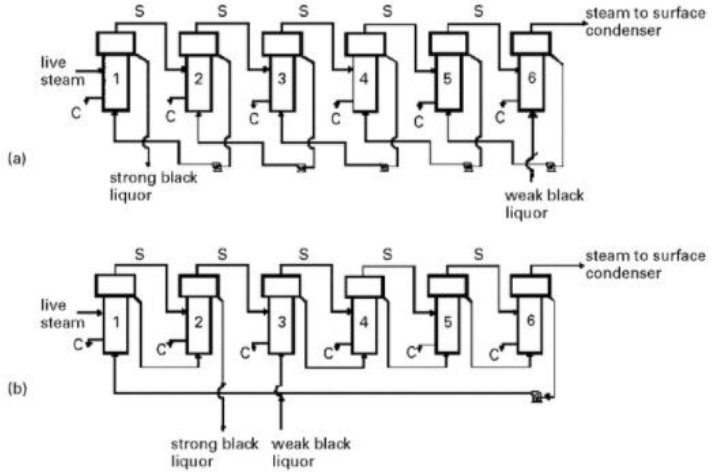

Figure 2. Different evaporation flow-types, where S is steam & C is condensate. a) counter-current b) mixed co-current. (Theliander 2009, 302)

2.2 Recovery boiler

The main objectives for the recovery boiler are steam production with the combustion of organic substances in black liquor, recovery of the inorganic cooking chemicals, and disposal of harmful substances efficiently. The combustion of organic wood compounds in black liquor produces heat in the furnace and with different heat surfaces in the boiler, water is converted into high-pressure steam. (Vakkilainen 2008, 12) This high-pressure steam is passed to the turbine, where it generates electricity for the mill's use. Mills usually have different types of extraction turbines and these turbines are also used for low- and medium-pressure steam extraction. The low- and medium-pressure steam are used in different stages of the pulp and paper mill (Vakkilainen 2005, 1-2). The modern pulp mills are energy self-sufficient, which means that the mill produces more energy than it consumes. This allows the surplus energy to be converted into additional revenue for example, by selling surplus electricity and heat to the national electricity grid or district heating network.

A modern recovery boiler in Figure 3, consists of several different heat transfer surfaces. The main heat surfaces are furnace, superheaters, boiler generating bank, and economizer. Over the years, recovery boilers have developed, and new improved properties and systems have been introduced to increase the boiler power and efficiency. A modern recovery boiler performance can be improved with the combustion of black liquor with higher dry solid content, preheating of combustion air and feedwater and, recovery of heat from the vent and

flue gases. Also, improved recovery boiler steam parameters have been introduced, but the increase in main steam values might require actions against heat exchange surface fouling and corrosion. These actions against fouling and corrosion can be more durable and expensive heat surface material and sootblowing systems for ash deposits in heating surfaces. (Hart 2021, 31-34 & Mansikkasalo, et al. 2015)

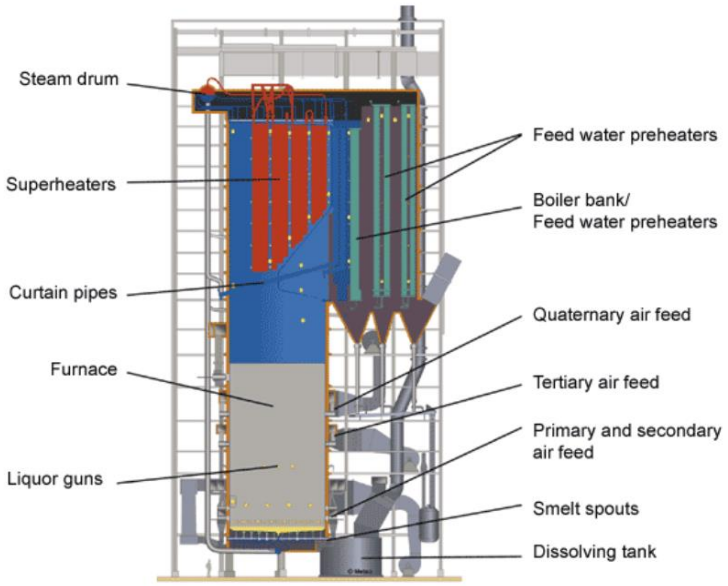

Figure 3. Structure and main components of a modern recovery boiler (KnowPulp, 2021)

The black liquor is sprayed as droplets to the bottom of the furnace to a char bed. On the way to the bed the droplets dry, ignite, and in the bed forms char on combustion. A reduction reaction occurs during char combustion. Carbon reacts with sodium sulphate ($NaSO_4$) to form sodium sulphide (Na_2S), which is one of the active cooking chemicals. The reaction forms carbon monoxide (CO) and carbon dioxide (CO_2) gases. Also, when char burns in the bed, reduction of sodium occurs if it contacts oxygen, which should be prevented with a new continuous layer of molten smelt on top of the bed. These reactions are presented in the following equations 1-3. (Vakkilainen 2008, 101-102)

$$Na_2S + 2O_2 \rightarrow Na_2SO_4 \quad (1)$$

$$Na_2SO_4 + 2C \rightarrow Na_2S + 2CO_2 \quad (2)$$

$$Na_2SO_4 + 4C \rightarrow Na_2S + 2CO \quad (3)$$

Most of the inorganic substances remain after the char combustion and form smelt. The smelt mostly consists of sodium carbonate (Na_2CO_3) and sodium sulfide (Na_2S). The smelt flows away from the furnace to a smelt dissolving tank via smelt spouts, which are located at the bottom of the furnace. (Krotscheck & Sixta 2006, 981)

2.3 Causticizing

The causticizing process is part of both, liquor, and lime cycle. The Causticizing plant converts molten smelt from the recovery boiler into white liquor with the burned lime. The causticizing plant enables a closed chemical cycle in the pulp mill, which is beneficial for mill economics and the environment.

The causticizing process receives smelt from the recovery boiler. In the dissolving tank, the smelt is dissolved into weak white liquor to form green liquor. From the dissolving tank, green liquor is pumped to the stabilization tank, where green liquor density and temperature are stabilized. After stabilization, the green liquor is clarified and pumped to the green liquor storage tank in the causticizing plant. From the storage tank, green liquor is fed to the slaker, in which the burned lime (CaO) from the lime kiln is also fed, to start the causticizing process. In the slaker, the burned lime first reacts with the water contained in the weak white liquor to form calcium hydroxide ($Ca(OH)_2$), according to equation 4. Then the reaction product, calcium hydroxide reacts with green liquor to form white liquor and lime mud according to equation 5. (Theliander 2009, 335-336)

$$CaO\ (s) + H_2O\ (l) \rightarrow Ca(OH)_2 \quad (4)$$

$$Ca(OH)_2\ (s) + Na_2CO_3\ (l) \rightarrow 2\ NaOH\ (l) + CaCO_3\ (s) \quad (5)$$

The slaked lime and sodium carbonate react rapidly in the slaker (Figure 4), but the reaction slows down after 15-30 minutes. Unslaked lime and grits are raked from the bottom of the slaker chamber to a classifier screw, which removes grits from the lime milk.

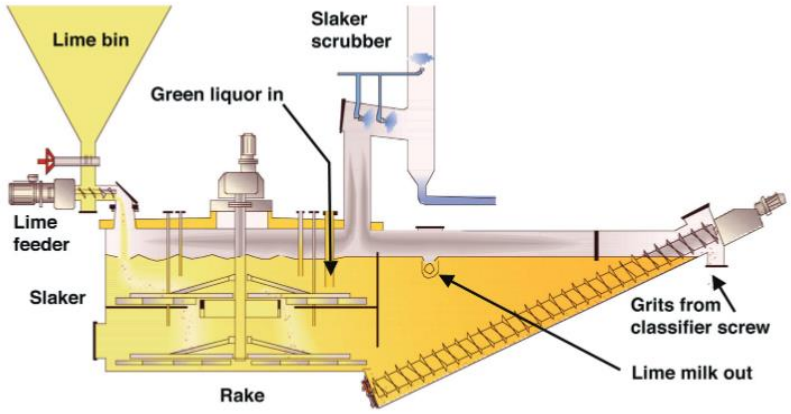

Figure 4. Slaker (Arpalahti, et al. 2008, 146)

The reaction continues to 70 % completion and the rest takes place in the causticizing vessels. (Arpalahti, et al. 2008, 147) A typical causticizing plant has three causticizing vessels. The purpose of these causticizing vessels is to complete the remained unreacted reaction after the slaker. The reaction is acquired to completion with more time. Typically, the first two vessels contain three chambers, and the last chamber contains one to two. Each chamber also contains an agitator. The number of chambers varies mill and equipment supplier specific. The agitators enable the liquor to have continuous contact with unreacted lime mud particles. Lime milk throughput time in causticizing tanks must be distributed evenly for a good causticizing result. (Arpalahti, et al. 2008, 147-148) The reaction is controlled by adjusting the lime feed. The target is that all the lime is converted into lime mud and the sodium hydroxide content in the lime milk is over 80 %. The sodium hydroxide content in lime milk is called causticizing efficiency. The causticizing efficiency is presented in equation 6 (KnowPulp, 2021). Typically, the causticizing plant tries to run with a causticizing efficiency of 82-84 % (Grace & Tran 2009, 19).

$$\text{Causticizing efficiency CE-\%} = \frac{NaOH}{(NaOH + +Na_2CO_3)} \quad (6)$$

After causticizing chambers, the lime milk, which consists of white liquor and lime mud (Ca_2CO_3), the white liquor is separated from lime milk. The most used method for white liquor separation is filtration. The target is to filtrate clear white liquor from lime milk, without any residual lime mud. In modern pulp mills, the most common and efficient filtration method is a pressurized disc filter. (Theliander 2009; Arpalahti, et al. 2008) The pressurized

disc filter is also used in lime mud washing and the principles of the disc filter are described more accurately in the next chapter 2.4.

2.4 Lime kiln

Lime kiln is part of the lime cycle. Lime is a chemical used to convert the green liquor from the recovery boiler back to white liquor via a causticizing reaction. In the lime kiln calcium carbonate ($CaCO_3$) formed in the causticizing reaction is burned and converted back into calcium oxide (CaO). The whole lime kiln process consists of lime mud handling, lime burning, and lime treatment after combustion. The lime kiln unit is presented in Figure 5. The lime burning process requires high temperature so external heat is brought to the process via a burner in the other end of the kiln. The size of a rotary lime kiln varies, depending on the kiln's capacity. A typical lime kiln's diameter is 4-4,5 meters and its length varies from 100 to 140 meters (Arpalahti, et al. 2008, 161). The diameter of the largest modern kilns can reach up to 5,5 meters (Manning et.al. 2021, 64).

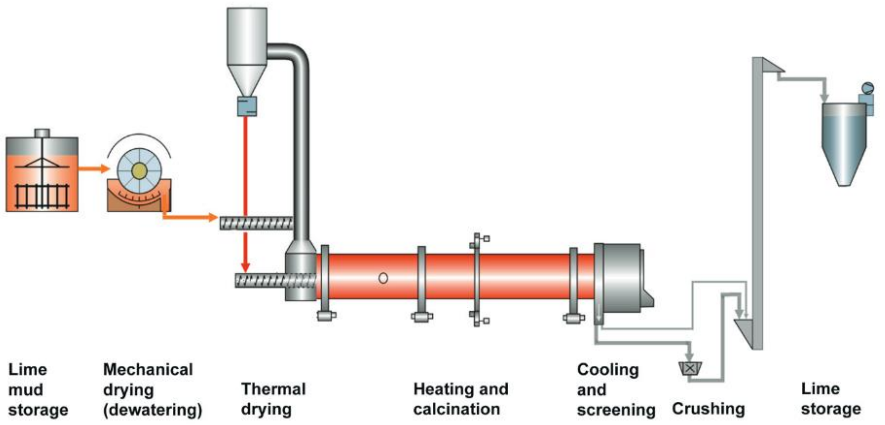

Figure 5. Lime kiln process (Arpalahti, et al. 2008, 162)

Most pulp mills use fossil fuels to produce heat for the lime burning process. Considering the whole mill, the lime kiln is typically the only user of fossil fuel during a normal operation. There are alternative non-fossil fuel options that the lime kiln process can use. These different fuels and ways of operation are reviewed in chapter 3.

Lime mud storage tank receives lime mud from causticizing white liquor filtration. From the lime mud storage, the lime mud is washed in a rotary drum filter to recover the last remains of soluble liquor. The wash filtrates (weak wash liquor) are used in the recovery boiler smelt

dissolving tank. (Krotscheck & Sixta 2006, 986) There are two types of lime mud filters used in a modern causticizing plant, disc, and drum filter. Figure 6 presents, how the pressurized disc filter consists of multiple disc-shaped filter elements. The elements rotate with a horizontal center shaft. In the bottom of the vessel is lime milk, which covers part of each filter disc. The disc surface is a filter cloth, which separates lime mud and white liquor with a pressure difference. The white liquor passes through the filter clothing and the lime mud stays on the cloth surface. The liquor goes via the center shaft to a pressurized tank, where gases and liquid are separated. The lime mud cake stays on the surface and it is washed through the round and the top layer of the mud is removed with a scraper before rotating back to the lime milk. (Bajpai, 2017; Arpalahti, et al., 2008)

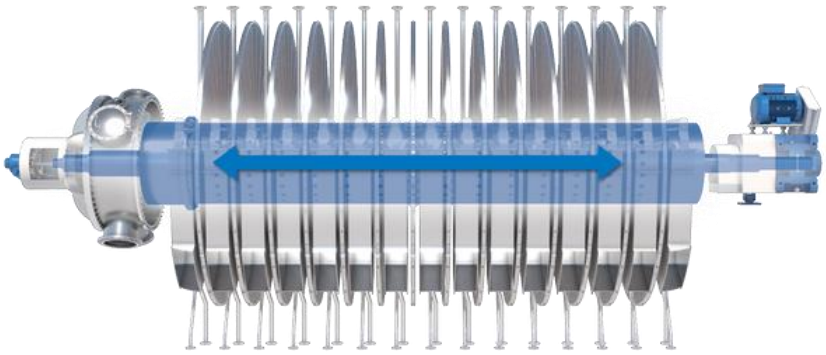

Figure 6. Center shaft with disc filters. (ANDRITZ, 2021)

The second option, the rotary drum filter works with almost the same principle as the disc filter. The washing liquid is distributed to the surface of the filter-clothed drum. The pressure difference allows the liquor to pass through the filter. The lime mud on the drum surface is washed and the liquor is replaced with air to achieve a decrease in the moisture content. At the end of the rotation, the lime mud cake is scraped from the surface like in a disc filter. (Theliander 2009, 353-354)

After washing lime mud contains 75-85 % of dry solids. Before combustion, the lime mud is dried. A more traditional way for lime mud drying is the chain section in the lime kiln cold end. The chains can be arranged with different variations but the main objective for the chains is to improve heat transfer from the relatively cold flue gases to lime mud. The chains increase the regenerative heat exchange surface and improve lime mud drying. In refurbished

and new lime kilns, external drying is a more common way, because of higher efficiency. The external dryers increase lime kiln capacity and volume because the length used for chains in a traditional system can be now utilized for lime mud preheating. (Manning, et al. 2021, 67) With an external dryer, the lime mud is dried in the cyclone with the help of hot flue gases formed at the burner end of the kiln. Lime mud with a suitable dry solids content flows with the hot flue gases into the cyclone, where the heat evaporates the rest of the moisture from the lime mud. If the dry solids content is too low after lime mud washing, the flue gases will not be able to lift lime mud into the cyclone, causing the lime mud to fall directly to the feed end of the kiln, lowering the kiln efficiency. (Järvensivu, et al. 2001, 591). The cold-end with lime mud washing and cyclone dryer is presented in Figure 7.

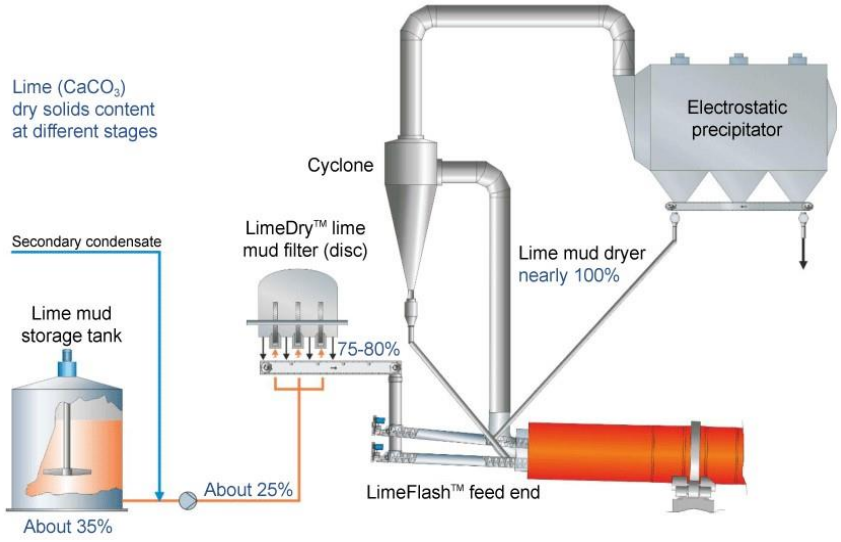

Figure 7. Lime kiln feed-end with lime mud washer and dryer. Dry solids content is also shown in the figure. (KnowPulp, 2021)

The cyclone discharges the dried lime mud into the lime mud conveyor screw and to the lime kiln. The kiln is inclined between 1,5 and 3° and rotates about 0,5 to 2 rounds per minute. The kiln is supported with riding rings, which are supported by a concrete base. The kiln is rotated with a large electric motor and gearbox. Lime mud is fed to the kiln's higher end. The angle and rotation of the kiln transport the lime forward in the kiln. During normal operation the lime pass time through the kiln is 1,5 hours to 4 hours. (Adams, 2008) The lime mud burning in the kiln can be divided into four zones (Theliander 2009, 355):

1. Lime mud drying
2. Lime mud heating to calcination temperature
3. Calcium carbonate $CaCO_3$ calcination
4. Lime cooling before leaving the kiln

As the lime proceeds in the kiln, the temperature of the solids increases as lime mud closes the firing end. The four phases of lime burning, gas, and solid temperatures are presented in Figure 8.

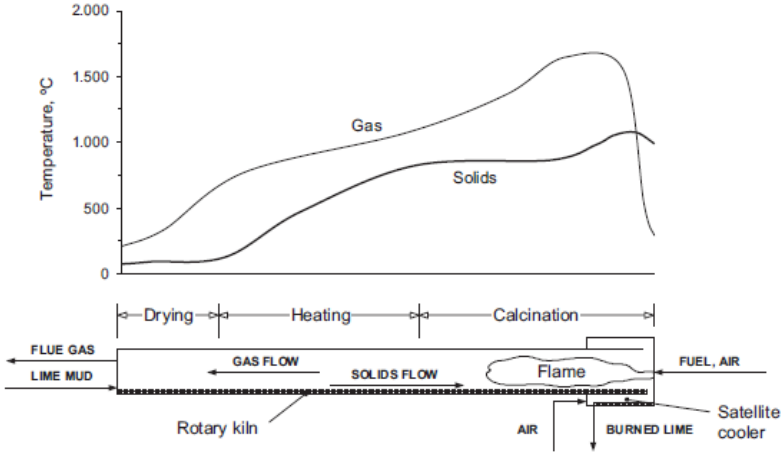

Figure 8. Different phases of lime burning and temperature of flue gases and lime solids. (Krotscheck & Sixta 2006, 991)

In the drying zone, the rest of the moisture in the lime mud evaporates with the direct heat exchange with hot flue gases. After the drying zone lime mud is heated up to 800 °C, which is the required temperature for calcination reaction. (Smook 2016, 157). In the calcination zone, calcium carbonate reacts to calcium oxide and carbon dioxide. At the end of the calcination zone, the lime mud temperature increases to 1100 °C. The reaction is presented in equation 7. The calcination reaction is an endothermic reaction, which means the reaction requires energy (heat) to take place.

$$CaCO_3\ (s) + \text{heat} \rightarrow CaO\ (s) + CO_2\ (g) \qquad (7)$$

Reburned lime quality can be indicated by a few indicators. The lime's residual carbonate and availability are commonly used as a quality indicator. A residual carbonate indicates the content of calcium carbonate (% $CaCO_3$), that did not calcinate. In normal operating

conditions the factors affecting the residual carbonate content are the size of the lime nodes and retention time in the kiln controlled by the kiln's rotation speed. Also, the lime mud feed to the kiln must be constant. The target for residual carbonate content is 1-3 % when reburned lime is strong, porous and it reacts as desired in the slaker. If residual carbonate content is too low, the lime loses its reactivity and if the content is too high, the lime reacts faster than desired in the slaker. (Theliander 2009, 128 & Hart 2021, 39) The second indicator, lime availability measures the content of calcium oxide (CaO) in the reburned lime. A typical value for lime availability is in the range of 80-95%. The value is affected by the level of impurities and residual carbonate. A sudden decrease in lime availability can be an indication of these materials in the lime cycle. The impurities in the lime cycle depend on wood species used for pulping, the efficiency of the causticizing process, kiln's firing conditions, fuels used for kiln firing, and refractory brick type. These impurities should be avoided because they cause operational issues for the whole lime cycle, and thus increases the need for make-up lime and operational costs. (Hart 2021, 40-41)

The heat is brought to the system by the burner in the hot end. The fuel used in the burner is typically heavy fuel oil, natural gas, or alternative non-fossil fuel. These fuel options are presented in chapter 3. The structure of a typical natural gas burner is presented in Figure 9. The structure of the burner might vary, depending on the fuel used in the lime kiln. Also, the burner can be positioned at the kiln to adjust the lime kiln conditions (KnowPulp, 2021).

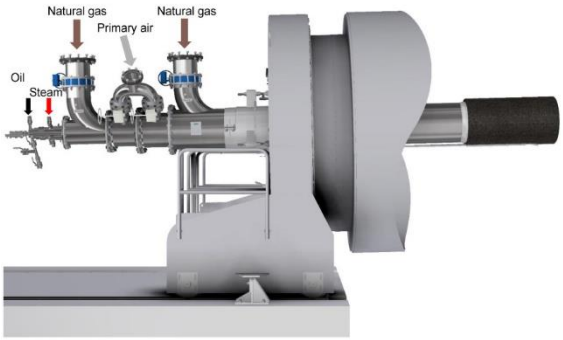

Figure 9. Lime kiln natural gas burner (KnowPulp, 2021)

At the end of the kiln near the burner, lime is heated to a temperature of 1000-1300 °C and then discharged to a lime cooler. (Theliander 2009, 359). The most used cooling systems are

sector, satellite, and stationary coolers. The basic principle of satellite and sector cooler is presented in Figure 10.

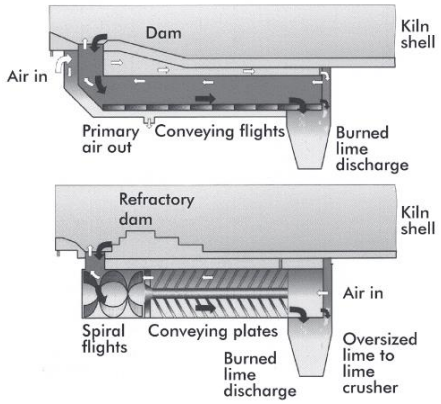

Figure 10. Sector and satellite cooler (Arpalahti, et al. 2008. 166)

The burned lime discharges over the dam and passes to the cooler. The air is brought to the systems counter-current with an induced draft fan. The purpose of the lime cooling systems is to recover heat from the lime to preheat secondary combustion air. The preheated secondary air can cover up to 80 % of the total amount of combustion air (Arpalahti, et al. 2008, 166). Heating the secondary combustion air with a lime cooler reduces lime kilns fuel consumption, which can be seen as a significant benefit (Francey, et al. 2011, 24). With an increased capacity in the kilns, the cooler has become the capacity restricting factor. With the stationary grate cooler, originally used in cement kilns, bigger capacities are achieved. In a stationary cooler, the air is brought to the system the same as in sector and satellite coolers, but the lime is discharged into a stationary grate (Figure 11), that moves lime forward with a special pattern. The efficiency of the stationary cooler is based on significantly better contact with reburned lime and air. (Schorcht, et.al. 2013) Regardless of the type of lime cooler, lime particles are screened, and larger particles are crushed to an even size. A lump crusher or a hammer mill can be used for crushing. The burnt, uniform-sized lime is then stored in a lime silo from where it can be reused in the slaking process.

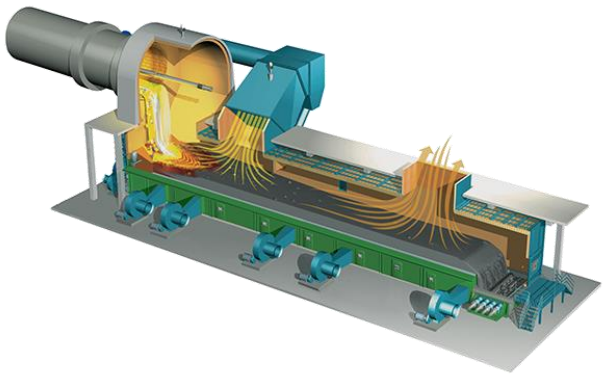

Figure 11. Stationary grate product cooler used in cement industry. (Peters 2021)

Due to the high temperature of the kiln, the inner metal surface of the kiln is lined with refractory bricks (Smook 2016, 158). In different kiln zones, the temperature can vary a lot and because of this, each zone has certain brick material and thickness. The refractory brick will increase kiln availability and decrease the need for maintenance. A typical lime kiln uses high alumina (Al_2O_3), mullite ($3Al_2O_32SiO_2$), or fireclay as refractory brick. In the case of higher flame temperature, more durable material is required, for example, magnesia (MgO). (Tran, 2007)

The most important emissions to air in the lime kiln are lime dust, particulate matter, total reduced sulphur (TRS), nitrogen oxides (NO_x), carbon dioxide (CO_2), and sulphur dioxide (SO_2) (Hynninen 2008, 128; Tran 2007, 2.3-6-7). The main factors affecting emissions from a lime kiln are fuel type, kiln temperature, solids retention time, and gas contact area. The emissions can be controlled with better lime mud washing and moisture removal, with the use of cleaner fuels, better combustion technologies, and flue gas handling systems. Particulate and dust emissions can be decreased with an electrostatic precipitator (ESP). This highly efficient equipment is widely used in lime kiln flue gas handling systems. The principle of ESP is based on a strong electric field, with positively and negatively charged collecting plates. Negatively charged ions are attracted towards positively charged ions, which causes that dust particulates are similarly charged as the ions, and collecting plates start to attract the particles. These particles are discharged from the collecting plates by removing charges, after which the particles fall into the dischargers at the bottom of the ESP. (Arpalahti, et al. 2008, 129-130) The SO_2-emissions can be problematic, and to solve this

problem some of the mills utilize scrubber to reduce SO_2-emissions. Typically, a mill that uses high sulphur content fuel and disposes of non-condensable gases in the lime kiln has problems with high sulphur emissions. Usually, these kinds of mills operate with scrubbers. A scrubber uses water droplets that collide with the gas. The water droplets absorb dust particles and sulphur dioxide into the scrubbing mixture. The mixture is separated from the gas with a cyclone. The most common scrubber type is the venturi scrubber, which is operated at very high efficiency. (Arpalahti, et al. 2008, 131)

The kiln operation can face problems if rings start to form into the kiln. Ring formation decreases kiln capacity and can result in an unplanned shutdown. The ring is formated as lime mud and solids cover the refractory brick with a layer of dust as they slide through the kiln. The dust layer keeps growing with time and restricts lime mud slide towards. (Monahan, et al. 2018, 285) It has not been possible to combine the formation of rings in a lime kiln with one specific thing. However, factors with a strong association with ring formation have been identified, such as moisture in lime mud, sodium, and sulfur content of lime mud, low performance and unstable flame, kiln temperature fluctuation, and fuel-containing contaminants. (Honghi & Barham 1991, 133; Monahan, et al. 2018, 285). In the lime kiln fuels chapter 3, the possible effects of lime kiln fuel on ring formation have been considered from the perspective of fuel quality, impurities, flame-, and temperature stability.

3 LIME KILN FUELS

In this chapter, different fuel options for the lime kiln are presented. Most of the existing lime kilns still, use traditional fossil fuels, natural gas, and fuel oil. However, attractive non-fossil fuel options have become more popular. The most significant drivers for the change are the cost reduction with alternative fuels, growth and difficult price predictability of fossil fuels, and the reduction of CO_2 emissions from the lime kiln. According to Francey et.al. (2009), the most potential options are product gas, which is produced by gasifying bark and sawdust, direct combustion of wood dust, or lignin firing. Also, some other interesting options are tall oil, pitch oil, turpentine, methanol, and hydrogen. All of these options are produced from residues and side-streams of the mill. In this chapter, these fuel options are considered from different aspects such as process requirement, properties, availability, operations, challenges, and economics. The most important properties for fuel combustion are presented in Table 1. Hart (2020A) states that adiabatic flame temperature and lower heating value determine a lot the lime kiln performance and it can be used to compare the kiln performance at some level (Hart 2020A, 263).

Table 1. Important lime kiln fuel properties (Alakangas, et.al. 2015; Arpalahti, et.al. 2008; Francey, et.al. 2009; Hart 2021; Kuparinen & Vakkilainen 2017)

Fuel	Lower heating value LHV [MJ/kg]	Adiabatic flame temperature [°C]
HFO	40-41,5	2210
Natural gas	50	2050
Product gas	5-30	1870-2000
Wood dust	14-19	1950
Lignin	23-26,5	1980
Tall oil	~35	2200
Pitch oil	30-40	1965
Turpentine	19-45	2075
Methanol	~20	2100-2200
Hydrogen	120	2210

The selection of lime kiln fuel affects widely the entire mill and its operations. The fuel affects the mill's emissions, the quality of the reburned lime, white liquor, pulp, and the control of the lime kiln. It also affects the mill energy balance, but it is discussed more later

in the experimental part. The stable operation of the kiln is a key factor in minimizing the negative effects of the above-mentioned things. Although alternative fuels have different technical requirements for the pretreatment system, storing, and burner, the basic principles of the kiln chemistry and basic structure remain the same as when using traditional fuels.

The economic profitability for most fuels depends on current fossil fuel prices and the number of investments and operational costs required for each process. For example, enabling the combustion of the product gas in a lime kiln requires the equipment for drying and gasification, and the high adiabatic temperature of hydrogen requires a more expensive refractory linin on the inner surface of the kiln. For some fuels, economic competitiveness is affected significantly by taxation, subsidies granted by the country, and the resale value of the fuel option as such.

3.1 Fossil fuels

Most of the existing pulp mill lime kilns use natural gas or fuel oil as its main fuel. The properties of these fuels are good for stable combustion and make the combustion easy to control. Although the use of fossil fuels is easy in lime kilns, their position has deteriorated. The high CO_2 emissions generated in fossil fuel combustion, tightening emission limits, increasing fossil fuel prices, and the trend of non-fossil pulp mills have weakened the position of these fuels. The mills have developed more cost and environmentally friendly ways to operate the lime kiln, which are replacing traditional fossil fuels.

3.1.1 Fuel oil and natural gas

Fuel oils can be divided into heavy and light fuel oil, according to its operating characteristics (Alakangas, et al. 2015, 180). Heavy fuel oil (HFO) is the most commonly used fuel in the lime kiln, but also light fuel oil is used in a lime kiln to preheat the kiln, in case of kiln start. Natural gas is also, widely used fuel in lime kilns. Natural gas is a gaseous fossil fuel that can be separated from natural sources or in connection with oil production. Natural gas contains mostly methane and small amounts of ethane, propane, butane, and nitrogen. For example, natural gas used in Finland contains 98 % methane. (Alakangas, et al. 2015, 186)

The kiln operation varies between HFO and natural gas. With these fuels, HFO is known to achieve higher capacity than natural gas. This is due to a higher luminosity of the heavy fuel

oil flame. The higher luminosity increases the efficiency of heat transfer between flame and bed material. Also, HFO's higher adiabatic temperature supports the achievement of higher capacity. (Manning 2021, 101-102) According to Manning and Tran (2015), the difference in luminosity and adiabatic flame temperature of the fuels cause a different temperature profile in the kiln. These temperature profiles are presented in Figure 12. The peak of the temperature profile moves further away from the burner when using natural gas, thus shortening the effective length of the kiln. Also, the kiln outlet temperature increases, and more heat escapes from the kiln, resulting in less effective heat at the same fuel input. (Manning & Honghi 2015, 475)

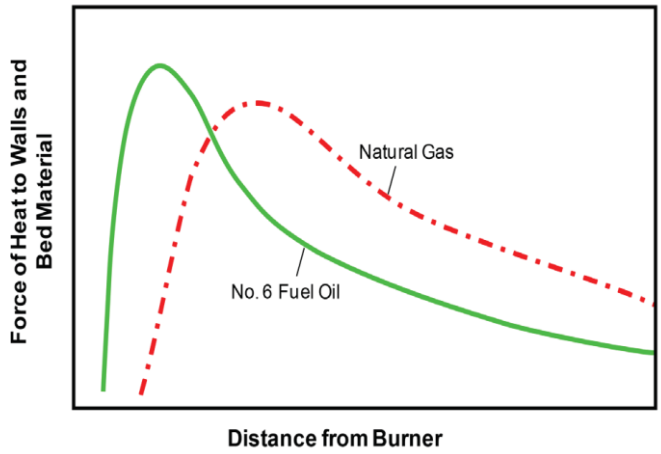

Figure 12. HFO and natural gas temperature profiles (Manning & Honghi 2015, 475)

Heavy fuel oil provides the highest capacity for lime kiln operation and the combustion is more efficient than for example with natural gas. Both HFO and natural gas has very high lower heating value. HFO lower heating value varies between 40-41,5 MJ/kg and natural gas has 50 MJ/kg (Alakangas, et al. 2015, 186; Kaasuyhdistys 2014, 7). The adiabatic flame temperature for oil is 2210 °C and for natural gas 2050 °C. Compared to alternative fuel options, which are based on biomass or pulping by-products, HFO and natural gas have considerably higher heating values and adiabatic flame temperature, which are important properties in lime burning. The popularity of these fuels in lime kilns is based on their wide availability and user experience, which makes kiln operation easier.

3.2 Biofuels

The biofuel solutions to replace fossil fuels in the lime kiln have been found from biomass and pulping by-products. In the case of these alternative fuels, fossil fuels can be replaced either partially, or even completely. These alternative biofuels can be produced from available wood residues and by-products from different stages of the pulping process. The wood residues such as bark and wood dust are generated in the wood processing department, where the wood is debarked, chipped, and screened. Also, a variety of pulping by-products have been operated and studied for lime kiln use. Most of these by-products are generated in the cooking process and extracted in the evaporation plant.

3.2.1 Biogas via gasification

The gasification of biomass is an attractive replacement for fossil fuel use in the lime kiln. Gasification gas (product gas) is used in several commercial pulp mills to replace fossil fuels entirely in the lime kilns. Metsä Fibre Joutseno pulp mill is an example, where lime kiln burning natural gas was replaced during normal operation with a bark product gas (Mäki, et al. 2021). The whole gasification process from raw material handling to lime kiln burner is presented in Figure 13.

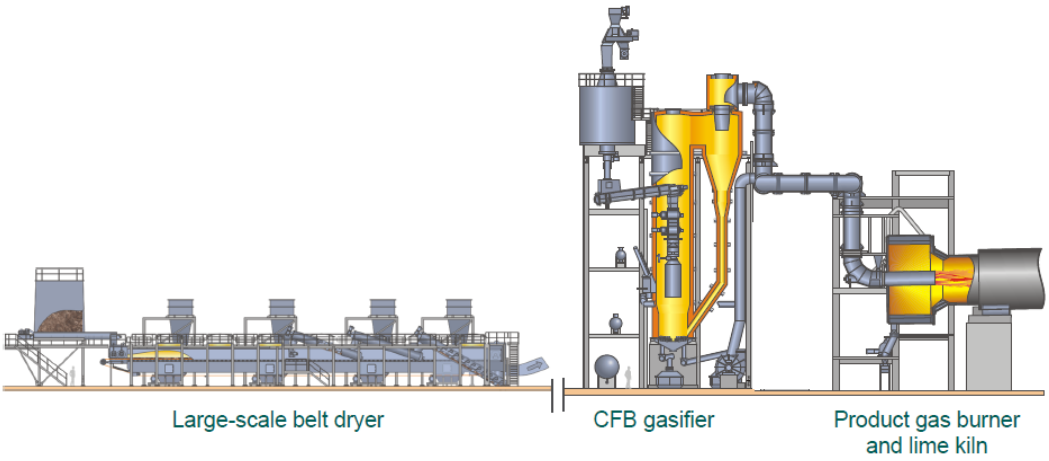

Figure 13. Overview of the lime kiln gasification system (Isaksson, 2017, 24)

The gasification process starts with the collection of wanted biomass in the wood yard. For example, the bark is commonly used in biogas production as a raw material. The bark is collected from the debarking and stored in a silo. After storage, the biomass is dried with the options of firing a side stream of the product gas or using flue gases and secondary heat

(low-pressure steam and hot water) from processes in the mill site (Huhtinen & Hotta 2008, 229). Drying of the biomass increases process efficiency and reduces the amount of flue gases (Isaksson, 2007). The most suitable gasifier for lime kiln operation is a circulating fluidized bed gasifier (CFB). The CFB gasifiers are sized better for lime kiln operation and it is more flexible with the particle size of the fed biomass. The suitable particle size for the biomass is around 6 mm, which is achieved with grinding before gasification. The moisture content of the biomass should be under 15 % to achieve efficient gasification. During a stable operation and suitable conditions, biomass moisture content can be reduced up to 5-10%. An average value for moisture content is about 10 % when efficient gasification and a lower risk of fire, are achieved during the drying process. Also, impurities, such as metals and sand should be removed from the biomass (Kuparinen 2019, 27; Vakkilainen 2018; Isaksson 2017).

A gasifier consists of cyclone, gas duct, fuel and bed material feeding, bottom ash discharge, and air supply system. The treated biomass is fed to the CFB gasifier reactor, which is at a temperature of 850-950 °C. In the reactor, some of the biomass is combusted to provide heat for a pyrolysis reaction, in which the biomass goes through the pyrolysis reaction and forms product gas. During pyrolysis, a large amount of oils and tars are generated into the product gas. (Hart 2021, 159) The preheated gasification air is fed to the gasifier from the bottom of the reactor. The cyclone in the gasifier is used to separate solids and the product gas. Solids, such as unreacted char, bed material, non-process elements (NPE), and ash are removed from the bottom of the reactor with a discharge system. The separated product gas flows to the gas duct from the top of the cyclone and the product gas is led to the lime kiln burner. (Taillon, et al. 2018, 86; Huhtinen & Hotta 2008, 230; Hamaguchi, et al. 2012, 2297)

The product gas properties vary, depending on gasification operation conditions, raw material type and properties, and whether oxygen or air is used as the gasifying agent (Francey, et al. 2009, 35). In the combustion process, fuel heating value plays a large role. According to Kuparinen & Vakkilainen (2017, 4034), the gasification gas lower heating value varies between 6-30 MJ/kg. The lower heating value of product gas sets a challenge for high enough kiln flame temperature. The adiabatic flame temperature for product gas varies from 1870 °C to about 2000 °C, which is significantly lower, compared to natural gas and fuel oil. To achieve the same hot-end temperature for the kiln, a higher firing rate is required with

product gas. Increased firing rate causes higher back-end temperature and flue gas losses. The product gas should be burned hot, right after the gasification to avoid an increase in fuel consumption. Also, the gasifier should be located near the lime kiln burner, so that the costs from the expensive refractory ducts are minimized and corrosion in the ducts is avoided. (Vakkilainen 2018; Kuparinen & Vakkilainen 2017, 4034) To achieve the required conditions for stable combustion, an additional 15 % combustion air is needed. This means that increased fan capacity is required. (Isaksson, 2007)

Considering the availability of bark, an average of 10 % of the volume of the softwood is bark, which means that the bark is largely available for mill use (Rasi, et al., 2019). In most eucalyptus cases, eucalyptus is debarked in the harvesting site, which means that in the eucalyptus mills some arrangements might be needed to increase bark availability. Also, if the mill burns bark in a power boiler for additional steam production, bark might be needed to be bought from outside the mill. Bark gasification also affects kiln availability. If problems occur in the gasification plant or drying process, it could cause an unplanned stop for the whole mill. That is why often a backup system with fossil fuel is used in case of malfunction or any other problem. The fossil fuel system as a backup system applies also to other fuels.

The combustion of product gas can cause some operational issues. Compared to fossil fuels, the kiln temperature control has a longer reaction time, which makes the stable operation more challenging. According to a study by Francey et.al. (2009), in some cases, a more stable flame and combustion have been achieved with product gas, than with fuel oil. (Francey, et al. 2009, 37) The product gas resembles fossil fuels in its other properties, which means that no major changes need to be made to the kiln burner. (Francey, et al. 2009; Mäki, et al. 2021; Hamaguchi, et al. 2012) The composition of bark and the varying quality can cause challenges in lime kiln operation and lime quality. Challenges such as gasifier bed agglomeration and stops in the drying phase can cause unwanted stops in production. Also, the high level of NPE in the bark can be carried with the gas to lime, which causes ringing in the kiln, and a decrease in the quality of lime and white liquor (Isaksson 2006). The decreased lime quality also increases make-up lime consumption. (Mäki, et al. 2021; Hamaguchi, et al. 2012; Kuparinen & Vakkilainen 2017) According to a study by Vakkilainen & Kivistö (2008), the combustion of the product gas in lime kiln increase make-

up lime demand by 8 kg per ADt of pulp, compared to heavy fuel oil. (Vakkilainen & Kivistö 2008, 29).

From an economic point of view, several factors affect the level of profitability. The cost of bark is significantly lower than natural gas and fuel oil, and with the use of the mill's raw material, significant savings can be achieved. Also, the risk of increasing fossil fuel prices can be avoided. Another major factor is the cost of investment. The gasification plant investment costs are relatively high, including storage facilities, drying plant, gasification plant, and the modification to fuel feeding system. The target country's policies and granted subsidies can make the investment more attractive and the low production of CO2 emissions increases savings from CO_2-emission costs. Additional costs are added with increased electricity and heat consumption and if the raw material delivery is outsourced. (Kuparinen 2019; Hamaguchi, et al. 2012; Mäki, et al. 2021)

3.2.2 Wood dust / Pulverized wood

The process for wood dust combustion in a lime kiln requires raw material collection, drying, grinding, and dusting prior to combustion. The raw material for wood dust is variable wood residues from wood yard operations, such as sawdust from a sawmill, bark from debarking, or too large chips left in the chip screen.

Drying the biomass is one of the requirements to achieve stable combustion. The drying equipment is typically a belt dryer. The heat can be brought to the system from flue gases from power, recovery boiler, or lime kiln. A more common and more fire-safe way is low-pressure steam or secondary heat from mill condensates. Biomass is grinded after drying and impurities, such as sand and metals are separated from the biomass. Then biomass is fed to the grinder and pulverized. The particle size is an important factor for better combustion, which should be unisize with a particle diameter of 1 mm or below. The small particle size also, makes the dust blow to the burner easier. A pulverized fuel burner is needed for dust combustion. (Kuparinen & Vakkilainen 2017, 4039-4041; Hart 2021, 152-153)

Low moisture content is an important property to improve combustion. The moisture content of the pulverized wood should be from 5-10 %. A low moisture content increases the fuel heating value and reduces flue gas volume in the kiln. (Kuparinen, et al. 2017) Wood residues consist high amount of ash, non-process elements, such as aluminum, silica,

phosphorus, and magnesium, which can be problematic in kiln combustion. The impurities gather in the lime cycle causing ring formation and increasing make-up lime demand. Also, a high level of ash may end up in the lime, which decreases lime quality. (Arpalahti, et al. 2008, 153-154; Kuparinen, et al. 2017) Compared to fossil fuels, the pulverized wood has a lower adiabatic flame temperature and heating value. The adiabatic flame temperature is 1950 °C and the lower heating value varies between 14-19 MJ/kg, depending on wood and residue type. (Alakangas, et al. 2015, 86; Kuparinen, et al. 2017)

Higher moisture content, lower adiabatic flame temperature, and lower heating value compared to fossil fuels, increase the required firing rate. This also decreases system efficiency, which leads to more flue gases are produced in the combustion process, and flue gas exit temperature increases (Kuparinen, et al. 2017). The impurities in wood dust make lime kiln and causticizing plant operation more difficult. The phosphorus in the fuel only reacts when it reaches the causticizing plant, which increases dead load lime that must be removed from the lime cycle. This increases the need for make-up lime. The combustion of wood residues has proven to have effects on lime quality and increasing ringing problems in the lime kiln. Also, the raw material treatment requires much more energy, than traditional fuels, and clogging of hammer mill and fuel feed system can cause unplanned system downtime. The dry wood dust accumulated in the system increases the risk of fire or dust explosion. (Kuparinen, et al. 2017; Hart 2021, 153; Francey, et al. 2009, 37)

3.2.3 Lignin

The market and potential for the use of lignin have increased globally. The lignin is dissolved in the cooking process from the wood chips to form black liquor with other organic substances. Lignin separation from the black liquor has become more attractive with the concept of bio-product mills, where all the side streams and potential bioproducts are considered. Lignin contains a large part of the energy contained in black liquor, which means that the strong black liquor, which is combusted in the recovery boiler does not contain as much energy. However, the extraction in modern pulp mills is possible, because of the self-sufficiency in terms of energy production. If the mill is energy self-sufficient and the mill's bottleneck is the recovery boiler, lignin extraction can be considered as one option. The lignin separation can enable more pulp production capacity and it can be used as a fuel in the lime kiln. The decreased heat load means that more black liquor can be combusted, which allows

for an increase in white liquor production. In the case of lignin extraction from black liquor, the amount extracted lignin must be examined on a case-by-case basis. The amount of lignin separated must be examined concerning the mill raw materials, load, and operations of energy production and recovery boiler. (Wallmo, et al. 2017, 2)

There are several different technologies for lignin separation in a pulp mill, but in this thesis, the acid precipitation technology was selected, because it is currently available on an industrial scale and is considered to be the most developed one (Kihlman 2021, 40). The LignoBoost method is a commercial precipitation process for lignin removal based on filtration and it is used for example, at Stora Enso's Sunila mill, where it is also used as a fuel in the lime kiln. Black liquor with 30-40 % dry solid content is led to the lignin plant into a precipitation vessel. Lignin is separated via the acidification process with sulfuric acid and carbon dioxide. Carbon dioxide gas is injected into the precipitation vessel, which lowers the pH. After precipitation, solid lignin is filtrated from black liquor in steps. The filtrated lignin cake contains also carryover black liquor. After filtration, the slurry is fed to re-suspension, where the lignin slurry is mixed with washing water and sulfuric acid (H_2SO_4). This lowers mixture pH to 2,5-4. The slurry matures in re-suspension and after maturing, the slurry is dewatered and washed with acidic wash water, which is pressed through the lignin cake to wash impurities and carryover black liquor from lignin. All the streams in the LignoBoost process are recovered back to the chemical recovery process. The LignoBoost process is presented in Figure 14 (Wallmo et al., 2017; Hamaguchi & Vakkilainen 2010 & Vakkilainen & Kivistö 2008)

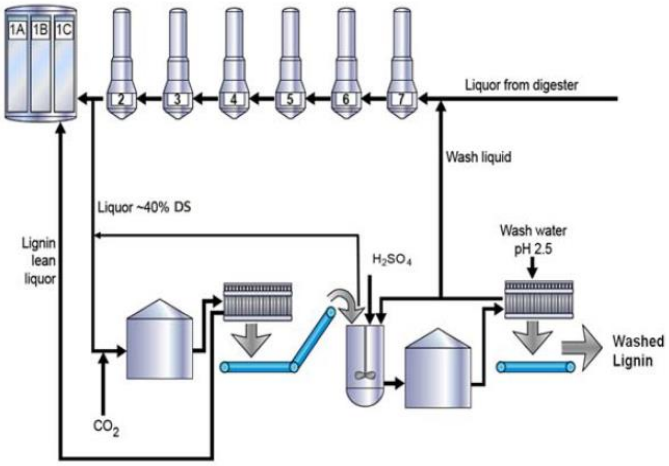

Figure 14. LignoBoost process overview (Wallmo, et al. 2017)

Before lignin combustion in the lime kiln, the lignin is dried to a moisture content below 10 % and pulverized. The pulverized dry lignin needs to be stored and conveyed to the kiln in an environment with a low oxygen level to prevent dust explosion (Hart 2021, 149).

When considering fuel properties, lignin's lower heating value is around 23-26,5 MJ/kg, which is higher than many other biomass-based fuel options. Also, the adiabatic flame temperature is 1980 °C, which is close to natural gas adiabatic flame temperature. According to Tomani, et.al. (2011), experience in mill demonstrates has shown that firing conditions are relatively close to natural gas firing. Also, operation control has been stated to be easy (Tomani, et al., 2011, 539).

From an operational point of view, the recovery boiler load should be considered accurate. To replace fossil fuels in a lime kiln, the recovery boiler should be the mill's bottleneck to achieve profitability economically as well as operationally. However, too high lignin removal rate can affect recovery boiler operation negatively. The lignin extraction affects the boiler operation with decreased black liquor heating value, recovery boiler heat load, superheating temperature, and lower furnace temperature. Hamaguchi and Vakkilainen (2010) state, that the removal rate limit for good and controllable operation is below 30 %. (Hamaguchi & Vakkilainen 2010) Lignin consists of a relatively high level of sodium and sulphur (1-3%), which could affect lime kiln operation with ring and ball formation. According to Hamaguchi et.al. (2011), if the lignin washing is done properly before combustion, the ring and ball formation should not be a major issue. The ring formation has been attempted to control by using high sodium salt concentrations in the lignin wash water. With lignin firing lime kiln, the make-up lime consumption increases by 1 kg per ADt of pulp, compared to heavy fuel oil (Vakkilainen & Kivistö 2008, 29).

Variable lignin quality can cause instability of kiln performance. With proper lignin preparation, good quality lignin can be achieved, which helps to achieve stable combustion. With powdered fuel, challenges occur in storing and conveying. Dry powdered fuel increases the risk of dust explosion and blockages in the storage, conveying and feeder systems can occur. (Tomani 2010, 56)

From an availability point of view, lignin is present in every mill. More lignin is available at the softwood-based mills than at the hardwood-based. In the case of eucalyptus, lignin

content is very close to softwood. In Table 2, the lignin contents for different wood species are presented. In Kraft pulping process, the yield of available lignin varies from 340-510 kg/ADt pulp (Mäki, et al. 2021, 5).

Table 2. The lignin content of different wood species (Gellerstedt, et al. 2013, 181; Hamaguchi, et al. 2012, 2291)

Wood specie	[% on wood]
Pine	28,2 %
Spruce	27,2 %
Urograndis	26,8 %
Grandis	26,1 %

From an economic perspective, a mill whose production is limited by a recovery boiler can achieve economic benefits by extracting lignin and increasing pulp production. (Manning & Honghi 2015, 478)

3.2.4 Tall oil

Tall oil is a by-product of pulping, which is produced from tall oil soap by acidulation. Crude tall oil is usually sold to the chemical industry, for the production of chemicals, and transportation biofuels, but some mills use it as a fuel for the lime kiln or recovery boiler. For example, in Northern Europe, tall oil is a very valuable product and, so it is not utilized for energy use, but in other places tall oil's value might be lower and therefore used for energy production.

Soap is extracted from pulpwood, mostly consisting of resin and fatty acids. Alkaline conditions in digester saponify resin and acids to crude tall oil soap. The soap dissolves to weak black liquor and in the evaporation plant, soap accumulates on the surface of black liquor. The soap is skimmed from the surface of the weak and intermediate black liquor tank. From a process point of view, soap separation is important to prevent soap foaming in the evaporation plant and to achieve stable combustion in the recovery boiler. Crude tall oil is yielded from the soap, using sulfuric acid to liberate resin and fatty acids. (Suhr, et al. 2015, 204; Alén, 2000, 74; Laxén & Tikka 2008, 360) A traditional one-stage tall oil soap acidulation is divided into four steps, soap heating, acid charge into soap, separation of other substances (brine, lignin, and calcium), and drying the tall oil. The separation occurs in a decanter,

where substances form their own layers, because of different densities. In the top layer, crude tall oil is pumped to a CTO storage tank and the other substances are brought back to different phases of the evaporation process. (Laxén & Tikka 2008, 372-374)

Crude tall oil has very similar combustion properties in the lime kiln with fuel oil. Tall oil has a very high lower heating value, which is around 35 MJ/kg. The adiabatic flame temperature is about 2200 °C, which is high and very similar to the fuel oil. In the literature, it is stated that the combustion and the heat transfer of flame and lime bed match with the fuel oil. (Arpalahti, et al. 2008, 176; Manning & Honghi 2015, 476)

One of the big questions for tall oil combustion in the lime kiln is the availability of the fuel. A typical yield is about 30-50 kg of tall oil/ADt of pulp (Laxén & Tikka 2008; Alén 2000, 74). The yield depends on the wood species, age of the wood, time of the year, and wood yard storage practices. Considering the wood species, softwood (pines) pulping produces tall oil with the highest yield. Tall oil is available also, with other softwoods, but in much less quantity and lower quality. The age of the wood and the length of the storage time affect the quantity of tall oil. If the wood is stored for a long time the tall oil loss increases significantly. (Foran 2005, 3.7-2; Peters & Stojcheva 2017, 6) Also, challenges from an availability point of view could occur if the lime kiln energy demand is based totally on tall oil production, because of the varying yield and quality. Even though tall oil is an important side stream for a pulp mill, the mill is not operated to optimize tall oil production, which will affect the reliability of the tall oil fuel in the lime kiln (Peters & Stojcheva, 2017).

The highly acidic nature of tall oil sets up for requirement for equipment and storage tanks, to achieve secure and stable operation. The equipment, such as storage tanks, conveying lines, and kiln burner must be designed to withstand the acidic environment and increased maintenance might be needed. Especially, in the kiln burner where the acidic nature is presented with high temperature, increased attention is needed. (Hart 2021, 145) Also, from an operational point of view, the varying quality, and properties can cause kiln overheating and refractory damage, which will increase the operating costs and need for maintenance. The changes in viscosity can also, lead to poor fuel atomization in the burner, which might lead to ring formation problems (Manning & Honghi 2015, 476).

From an economic point of view in Finland, the combustion of tall oil in the lime kiln is not profitable at all. In Finland, the use of tall oil for industrial heating purposes is an activity under the excise duty act. Thus, tall oil is a fuel subject to excise duty, even if it is used for purposes that justify the use of duty-free fuel, such as a lime kiln. (Verohallinto, 2021) Also, in Finland, the sales value of tall oil is so high that it is not profitable to burn it. Tall oil is important revenue for the mills, and the tall oil firing should be considered accurately (Hart 2020A, 264).

3.2.5 Tall oil pitch

Tall oil pitch is a by-product of crude tall oil distillation to biodiesel. The tall oil produced in the pulp mill is not often used as it is. Typically, it is sold to biorefineries, that produce biodiesel from fatty acids in tall oil. The tall oil is distilled into different components in a fractionation column. The components are tall oil heads, tall oil fatty acids (TOFA), distilled tall oil (DTO), tall oil rosin (TOR), and tall oil pitch (TOP) (Laxén & Tikka 2008, 379). The tall oil pitch is used as a fuel, because of its high heating value. For example, Metsä Fibre Rauma is burning TOP in the lime kiln (Suhr, et al. 2015, 345). The crude tall oil produced in the mill is sent to the Forchem tall oil distillery next to the mill, from where, after refining, the pitch oil is returned to Metsä Fibre mill site for combustion (Aho, et al. 2013, 52). In Europe, an average share of TOP in CTO is 28 % depending on tall oil quality, and in the US 16 % (Alén 2000, 74; Cashman, et al. 2015, 1111).

Tall oil pitch properties are favorable for being an additional fuel for the lime kiln. Pitch oil's lower heating value varies around 30-40 MJ/kg (Aluehallintovirasto 2019, 29), which is almost as high as fuel oil. The adiabatic flame temperature for tall oil pitch is about 1950 °C, which is about 200 °C lower than fuel oil (Hart 2021, 143). As mentioned, tall oil pitch is generated from the crude tall oil in the distillation process, so it will be available for lime kiln combustion in areas, where a tall oil refinery is located near the mill site. If the biorefinery is inside the mill, it will be easy to transport for kiln use.

3.3 Other alternatives

The other alternative fuels include mill’s minor by-products and side streams. In this chapter, other alternatives methanol, hydrogen, turpentine, and non-condensable gases are reviewed. In the case of these other alternatives, by-products such as methanol and turpentine are

available for sale as such, but they can also be used as fuel. Hydrogen is rarely used technology for lime kiln use, because of its difficult handling and exceptional properties, but it has been proven to work in a lime kiln at least to a certain extent. Lime kilns have been used for the disposal of non-condensable gases, but the current trend has been declining and the trend has moved toward recovery boiler. Despite these things, mentioned options are considered in this section as with other fuel options in previous chapters.

3.3.1 Methanol

Methanol is a pulp mill by-product, which is produced during the cooking process, in which it binds to the weak black liquor. In the evaporation plant, the methanol condenses to foul condensates and it is purified with a condensate treatment system. The most commonly used condensate treatment method is steam stripping. The condensate treatment occurs in the stripping column, where condensates flow counter current to live steam or vapor. Methanol is released from the foul condensate into a stripper-off gas (SOG). The steam stripping enables treated condensates to be reused for example in brown stock washing and causticizing plant. To recover the methanol from SOG, the gas is sent to the liquid methanol system. This process decreases SOG moisture content from 65 % to 20 %. Gas from stripper and steam are injected into the lower part of a vessel. There is a cooling water circulation in the upper part of the vessel. The injected steam separates methanol from the gas in the lower part and then it flows through the upper part cooling circulation. The cooling water in the upper part maintains a temperature, which is below the condensation point of water but above the boiling point of methanol. The vapor containing methanol, flow to through a separate cooler vessel in which the vapor and methanol mixture condenses to liquid with an 80 % concentration of methanol. (Suhr, et al. 2015; Valmet 2018, 2-5)

Liquid methanol has an effective heating value of 21 MJ/kg, which is higher than most biomass-based fuels. Compared to traditional fossil fuels, methanol has about half of the energy content of fossil fuels. Methanol adiabatic flame temperature varies between 2100-2200 °C, which is very close to heavy fuel oil. From a properties point of view, liquid methanol can be combusted in a lime kiln to replace a partial amount of fossil fuels or as a support fuel for biofuel option. With similar combustion properties, only small changes to the kiln equipment are required. (Arpalahti, et al. 2008, 176)

The technology for a liquid methanol treatment system is relatively new, and therefore little information is available on its utilization in the mill environment. Nevertheless, the literature states that methanol can be used as additional fuel to replace some of the fossil fuel consumption. Also, a typical mill does not produce enough methanol to cover the whole energy demand of the lime kiln. A typical methanol recovery process produces about 10-15 % of the kiln total heat input (Hart 2021, 147). The amount of methanol produced varies with the wood type used for pulping from 7 to 15 kg methanol per ADt pulp. Pulping with hardwood produces about 10-15 kg of methanol per ADt of pulp and with softwood about 7-10 kg of methanol per ADt of pulp (Valmet 2018, 2; Zhu, et al. 2000).

To enable liquid methanol combustion in the lime kiln, significant changes in combustion equipment are not needed. The burner is designed with the same similar principle, but a separate nozzle for methanol is needed. The nozzle is required for proper atomization of the liquid methanol, to achieve stable flame and combustion. Also, good filtration is necessary before the burner nozzle to avoid blockage. (Arpalahti, et al. 2008, 176)

According to European Commission, a stripper system for a 1500 Adt/d mill, the investment is about 2-2,5 million euros (Suhr, et al. 2015, 276). From an economic perspective, the removal of methanol decreases the operational costs of an effluent treatment plant. It also decreases water consumption and potential fossil fuel costs. (Valmet, 2018) In Finland, synthetically produced methanol is tax-free, when it is used for electricity production or first use. Methanol produced as a by-product of the forest industry is considered non-synthetic and is classified as biofuel oil, so therefore it is under taxation. (Verohallinto 2021)

3.3.2 Hydrogen

The alkaline electrolysis process is currently the most developed industrial hydrogen production method. The basic electrolysis reaction is presented below.

$$2H_2O\ (l) + \text{energy} \rightarrow 2H_2\ (g) + O_2\ (g) + \text{heat} \qquad (8)$$

The function of a single electrolysis cell is presented in Figure 15. The cell consists of a liquid electrolyte, separator, and two electrodes, anode, and cathode. The electrolyte is 20-40 wt% aqueous potassium hydroxide (KOH) and the separator is a microporous diaphragm. The material used for electrodes is typically nickel. A current is applied to the electrolyzer

cell when in anode and cathode different reactions occur. In the cathode, water decomposes into hydrogen and hydroxide ions, and in the anode, hydroxide decomposes to oxygen and water. (Lehner, et al. 2014, 24-25; Zeng & Zhang 2011, 309-310)

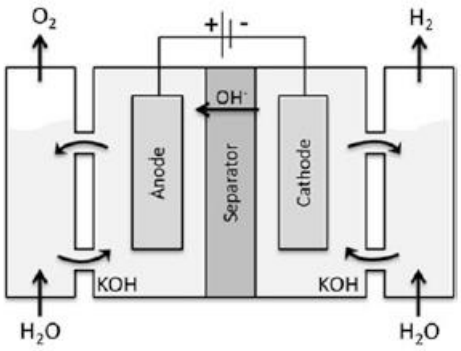

Figure 15. Alkaline electrolysis cell principle (Lehner, et al. 2014, 25)

In industrial applications, single electrolysis cells are stacked in parallel or series. In terms of power consumption, electrolysis application varies from kW to hundreds of MW. (Zeng & Zhang 2011, 319) Modern pulp mills are self-sufficient in energy production, which enables hydrogen production in pulp mills, without the purchase of electricity from external electricity producers. Hydrogen production decreases the availability of electricity sold to the grid. The additional revenue loss from electricity sales is compensated with the saving in fossil fuel use in the lime kiln and oxygen production in the mill site. (Lehner, et al. 2014, 25) Generally, a pulp mill water pretreatment plant can be assumed to be suitable for the electrolyzing process (Kuparinen 2019, 34).

Hydrogen product gas heating value is about 120 MJ/kg, which is very high compared to traditional fuels and the adiabatic flame temperature is 2210 °C. Hydrogen combustion produces a very small amount of emissions. (Kuparinen & Vakkilainen, 2017) The density of hydrogen is very low, which means that the volume in combustion needs to be high. Although hydrogen has a high lower heating value, its low density causes the hydrogen to have a low energy density (MJ/m^3). As a lime kiln fuel, the feed volume of hydrogen to the burner needs to be high. Very low density and high flammability also cause operational issues for storing and conveying (Boudellal 2018, 61; Hart 2020B, 277).

From an availability point of view, the most important factor is the mill's energy self-sufficiency. Electrolysis requires electricity and treated water. To achieve more profitable

electrolysis electricity should be brought to the process from the pulp mill's excess electricity. If the level of self-sufficiency is not enough, the rest of the electricity could be bought from the national grid, which is more expensive and so, less profitable. Also, treated water is available in mill sites and existing water treatment technology is adequate for the electrolysis process (Hart 2021, 164). In the electrolysis process, hydrogen is the main reaction product, but also oxygen is produced as a side product. This can be beneficial for pulp mills because oxygen is used in the oxygen delignification process and pulp bleaching. The oxygen produced by electrolysis could be used to replace oxygen produced by the separate oxygen plant. (Kuparinen & Vakkilainen, 2017). It is also possible that hydrogen is brought to the plant externally produced. This kind of solution is utilized in Stora Enso Oulu mill, where the lime kiln uses mainly hydrogen and pitch oil as fuel. The hydrogen is brought to the mill site from the nearby chemical plant, where it is produced as a by-product. (Kuparinen 2019, 34)

The properties of hydrogen make storing challenging. The low-density gas requires airtight storage and the high flammability increases the risk of explosion. To minimize the risk of storage issues, the integration of lime kiln and hydrogen production should be online. An online system makes the process simpler and safer. The hydrogen production facilities should be near the kiln to prevent the risk of explosion, gas leaks, and gas duct costs. (Kuparinen, 2019) The hydrogen feed to the kiln requires a specific valve and injection system to the burner. Because of the high adiabatic flame temperature and flammability, high exit velocity in the burner for hydrogen is needed to avoid burner tip overheating. Due to high flame temperature, kiln refractory lining needs to be a more durable material against heat than with other fuels and also, the additional water use required for electrolysis increases the mill's water treatment plant load. (Hart 2021, 164)

From an economic point of view, hydrogen production requires big investments for electrolysis plant, storage, and conveying system. Alkaline electrolysis is a relatively new and developing technology, and the price for the investment is high. At the moment, with the current electricity prices, hydrogen does not seem to be profitable at this scale. In the future, as technology develops and becomes more widespread, the investment costs of hydrogen production may decrease, thus increasing the economic viability of the technology. Also, if the

price of electricity falls or a fair rise in fossil fuel prices can increase the economic profitability of hydrogen (Kuparinen & Vakkilainen 2017, 4044).

3.3.3 Turpentine

In softwood pulping, the cooking process evaporates terpenes from wood, which are volatile agents in pitch. The terpenes are collected from cooking exhaust gases and other gases to be recovered as turpentine. The recovery is done with steam condensation. A surface condensation separates turpentine from the non-condensable gases. The turpentine and condensation water is discharged into a decanter, where water and turpentine form their own layers, because of a density difference between these substances. Turpentine is then collected from the decanter with an overflow. (KnowPulp 2021; Smook 2016, 163-164)

From an availability perspective, only softwood consists of terpenes. When considering the availability of softwood turpentine, pine trees contain more turpentine than spruce trees. The amount of turpentine in the pulpwood depends on wood growth conditions, wood chip storage methods, wood age, and duration and temperature of wood chip presteaming before digester. (KnowPulp 2021) A typical yield for pinewood turpentine extraction varies in 2-15 kg of turpentine per ADt of pulp and spruce trees yield varies 2-3 kg of turpentine per ADt of pulp. (Suhr, et.al. 2015, 204; KnowPulp 2021) In normal operation conditions, turpentine can cover about 10 % of the kiln total heat input, and turpentine is fired with a multifuel burner through the main nozzle (Hart 2021, 148).

3.3.4 Non-condensable gases

In Kraft pulping process, non-condensable gases (NCG) are released in processes where black liquor is handled. These harmful gases have an odour, which is hazardous for humans and the environment. NCG can be disposed of in two ways, thermal oxidation, or absorption with scrubbers.

Non-condensable gases are divided into concentrated malodorous gases (CNCG) and diluted malodorous gases (DNCG), according to sulphur content. CNCG are originated in the cooking phase, evaporation plant, and stripping of foul condensates with a sulphur concentration over 5 g/Nm^3 (Suhr, et.al. 2015, 244). The CNCG can be combusted in a recovery boiler and lime kiln. The current trend for CNCG combustion has changed towards preferring recovery

boiler for disposal. The reason for this current trend is operational problems, with ring formation and sulphur emissions in a lime kiln, when burning CNCG (KnowPulp 2021). The dilute malodorous gases are collected from different phases of fiber line and chemical recovery from black liquor tanks, brown stock washing, screening equipment, and evaporation, tall oil, and causticizing plants with a sulphur concentration under 0,5 g/Nm^3. (Suhr, et.al. 2015, 244-245)

The NCG is hazardous for the environment and people if the gases are not properly handled. Attention must be paid to the design and operation of NCG handling systems. Gas leaks are dangerous, and leaks must be avoided. The toxic hydrogen sulphide in gases can be fatal for humans. The NCG requires careful handling and any equipment that can cause sparks and the presence of oxygen must be avoided because of high flammability and explosiveness (Lin, 2005, 3.6-8).

From an availability perspective, the concentrated malodorous gases are available in low volume with a high sulphur concentration and the dilute malodorous gases in higher volume with a lower sulphur concentration. The higher volume is because DNCG contains for the most part air and steam.

A common way of NCG disposal is combustion in recovery boiler and lime kiln or scrubbing the NCG with a spray tower or packed column. On average, malodorous gases can be used to replace 15 % of the fossil fuel consumption in the lime kiln (Suhr, et.al. 2015, 244; KnowPulp 2021) The 15 % replacement limit has been proven to be maximum to prevent temperature fluctuation in kiln flame, bed material and refractory lining in flame area. Co-combustion of NCG with other fuels over this limit, also decreases the main fuel heating value and main fuel volumetric flow, because of the level of moisture in NCG. The dilute malodorous gases are typically injected into the kiln as a mixture with primary combustion air. (Arpalahti, et al. 2008, 176). According to Lin (2005), it has been found beneficial to scrub NCG with white liquor before combustion in a lime kiln to reduce sulphur components from the NCG stream. (Lin 2005, 3.6-6-8) This action prevents ring and ball formation and reduces sulphur-emissions, but in any case, when NCG are burned in the lime kiln, the kiln's TRS and NO_x-emissions increase (Tran, et al. 2004)

3.4 Summary of the fuel options

The popularity of fossil fuels, heavy fuel oil, and natural gas is based on fuel availability, security, and ease of operation. Forest companies have started looking for alternative fuel options for fossil fuels, whose prices are rising steadily over time. The biggest drivers for the change are economic benefits and reduced CO2 emission. These alternative fuel options have been embraced from pulp mill wood residues and by-products.

Wood residues in a pulp mill are generated during different wood handling processes, for example, debarking and chipping. Wood-based fuels, gasification product gas, and wood dust have been used as the main fuel to replace fossil fuels entirely. To have a successful operation with these fuels, biomass drying, and handling are important steps to achieve it. Because of the high moisture content, biomass requires drying to moisture content as low as possible to achieve more efficient combustion and higher heating value. The properties of wood fuel vary from traditional fossil fuel, which sets requirements for combustion. A higher firing rate is needed with wooden fuels, because of the lower heating value and adiabatic flame temperature. This increases flue gas exit temperature, heat losses, fuel consumption, and the need for fan capacity. Also, the amount of non-process elements and impurities in the lime kiln increases, which causes a higher risk of ring formation. The ring and ball formation is prevented by opening the lime cycle and removing the deadload lime, which means that the consumption of make-up lime increases. This issue is faced with all the alternative fuel options. From an economic point of view, economic benefits are achieved, but wood fuel requires investments for fuel treatment, such as drying equipment, storage capacity for biomass, and gasifier in case of gasification. Also, in the case of eucalyptus mills, where wood residue availability is lower, the wood handling department, might require changes.

The pulping by-products are extracted from different side streams during different phases of pulping process and chemical recovery. Lignin is a pulping by-product, that has the potential of completely replacing fossil fuel. Often, the other by-product fuels, tall oil, tall oil pitch, turpentine, methanol, and hydrogen are used to cover some of the kiln's energy demand, but not completely. The lignin can be extracted from the black liquor to cover the whole kiln heat demand, but its profitability should be considered especially in a case where the recovery boiler is a bottleneck in the mill. When the recovery boiler works as the bottleneck, the lignin separation allows additional pulp production, which adds additional revenue with

increased production and savings from covering fossil fuel costs. The lignin separation plant requires investments and pretreatment before combustion, which increases the capital costs, but it has been stated that good performance has been achieved with lignin combustion. In the case of the other by-products, the fuels are not available at a level where they could be used to cover the whole kiln energy demand, or the sales value of the product is higher than the value in use. Also, some of these fuels increase operational issues, when used on large scale.

4 CASE STUDIES

The aim of this thesis is to calculate energy balances for different lime kiln fuel cases and to examine the effects of the fuel on the mill's energy balance. For the calculation of energy balances, the fiber and chemical balances are solved using the pulp mills' main fiber and chemical balance tool in Excel. The main dimension balances are calculated for softwood pulp mills and hardwood eucalyptus pulp mills. The softwood pulp mills are referred to as Mill A and, the eucalyptus pulp mills as Mill B. The results are calculated with the main balance dimensioning- and energy balance tool. The main balances are calculated to determine the capacities of the mill's department capacities and the available fuel flows within the mill site. Department capacities and flue flows are used to solve heat balance and electricity consumption for each case, using known reference values for department-specific heat and power consumption and heat generation.

In the case of a Northern Bleached Softwood Kraft mill, two main balances are formed, for the base case with an oil-fired lime kiln, and a mill with a gasification process, bark product gas firing lime kiln. The energy balance cases are presented below. A total of three energy balances are calculated for softwood cases.

In the case of a Southern Bleached Hardwood mill, four main balances are formed, for the base with an oil-fired lime kiln, two different gasification options with product gas firing lime kiln, and for a lignin firing lime kiln, in which lignin is separated from black liquor in evaporation plant. The energy balances are calculated for each of the four main balances. More accurate case descriptions are presented below.

Northern softwood cases:

Mill A1:

- Lime kiln fired with heavy fuel oil
- All the bark, wood rejects, and sludge are burned in the bark boiler

Mill A2:

- Lime kiln fired with bark product gas
- A required amount of bark is gasified into product gas to cover the lime kiln heat demand

a. Rest of the bark, wood rejects, and sludge are burned in the bark boiler
b. Rest of the bark and all the wood rejects are sold as such

Southern hardwood cases:

Mill B1:
- Lime kiln fired with heavy fuel oil
- All the bark, wood rejects, and sludge are burned in a bark boiler

Mill B2:
- Lime kiln fired with product gas
- No bark boiler

a. All the bark, wood rejects, and additional bark from site debarking is gasified into product gas
b. All the bark, wood rejects, and additional eucalyptus wood chips are gasified into product gas

Mill B3:
- Lime kiln is fired with lignin, which is separated from the black liquor in the evaporation plant
- All the bark, wood rejects, and sludge are burned in a bark boiler

4.1 Initial values for main balances

The main dimension balance is calculated with the Microsoft Excel tool, which calculates the mill's fiber and chemical balances using the initial values given. The initial values are based on reference data from existing mills and projects, but the values are slightly modified and rounded to fit the cases. The decided initial value for wood processing and characteristics are presented in Table 3 for both mills. Further initial values for each main fiber and chemical balance are presented in the appendices.

Table 3. Initial values for wood processing and characteristics for both softwood and hardwood mills

	Unit	Mill A	Mill B
Wood processing			
Wood total losses	V-%	2,7	2,3
Wood characteristics			
Wood moisture	w-%	50	33
Bark moisture	w-%	60	33
Wood density	kgBD/m^3sub	415	525
Bark density	kgBD/m^3sub	315	330
Bark content, unbarked wood	V-%	11	10

The pulping process initial values for Mill A's main dimensions are presented in Table 4. The table shows the more relevant initial values for the main balance sheets for each process section. For each case, the annual pulp production capacity is 1000 000 ADt of bleached pulp. The mills are assumed to be stand-alone pulp mills, without a paper mill.

Table 4. The initial value for northern softwood mills

	Unit	Mill A1	Mill A2
Production			
Annual production	ADt/a	1000 000	1000 000
Operating days	d/a	355	355
Unbarked wood	%	100	100
Cooking			
Kappa number		32	32
Cooking yield	%	47	47
Oxygen delignification			
Kappa number		15	15
Alkali charge, OWL	kg/ADt	27	27
Oxygen charge	kg/ADt	25	25
Evaporation plant			
Dry solids concentration, black liquor out	%	83	83
White liquor plant			
Lime availability	%	80	77

The values for both cases of Mill A are the same, but the lime availability is higher when the kiln is firing oil, compared to product gas firing, when the lime availability is set to 77 %. The reason for lower lime availability is fuel impurities and worse combustion conditions for calcination. The product gas includes a higher amount of impurities, which are transferred from the fuel to the lime kiln, which causes more dead lime and cycle opening. Also, the burner flame temperature is lower, and temperature fluctuations are greater due to changing fuel quality and these things cause weaker heat transfer between the flame and the lime bed.

The initial values for Case Mill B are presented in Table 5. The initial values are based on reference eucalyptus pulp mill. In the case of gasification mill B2, the initial values are the same between cases, except for the amount of unbarked and forest debarked wood entering the mill. The figures are presented in the same cell. At mill B2a, the ratio of unbarked and forest debarked wood is 52/48, while with mill B2b, all the wood arriving at the mill is forest debarked.

Table 5. The initial values for southern eucalyptus mills

	Unit	Mill B1	Mill B2 a/b	Mill B3
Production				
Annual production	ADt/a	1000 000	1000 000	1000 000
Operating days	d/a	355	355	355
Unbarked wood	%	0	52/0	0
Forest debarked wood	%	100	48/100	100
Cooking				
Kappa number		18	18	18
Cooking yield	%	53	53	53
Oxygen delignification				
Kappa number		10,5	10,5	10,5
Alkali charge, OWL	kg/ADt	22	22	22
Oxygen charge	kg/ADt	18	18	18
Evaporation plant				
Dry solids concentration, black liquor out	%	80	80	80
Lignin separation	kg/ADt	0	0	70
White liquor plant				
Lime availability	%	80	77	77

In the base case, the wood to the mill has been assumed to be forest debarked. Also, the lime availability is set to 80 % with oil firing and 77 % for gasification and lignin. The same things are valid with mill B cases as for mill A, in terms of kiln combustion conditions and fuel impurities, which are the reason for lower lime availability for product gas and lignin.

4.2 Main balance results

The results of the northern softwood mill's main dimensions are presented in Table 6. In northern softwood mills, logs are debarked at the mill site. For gasification raw material, there is typically a surplus amount of bark. The required amount of bark for gasification is calculated in the energy balances to cover the whole heat demand of the lime kiln. Further results of the main balances are presented in the appendix.

Table 6. The main balance results for northern softwood mills

		Mill A1		Mill A2	
	Unit	Average	Design	Average	Design
Chipping	m^3sub/h	569	759	569	759
Bark	BDt/d	455		455	
Wood residues	BDt/d	149		149	
Cooking	ADt/d	2985	3317	2985	3317
Brownstock	ADt/d	2874	3194	2874	3194
Bleaching	ADt/d	2817	3130	2817	3130
Drying	ADt/d	2817	3314	2817	3314
Evaporation	tH_2O/h	1025	1191	1025	1191
Recovery boiler	tDS/d	5043	5603	5043	5603
Causticizing	m^3WL/d	11341	12601	11341	12601
Lime kiln	tCaO/d	844	938	877	974
Bark boiler	BDt/d	644	716	358	397

As mentioned, with lower lime availability, the case mill A2 with product gas firing lime kiln, the required lime kiln capacity is higher. Also, the bark boiler capacity varies between cases, as in base case mill A1, all the bark and wood residues are burned in the bark boiler. In mill A2 options, both options require 286 BDt/d of bark for gasifier and in option mill A2a, 358 BDt/d is burned in a bark boiler, and in option mill A2b, the mill does not include bark boiler, and the bark and wood residues are sold.

The main balances of case mill B are presented in Table 7. In the case of mill B results, the two different gasification cases with additional bark and additional wood fiber, the results vary a bit. The amount of bark and wooden residues in the base case does not cover the

required amount of raw material consumption for gasification and lime kiln heat demand, so additional raw materials have to be brought into the systems.

Table 7. The main balance results for southern eucalyptus mills

		Mill B1		Mill B2 a / b		Mill B3	
	Unit	Average	Design	Average	Design	Average	Design
Chipping	m^3sub/h	408	647	408/418	647/662	408	647
Bark	BDt/d	75		210/75		75	
Wood residues	BDt/d	67		67		67	
Additional wood	BDt/d	0		0/122		0	
Cooking	ADt/d	3004	3338	3004	3338	3004	3338
Brownstock	ADt/d	2896	3218	2896	3218	2896	3218
Bleaching	ADt/d	2824	3137	2824	3137	2824	3137
Drying	ADt/d	2817	3238	2817	3238	2817	3238
Evaporation	tH_2O/h	784	949	784	949	805	975
Lignin separation	t/d	0	0	0	0	197	232
Recovery boiler	tDS/d	4038	4487	4038	4487	3852	4280
Causticizing	m^3WL/d	8550	9500	8550	9500	8550	9500
Lime kiln	tCaO/d	681	756	707	786	707	786
Bark boiler	BDt/d	182	202	0	0	182	202

In the bark gasification case mill B2a, the required amount of unbarked wood to the mill is calculated to be 52 % of the received wood. The amount of incoming additional bark in 52 % of unbarked wood meets the need for gasification raw material and lime kiln heat demand. In the case of additional wood gasification case mill B2b, 100 % of the wood is forest debarked, and the additional 122 BDt/d of eucalyptus wood is calculated and added to the wood consumption to cover the raw material need for gasification and so lime kiln heat demand. In the case of mill B3, 70 kg of lignin is separated from black liquor per ADt of pulp, which means an average of 197 tons of lignin per day.

As a result of different gasification raw materials for mill B2 options, the chipping capacity, bark from the wood varies. In mill B2a, the amount of bark is higher than in other cases, because of the 52 % unbarked wood arriving in the mill. In the mill B2b option, chipping capacity is higher, because additional wood is chipped to cover the gasification raw material demand. With the lower lime availability in gasification options and lignin firing option, the required lime kiln capacities are higher, than with heavy fuel oil.

The elevation in evaporation capacity in lignin case mill B3 is due to black liquor viscosity reduction, which is affected by lignin extraction. The decreased viscosity increases evaporator heat transfer, and as a result evaporation capacity is increased. (Kihlman 2021, 16) The separation of lignin also reduces the amount of organic matter in the black liquor burned in the recovery boiler, which leads to a reduction in the capacity requirement of the recovery boiler.

5 CASE MILL ENERGY BALANCES

The aim of calculating energy balances is to compare the effects of lime kiln fuel alternatives on the case mills' energy balance. The energy balances are calculated using the energy balance tool in Microsoft Excel and WinSteam Excel add-in, which is a steam property calculator. In this chapter, important initial values for energy balance calculations and results are presented. The results include steam and power consumption and generation, as well as the availability of surplus electricity.

5.1 Fuel properties and lime kiln specific heat demand

The calculation of fuel consumptions for each case requires, that lower heating values and specific lime kiln heat demand are decided for each fuel. In the case of heavy fuel oil and lignin, lower heating value is based on literature. Values for both wood species and barks are selected based on an online database called Phyllis (Phyllis 2021), which has information on the composition and properties of different biomass. For each wooden fuel, the average of the lower heating value of the suitable samples was selected for use in the calculations. In the case of product gas, the lower heating value is hard to define, because of the varying quality and properties of the gasification raw material. For this reason, the lower heating value of the product gases was estimated to be slightly lower than the lower heating value of the raw material fed to the gasification. The selected heating values are presented in Table 8.

Table 8. Selected lower heating value for energy balances

	Unit	LHV, dry af
Softwood	MJ/kg	19,4
Softwood bark	MJ/kg	19,7
Eucalyptus	MJ/kg	18,0
Eucalyptus bark	MJ/kg	16,0
Heavy fuel oil	MJ/kg	40,0
Product gas (SW, bark)	MJ/kg	17,9
Product gas (euca, bark)	MJ/kg	15,0
Product gas (euca, wood)	MJ/kg	15,8
Lignin	MJ/kg	23,0
Black liquor	MJ/kgDS	14,0
Black liquor, Mill B3	MJ/kgDS	13,7

Calculations of lime kiln fuel consumption also require values for the specific heat demand of the lime kiln. Different combustion properties of fuel alternatives affect the specific heat demand of the kiln calcination reaction. The values for kiln heat demand are based on experience from past projects and are presented in Table 9.

Table 9. Specific lime kiln heat demand for each lime kiln fuel

Fuel	Specific kiln heat demand [MJ/tCaO]
Heavy fuel oil	5800
Product gas	6300
Lignin	6340

The reason for varying specific lime kiln heat demands are fuel's adiabatic flame temperatures, flame performance, and higher feed-end temperature due to poorer heat transfer in the kiln. To achieve the same production rate with different fuels, the calcination reaction requires a different amount of heat with fuels with a lower performance.

5.2 Steam and water properties for energy balances

The values for steam and water in the energy balance calculation are presented in Table 10. For both softwood and eucalyptus mills' energy balances, the same steam and water values are used.

Table 10. Steam and water values for energy balances

	p [bar]	T [°C]	h [kJ/kg]
Recovery Boiler	104,0	505,0	3383
Bark Boiler	104,0	505,0	3383
Steam to Turbine	102,0	505,0	3386
MP2 steam	29,0	300,0	2997
MP1 steam	11,5	195,0	2806
LP steam	5,0	153,0	2751
Process condensate		90,0	377
FW in tank for BB	3,9	143,0	602
FW for BB	115,0	143,0	609

FW in tank for RB	3,9	143,0	602
FW for RB	115,0	143,0	609
FW for RB after MP1 preheater	115,0	182,0	777
FW for RB after interheater	115,0	209,0	897
FW for RB after MP2 preheater	115,0	230,0	992

For each case, some assumptions were made. In each case, it is assumed that there is no district heating. In each case, two turbines are included in the mill, condensing turbine, and back-pressure turbine. The process steam pressure and temperature levels are the same for each case and, the levels are set at 29 bar and 11,5 bar for intermediate steam pressures and 5 bar for low-pressure steam. Both boilers are assumed to be high-performance boilers, that are capable of 104 bar and 505 °C high-pressure steam values.

5.3 Energy balance results

The lime kiln fuel consumption for each case is presented in Table 11. The lime kiln total heat demand is calculated with the known specific heat consumption presented in Table 9 and lime kiln capacity from main balance calculations. As mentioned earlier, the lower heating value of each fuel is selected based on references, the fuel consumption for each lime kiln case can be calculated. The gasifier losses are noticed in the lime kiln specific heat demand, so it is not noticed in the gasifier calculations. With each fuel option, the fuel availability is set to cover the whole lime kiln heat demand in a normal operating situation. Considering the product gas cases, it is good to note that, the fuel consumption is presented as the required amount of wood dry matter for product gas production. In the case of HFO and lignin, the amount of moisture in the fuel has been considered in consumption. The cases of malfunctions and stoppage time, in which fossil fuels might be used for kiln preheat and ignition is not noticed in these situations.

Table 11. Each case's lime kiln fuel consumption

	Main fuel option	Unit	Total fuel consumption [unit/d]	Specific fuel consumption [unit/tCaO]
Mill A1	HFO	t	122	0,15
Mill A2a	product gas	BDt	286	0,33
Mill A2b	product gas	BDt	286	0,33

Mill B1	HFO	t	99	0,15
Mill B2a	product gas	BDt	275	0,39
Mill B2b	product gas	BDt	261	0,37
Mill B3	lignin	t	198	0,28

As examining specific fuel consumptions, it is noted that with product gas and lignin the specific fuel consumption at least doubles or even more, compared to heavy fuel oil. Factors affecting this are the lower heating value of product gas and lignin, the higher capacity of the lime kiln due to lower lime availability, and the higher specific heat demand for calcination.

The results of the northern softwood pulp mill are presented in Table 12. The results consist of essential figures for steam consumption, steam generation, and electricity consumption and generation. The amount of surplus power can be calculated as the difference between power generation and consumption. Surplus electricity can be sold to the national grid for additional revenue. For each case, in both mill types, only one energy balance is calculated with an annual average, and seasonal differences were not considered in these cases. A summary of the energy balance results is presented in the appendix.

Table 12. Main energy balance results for softwood pulp mill cases

		Mill A1	Mill A2a	Mill A2b
Specific steam consumption	GJ/ADt	11,8	12,1	12,1
Total steam consumption	kg/s	270	253	233
-MP2 29 bar	kg/s	10,1	10,1	10,1
-MP1 11bar	kg/s	52,1	52,1	52,1
-LP 4,5 bar	kg/s	110,3	114,5	113,1
-Condensing tail	kg/s	94,2	74,0	55,8
Total steam generation	kg/s	267	251	231
-Recovery boiler	kg/s	229,4	229,4	229,4
-Bark boiler	kg/s	36,1	19,6	0,0
-Desuperheating	kg/s	1,8	1,8	1,8
Specific power consumption	kWh/ADt	655	643	625
Total power consumption	MW	76,8	75,5	73,4
Specific power generation	kWh/ADt	1704	1534	1355

Total power generation	MW	200	180	159
Specific power surplus	kWh/ADt	1049	890	729
Power surplus	MW	123,2	104,5	85,6

Results for southern eucalyptus pulp mill cases are presented below in Table 13, and the figures are presented in the same way as for the softwood cases. The same principles apply to Table 13, in terms of surplus power and other information.

Table 13. Main energy balance results for hardwood pulp mill cases

		Mill B1	Mill B2a	Mill B2b	Mill B3
Specific steam consumption	GJ/ADt	10,2	10,3	10,3	10,1
Total steam consumption	kg/s	202	193	193	187
-MP2 29 bar	kg/s	8,0	8,0	8,0	7,6
-MP1 11 bar	kg/s	44,0	44,0	44,0	42,7
-LP 4,5 bar	kg/s	95,7	96,5	96,4	96,1
-Condensing tail	kg/s	52,1	42,2	42,2	38,3
Total steam generation	kg/s	200	191	191	185
-Recovery boiler	kg/s	189,3	189,3	189,3	174,7
-Bark boiler	kg/s	9,2	0,0	0,0	9,2
-Desuperheating	kg/s	1,6	1,6	1,6	1,5
Specific power consumption	kWh/ADt	558	553	553	563
Total power consumption	MW	65,4	64,9	64,9	66,1
Specific power generation	kWh/ADt	1188	1099	1099	1056
Total power generation	MW	139,5	129,0	129,0	124,0
Specific power surplus	kWh/ADt	631	546	547	494
Power surplus	MW	74,1	64,1	64,1	57,9

6 CONCLUSIONS ON THE EFFECTS OF THE FUEL OPTIONS ON ENERGY BALANCES

The aim of the experimental part is to examine the lime kiln fuel options' effects on the energy balances of the case mills. In total seven energy balances were calculated for Nordic softwood and Southern eucalyptus mills with different options for lime kilns. The predefined case mills are presented in chapter 4, and in this chapter, these cases are compared with each other. Fuel options effects on each case mills energy balance are discussed from steam consumption, steam generation, power consumption, and power generation point of view. The cases are also considered from an economic perspective, examining the differences in operational costs, and required investments.

6.1 Steam and heat balance - Mill A cases

From the steam consumption perspective between softwood Mill A, the difference in steam consumption comes with the bark dryer, gasifier and, bark boiler. Also, the higher specific lime kiln heat demand and lower lime availability, which is noticed in cases Mill A2a and A2b with higher kiln capacity increase the heat demand for the lime kiln. In the case of Mill A1, the lime kiln is fired with heavy fuel oil, so heat is not needed for the bark dryer and gasifier. Although, the bark boiler capacity is higher compared to other cases. Mill A1 has the lowest steam consumption with 11,75 GJ/ADt. In cases, Mill A2a and Mill A2b, the bark dryer, and gasifier have almost the same steam consumption with only a small difference in bark boiler steam consumption. The Mill A2b (12,07 GJ/ADt), does not have a bark boiler, because the bark is sold, and therefore the steam consumption is lower than Mill A2a (12,09 GJ/ADt). The bark dryer utilizes secondary heat sources, but also both Mill A2 cases require 5,1 kg/s (0,37 GJ/ADt) of low-pressure steam as an addition to secondary heat sources, which is the biggest affecting factor for softwood case mills steam consumption. To sum up total steam consumption, the differences occur only in LP-steam consumption. Mill A1 consumes 110,3 kg/s of LP-steam, Mill A2a consumes the highest amount with 114,5 kg/s with the increase due to bark boiler and dryer and Mill A2b consumes 113,1 kg/s with the increase due to dryer.

From a lime kiln heat consumption point of view, the cases differ some. In the base case, the lime kiln consumes 483 GWh/a, and in both gasification cases, the kiln heat consumption is 545 GWh/a. As mentioned, this is due to higher specific heat consumption and lime kiln production capacity in gasification cases Mill A2a and Mill A2b.

Steam is generated with a recovery boiler in every softwood case and with a bark boiler in base case Mill A1 and Mill A2a. In each softwood case, the recovery boiler (including recovery boiler and desuperheating) generates the same amount of 231,2 kg/s of high-pressure steam. In the base case Mill A1, the bark boiler produces 36,1 kg/s of high-pressure steam, so the total steam generation for the base case is 267 kg/s. In the gasification case Mill A2a, total steam generation is 251 kg/s, as the bark boiler produces 19,6 kg/s of high-pressure steam. As mentioned, Mill A2b does not have a bark boiler, so the total steam generation is 231 kg/s. Process steam is extracted from the back-pressure turbine and the surplus high-pressure steam flows to condensing tail, which is used for power generation, which is discussed in chapter 6.2.

6.2 Power balance - Mill A cases

The specific power consumption for Mill A base case is 655 kWh/ADt (76,8 MW). Departments that make a difference in electricity consumption are presented in Figure 16. Mill A2a has a total power consumption of 643 kWh/ADt (75,5 MW), and the other gasification case Mill A2b without bark boiler has a total power consumption of 625 kWh/ADt (73,4 MW). The most significant difference comes from the bark dryer and gasifier with 4,3 kWh/ADt, between the base case and gasification options. In each case, the bark boiler has different capacities, because of different bark and wood residue availability. Mill A2b does not have a bark boiler, which results in a 24 kWh/ADt difference, compared to the base case. There is also a difference in the electricity consumption of cooling water pumping. The origin of the cause comes from the difference in the size of the bark boiler and as a result the amount of required cooling water pumping to the turbine condensers.

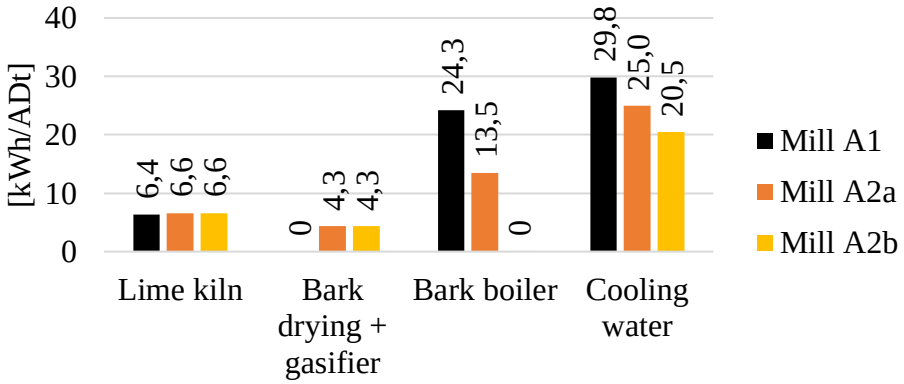

Figure 16. Softwood mill cases' departments with power consumption differences

Each softwood mill case is self-sufficient in power generation and has surplus power for sales to the national grid. In each case, the mill has a back-pressure and condensing turbine, in which the back-pressure turbine generates electricity with 73,5 MW power. The amount of power generated in condensing turbine depends on steam generation and consumption. With the left-over high-pressure steam, power is generated with the condensing turbine. In Mill A1, 134,7 kg of high-pressure steam flows to the condensing turbine generating electricity with 126,2 MW power. With the same principle, Mill A2a generates electricity with 106,4 MW power as 118,6 kg of high-pressure steam flows to the condensing turbine. In Mill A2b, 99,4 kg of high-pressure steam flows to the condensing turbine generating 85,7 MW of electricity.

As the production capacity and mills' annual running time is known specific heat generation can be calculated for each case. For each case mill, specific power generation and surplus power are presented in Figure 17. As the power generation in each mill is higher than power consumption, each case mill is self-sufficient in terms of power generation. Mill A1 has the highest degree of power self-sufficiency with 260 %, Mill A2a 238 %, and Mill A2b 217 %. Differences in power generation come from the bark boiler because the recovery boiler has the same parameters in each case. In Mill A1, all the bark and wooden residues are burned in the bark boiler, which means the bark boiler has more fuel available and allows for higher capacity. In gasification cases, the required amount of bark goes via gasifier to the lime kiln, which means less fuel is available. In the case of Mill A2a, surplus bark is burned in the bark boiler, and in Mill A2b, surplus bark is sold and so it does not have a bark boiler. Therefore, Mill A2a has a higher power generation capacity, compared to Mill A2b.

With a higher power self-sufficiency rate, more power is available for sales. Surplus power increases additional incomes for the mill. Although Mill A1 has the highest power consumption, it generates more power, than gasification case mills, which leads to a higher self-sufficiency rate. Potentially Mill A1 has 1049 GWh annually for sales as Mill A2a has 890 GWh and mill A2b 729 GWh. Revenues from electricity sales are discussed further from the perspective of the mills' economic profitability in chapter 6.5.

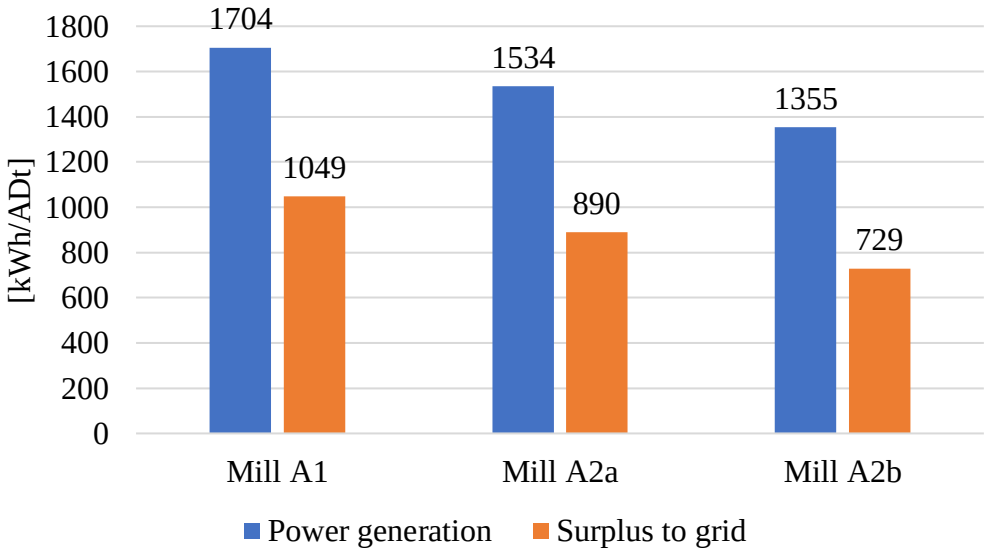

Figure 17. Softwood cases' specific power generation and surplus power available for sale

6.3 Steam and heat balance - Mill B cases

In the southern eucalyptus base case, Mill B1's specific steam consumption is 10,19 GJ/ADt. With bark boiler included in the mill, and lime kiln fired with heavy fuel oil, Mill B1 has lower steam consumption, than gasification cases Mill B2a and mill B2b. As the bark dryer is a more significant steam consumer (0,09 GJ/ADt), the case Mill B2 both options with additional bark and additional wood have higher steam consumption with 10,27 GJ/ADt and 10,26 GJ/ADt. This little difference between these options comes from bark gasifier dryer capacity, which varies because of the different properties of eucalyptus wood and bark. The total steam consumption in Mill B3 is 10,13 GJ/ADt. Mill B3 does not have drying and gasifying processes for bark, although the lignin separation requires a higher capacity for evaporation plant, which increases the MP1 and LP-steam consumption, by 0,08 GJ/ADt. As lignin is separated, the organic substances and so the heating value of black liquor

decreases, which causes lower recovery boiler capacity. This decreases the total heat load of the recovery boiler, and also decreases the recovery boiler's MP1- and MP2-steam consumption, by 0,14 GJ/ADt. Mill B3 has a bark boiler with the same capacity as Mill B1, so in this perspective, it is no different from the base case. To sum up steam consumption in eucalyptus case mills, comparing the base case and gasification cases, the differences occur in LP-steam consumption, which is due to gasifier, dryer, and bark boiler heat demand. With the case Mill B3, there are differences in MP2, MP1, and LP-steam consumptions compared to the base case. The smaller required heat load of the recovery boiler affects the consumption by lowering the MP2 and MP1-steam consumptions. On the other hand, higher evaporation capacity increases LP-steam consumption. In total the recovery boiler has a greater effect on the consumption, which leads to a result, that Mill B3 has lower specific steam consumption than base case Mill B1.

From a lime kiln heat consumption point of view, the cases differ from each other likewise in northern softwood mills. The specific lime kiln heat demands vary with each fuel, which is presented earlier in chapter 5, Table 9. Also, both options of gasification Mill B2 and lignin firing lime kiln Mill B3 are set with lower lime availability, and so calculated with higher lime kiln capacity, which increases heat demand. The base case Mill B1, consumes 390 GWh/a, with the lowest heat consumption in the lime kiln. Both gasification options, Mill B2a and Mill B2b have the same heat consumption with 439 GWh/a, and the lignin firing kiln, Mill B3, has the highest heat consumption in the lime kiln with 442 GWh/a.

Steam is generated with recovery boiler and bark boiler in base case Mill B1 and lignin separation case Mill B3. In gasification cases, all the bark and wooden residues are gasified for lime kiln fuel, and therefore, both Mill B2 options do not have a bark boiler for steam generation. Considering recovery boiler steam generation, cases Mill B1 and Mill B2 options generate 191,3 kg/s of high-pressure steam. In the case of Mill B3, lignin is separated from the black liquor, which affects the heating value of the strong black liquor. The extracted lignin decreases available dry solids in black liquor. In this case, the black liquor heating value is estimated to be 13,70 MJ/kgDS. In other cases, the black liquor lower heating value is estimated to be 14,00 MJ/kgDS. As a result of lignin separation, the required capacity of the recovery boiler and steam production per liquor solids decreases. As mentioned earlier, other cases produce 191,3 kg/s of high-pressure steam, and exceptionally Mill B3 produces

176,4 kg/s. With cases included with bark boiler Mill A1 and Mill A3, the bark boiler produces 9,2 kg/s of high-pressure steam. Compared to the Nordic softwood mill bark boiler, the capacity is much lower due to lower fuel availability, and the steam generation effect is not so high.

6.4 Power balance - Mill B cases

When considering the power consumption of base Mill B1, the specific power consumption of Mill B3 is 558 kWh/ADt. With both gasification cases Mill B2a and Mill B2b, the specific power consumption is 553 kWh/ADt. The last eucalyptus Mill case B3 has a specific power consumption of 563 kWh/ADt. The departments that affect the energy balance are presented in Figure 18. Considering the evaporation plant, the Mill B3 has a higher capacity, than the base case and gasification cases. The difference is due to the higher evaporation capacity for lignin separation from black liquor. In the case of Mill B3, the lignin separation also affects the power consumption of the recovery boiler. The separated amount of lignin from black liquor decreases the heating value of the black liquor, which also decreases the required recovery boiler capacity. Lower boiler capacity decreases the power consumption by 5 kWh/ADt but also decreases the total amount of steam produced in it. As the power consumption in the recovery boiler decreases, the lignin separation process LignoBoost and lignin drying to suitable moisture for combustion in a lime kiln, requires 12 kWh/ADt of more power, compared to the cases without lignin separation. As Mill B2 options consist of drying and gasifying process, those mills consume 4 kWh/ADt. As the gasification option consumes power on bark drying and gasifier, they do not have a bark boiler, which consumes 7 kWh/ADt, in the base case and Mill B3. Also, little differences in cooling water pumping power consumption occur due to the same reason as in softwood cases. Different recovery- and bark boiler capacities lead to a difference in the pumping volumes of the cooling water for the turbine condenser and thus to different power consumptions. The main departments that make a difference in power consumption are shown in Figure 18.

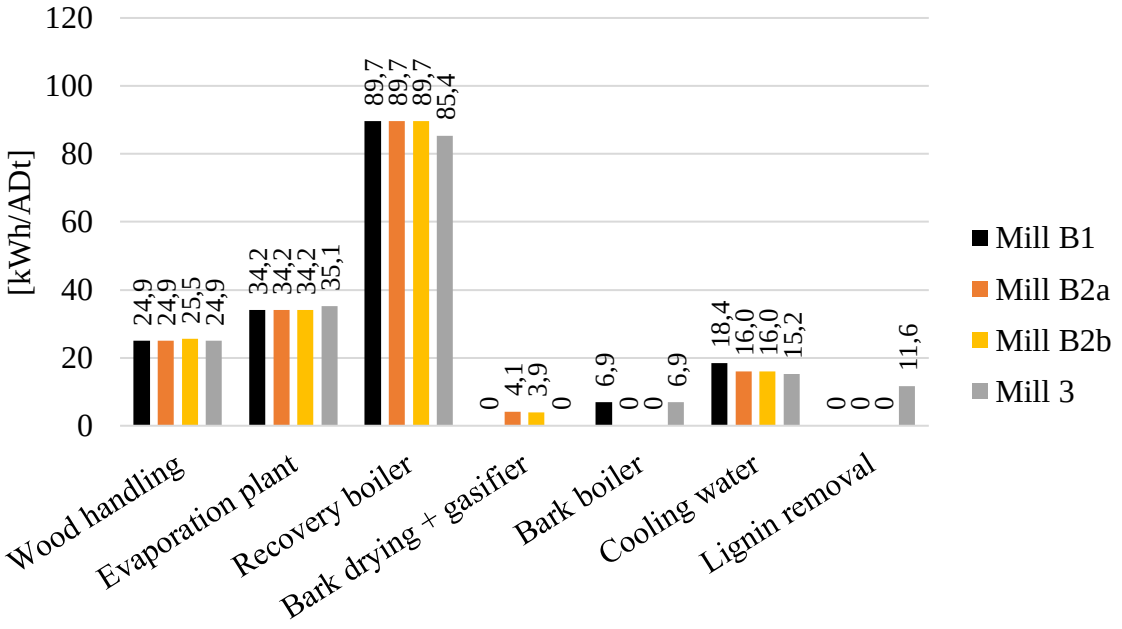

Figure 18. Eucalyptus mill case departments that differ in power consumption

In the eucalyptus case mills, each mill generates more power, than it consumes, which means surplus high-pressure steam is used for electricity generation. This surplus electricity is sold to the grid as in softwood cases to get additional revenue for the mill. In Figure 19, specific power generation capacity and available surplus electricity for sales is presented. In each case, the back-pressure turbine generates electricity with 65,4 MW power. Power is also generated with the condensing turbine. The amount of power generated with the condensing turbine varies from case to case. In eucalyptus base case Mill B1, 83,4 kg of high-pressure steam flows to the condensing turbine, generating 74,1 MW power. With the Mill B2 both options, condensing turbine generates the same amount of 63,6 MW, with 74,2 kg of high-pressure steam. In the last eucalyptus case Mill B3, where lignin is separated and thus required recovery boiler capacity decreased, 68,7 kg of high-pressure steam flow to the condensing tail generating electricity with 58,3 MW power. Likewise, in softwood case mills, every eucalyptus mill is also self-sufficient, in terms of power. The power self-sufficiency rate for the base case is 213 %. With both gasification options, the rate is 199 % and with the lignin, it is 188 %.

Considering the available surplus electricity for sales, the base case has the highest capacity with 631 kWh/ADt, which is annually 631 GWh with the assumed pulp production capacity. Compared to Mill B2 options, the difference comes from the power generated with the bark

boiler, which both generate 546 kWh/ADt surplus electricity, which is 85 GWh per year less electricity for sales. In the case of Mill B3, there are 494 kWh/ADt of surplus power available and it is lower due to lower recovery boiler capacity and higher power consumption, compared to the base case. Annually, this means that there are 137 GWh less power sales, compared to base case Mill B1.

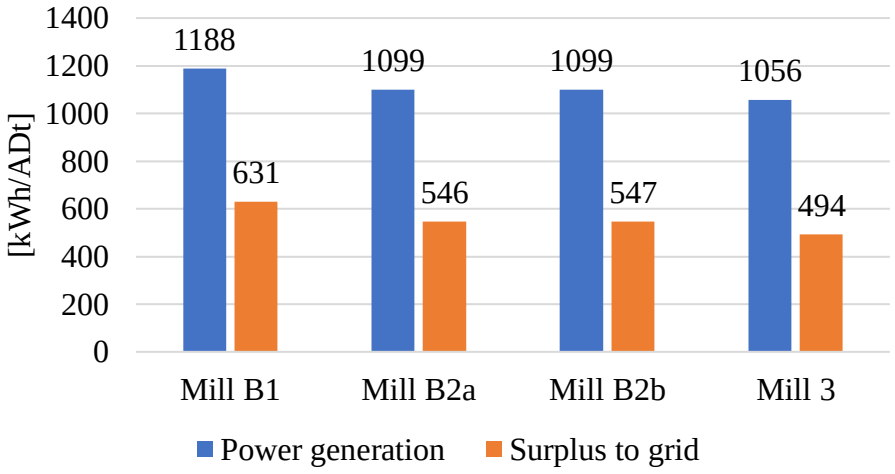

Figure 19. Eucalyptus case mills' specific power generation and surplus power available for sales

6.5 Cost comparison

Case mills are compared with each other from an economic point of view. In this chapter, operational expenses (OPEX) and capital expenditures (CAPEX) are discussed. The capital expenditures are roughly estimated by AFRY's cost accounting. Operating expenses are based on calculated balances and estimated prices of each expense and income. Both base case mills are used as a comparison for alternative cases and the costs are examined as a cost difference. The considered capital expenditures are focused on processes that enable alternative fuels to be burned in a lime kiln and boiler and turbine modifications. The operating expenses compare increased costs and lost revenues to savings and additional revenue from side streams.

6.5.1 OPEX, Operating expenses and incomes

The operating expenses focus on comparing lost electricity sales income, increased chemical and wood costs, savings from HFO replacement, emission trading, and additional revenues from bark sales. For OPEX calculation, the location selected for the Nordic softwood mill is

Finland and Brazil for the southern eucalyptus mill. The prices used in OPEX invoices are tied to the latest available prices. It should be noticed that changes in prices may significantly affect the results. The designated countries' national taxes and practices have also been taken into account in the invoices.

Table 14. Assumed prices for OPEX calculations

Nordic electricity	45 €/MWh
South American electricity	40 €/MWh
Bark	10 €/MWh
Eucalyptus wood	25 €/m^3sub
CO2-credit	60 €/t CO_2
Carbon dioxide	60 €/t CO_2
Sulfuric acid	70 €/t H_2SO_4
HFO in Nordics	650 €/t
HFO in South America	350 €/t

The amount of CO_2-emission was calculated with a specific emission factor, based on a reference by Isakkson (2007). The price of Nordic HFO is calculated as the sum of the HFO import price and the excise duty. The electricity price for Brazil was estimated as an average of the year 2020 and beginning of 2021 because at the moment in Brazil the price of electricity is exceptionally high due to low rainfall and shortage of hydropower. The situation may change rapidly as the rains arrive, which is why the price is estimated for the longer term, so simulated normal situation. (Reuters 2021) The price of Nordic electricity is calculated as a 2021 yearly average, for the period from January to July (Nord Pool 2021).

The Nordic softwood cases are presented in Table 15. The figures are presented as a cost difference compared to the base case.

Table 15. Nordic softwood mill cases' annual operating expenses compared to the base case

		Mill A1	Mill A2a	Mill A2b
Electricity sales	mil €	0,00	-7,2	-14,4
HFO savings	mil €	0,00	28,2	28,2
Bark sales	mil €	0,00	0,00	4,6
CO2 credit saving	mil €	0,00	8,1	8,1
Summary	mil €	0,00	29,1	26,5

As reviewing operating expenses, the price of electricity and heavy fuel oil has the biggest impact on operational viability. Emission trading also plays a role in annual savings, as the use of heavy fuel oil requires the acquisition of CO2 emission permits. As the softwood mill is assumed to be in Finland, it is part of the European Union's Emission trading system (ETS). Comparing the two gasification cases, with the current price of electricity, sawdust, and bark sales price, Mill A2a seems more profitable from the operational cost perspective. However, it can be noticed that in both softwood cases, compared to heavy fuel oil base case, the alternative fuel cases achieve significant leverage when considering the operational expenses. In addition, the operational costs of the replacement method for sludge disposal in the bark boiler have not been included in the calculations.

In Table 16, southern eucalyptus cases are presented, compared to the base case. The costs differ slightly from the costs of the softwood mills. Increased chemical costs have been added to the table based on the fact that lignin separation increases chemical consumption (carbon dioxide CO_2 and sulfuric acid H_2SO_4) for the separation process. In the case of Mill B2b, a higher amount of wood is consumed, compared to other cases. Also, CO_2 credits have been removed from the table, because at the moment Brazil does not have an operating emission trading system. However, the implementation of the ETS system is planned and may enter into force in the future (World Bank Group 2021, 22).

Table 16. Southern eucalyptus cases' annual operating costs compared to the base case

		Mill B1	Mill B2a	Mill B2b	Mill B3
Electricity sales	mil €	0,00	-3,4	-3,4	-5,5
Chemical cost	mil €	0,00	0,00	0,00	-1,9
Wood cost	mil €	0,00	0,00	-2,1	0,00
HFO savings	mil €	0,00	12,3	12,3	12,3
Summary	mil €	0,00	8,9	6,8	4,9

Like in northern softwood cases, southern eucalyptus cases' the most important operating expenses are heavy fuel oil cost and electricity sales. The biggest impact on operating costs is caused by the price of heavy fuel oil. The use price of heavy fuel oil is considerably lower, as its price does not include excise tax, unlike in the case of the Nordic mill cases. Also, the lack of an emission trading system decreases the profitability. If a functioning ETS would be implicated in South America, the carbon-free options would become more attractive from

an economic point of view. Also, in the cases Mill B2a and Mill B2b without bark boiler, the operational costs do not include the additional expenses of sludge disposal. The case Mill B3, has the lowest annual savings, because of the highest lost share of power surplus income due to lower recovery boiler power steam generation. Also, lignin separation adds costs as chemical consumption increases. One way to mitigate the chemical cost would be the internal chemical production of sulphuric acid and carbon dioxide with carbon capture (Kihlman 2021, 14). This would support the biorefinery concept, using the mill's raw material flows, and would likely have an impact on the mill's chemical balances, especially Na/S-balance. In addition, it should be noticed, that with lignin separation increased pulp production is a possibility. The revenue from additional pulp production would increase profitability significantly compared to other cases, which has not been examined in this thesis.

6.5.2 CAPEX, Capital expenditures for upgrade and new mill

In this thesis, the evaluation of capital expenditures is done through the savings from operating costs, the payback period, and the interest rate. The additional income from the change in alternative cases operating costs is used to calculate, at different payback periods and interest rates, the maximum amount of investment in order to make the investment financially viable. The required investments for each case are estimated by AFRY's costs accounting team and those numbers are compared with the calculated maximum investment threshold. The maximum investment limit is calculated with the annuity method. In the annuity method (equation 9), the basic investment is divided into equal annual costs (annuities) for the payback time. The principal will be repaid within the agreed payback time in equal annual installments. The acquisition cost is divided for the different years of the payback time using an annuity factor. In this case, the annual net return is divided by the annuity factor to calculate the maximum value of the investment (equation 10). The size of the investment must be below this figure for the investment to be considered profitable.

$$S - c_{n,i} \cdot I > 0 \tag{9}$$

$$I < \frac{S}{c_{n,i}} \tag{10}$$

where, S is annual net income [million €], $c_{n,i}$ is annuity factor and I is investment [million €].

In Table 17, the maximum value for the investment is presented with different payback times and rates of interest. In this thesis, the investments are calculated to be repaid by the net savings from fossil fuel replacement. The annual net income is the OPEX costs difference, compared to the base case, from Table 15 and Table 16. When using the annuity method, the annual income should be fairly even. In this case, the amount of annual revenue might vary a little, but it is assumed to be even enough, as the calculations are annual averages, and thus the effects of small fluctuation decrease in the long term. However, it is good to take this into account when assessing the profitability of the investments and the maximum limit.

Table 17. The maximum limit value for each cases' investments, in terms of payback time and rate of interest. The values are presented in million €.

Payback time [a]	Rate of interest	Mill A2a	Mill A2b	Mill B2a	Mill B2b	Mill B3
5	0,0 %	146	133	44	34	24
5	5,0 %	126	115	38	29	21
5	10,0 %	110	101	34	26	19
10	0,0 %	291	265	89	68	49
10	5,0 %	225	205	69	53	38
10	10,0 %	179	163	55	42	30

With the upgrade mill consideration, the maximum values calculated with the 5 years pay-back time is used as reference. With softwood gasification cases, it can be stated that the amount of required investments for the upgraded mill are well below the maximum values at all rates of interest, presented in Table 17. Upgrade from base case to Mill A2a and Mill A2b, requires investments only for belt dryer and gasifier. However, in the case of an up-grade mill for alternative softwood and eucalyptus cases, it is assumed that the existing lime kiln has the capacity for an increase in lime production. If the lime kiln capacity is fully utilized before the upgrade, it will require separate investments that have not been taken into account in these upgrade cases. As examining the AFRY's investment estimates, the values for required upgrade investments are many times smaller than the maximum investment limit values. In conclusion, based on these estimates, an upgrade from heavy fuel oil to product gas can be considered a profitable fuel alternative for northern softwood mill cases.

With hardwood gasification cases and lignin separation cases, the investment limits are much lower, because of the lower annual additional incomes, compared to the base case. Upgrade from base case to Mill B2a and Mill B2b, requires investments for belt dryer, gasifier, an upgrade for hog fuel silo, and wood yard investments for debarking drum in Mill B2a and chipping line in Mill B2b. However, even with the 10 % rate of interest and so the lowest maximum investment limits, the AFRY's estimates are slightly lower. In conclusion, based on these estimates and calculations, the product gas options can be considered as a potential and economically competitive fuel alternative for an upgraded eucalyptus mill.

With Mill B3, the AFRY's estimate for upgrade is higher, than the maximum investment limits with every rate of interest. However, it should be stated, that with lignin separation additional pulp production is possible with Mill B3. As a result, additional pulp production would increase additional annual OPEX savings and thus profitability. However, in this study, it is not taken into consideration and should be studied further for more accurate conclusions.

In the case of a new mill, the investment comparison is performed by comparing all the departments to which are affected due to the different lime kiln fuel options and case definition. For a new softwood mill compared to base case Mill A1, with the defined lime kiln, bark boiler, and turbine configurations, Mill A2a requires higher investments for dryer, gasifier, and lime kiln, and lower investments are needed for bark boiler and condensing turbine. To sum up the difference, between Mill A2a and base case, CAPEX difference in case of new mill's total investment is smaller than the annual OPEX savings, which means despite the higher required investments, the annual savings cover the higher investment in short term. In the case of Mill A2b, the required investments in the case of a new mill are lower, compared to the base case, because there is no bark boiler in Mill A2b. In conclusion, for Mill A2b, the total investments for a new mill are lower, compared to the base case and also, significant savings can be achieved with the replacement of heavy fuel oil. Based on these calculations, investment estimates, and case definitions, it can be stated that the alternative fuel mill options are more profitable also in the case of a new mill.

With eucalyptus product gas cases, the annual OPEX savings cover the increased total investment difference in the case of a new mill. In the product gas cases, higher investments are required in fuel storage, belt dryer, gasifier, lime kiln, and wood yard equipment.

However, in Mill B2a and Mill B2b, the absence of a bark boiler affects the new mill's investments by reducing the investment difference, compared to the base case. In addition, when looking at the results in cases Mill B1 and Mill B3, it was found that the size and the estimated investment cost are not profitable for this production rate and fuel availability. Typically, in eucalyptus pulp mills, with lower fuel availability, the bark boiler acts as a sludge disposer and in reality, the size of the investment would be smaller. Even when this is taken into consideration, it can be still stated that the OPEX savings from relatively short-term operation cover the higher investment costs difference for both cases.

In the case of Mill B3, lower required investments occur in the recovery boiler and condensing turbine. However, the investment for the LignoBoost plant is very high, and thus, based on Table 17 values, the investment difference would be covered only with the 0 % rate of interest and 10 year payback time. However, it is good to note that the additional pulp production possibility has not been taken into account and its impact on profitability can be significant.

Also, when comparing the eucalyptus cases, it can be clearly stated that an operating ETS system or CO_2-taxation would reduce the competitiveness of fossil fuel options and thus improve the profitability of lime kiln fuel alternatives. As mentioned earlier, such a system is planned, for example in Brazil, which is the designated country in this thesis. The ETS system implementation would have a significant impact on the eucalyptus cases economic review of this study. In a summary, the alternative fuels in the lime kiln can be considered as an economically profitable option in the case of upgrade mill and design of a new mill. In addition, the competitiveness of these alternative fuels will improve if the price of fossil fuels, carbon emissions credits, and energy continues to rise.

7 SUMMARY

The aim of this thesis was to introduce alternative lime kiln fuels for the replacement of traditional fossil fuels and to examine the effects of alternative lime kiln fuels, on the pulp mill energy balance. Also based on the results of mill energy balances, the profitability of the alternative fuel options were examined, based on net operating expense difference, and required investments. The results were compared to a pulp mill, with heavy fuel oil firing lime kiln.

The literature section introduced the basic principles of the chemical recovery and lime kiln and introduced alternative fuels to replace the fossil fuel traditionally used in a lime kiln. The examined alternative fuel options were all based on the fuels available inside the mill site. The review of the fuels was based on literature sources. For many examined fuels, the potential to replace fossil fuel options, in whole or partly, was noticed. Examining the fuel alternatives, the most significant factors were stated to be the properties of the fuel and the effects to lime burning and lime cycle, the adequate and even availability of fuel, and the economic impact. A few fuel alternatives were selected for further consideration in the experimental section.

In the experimental part, in total, seven different energy balances were calculated for two different mill types, northern softwood, and southern eucalyptus pulp mill. For the energy balance calculations, main fiber balances were required to calculate, to solve the available fuel flows and department capacities. Main fiber- and energy balances were calculated to cases with, fuel oil firing lime kiln, product gas firing lime kiln produced by gasification with few different configurations, and lignin firing lime kiln, separated from black liquor. The alternative fuel energy balances were compared with the fossil fuel firing base case, and the fuel options effects were based on the difference compared to the base case.

The main conclusions considering the effects of product gas option to energy balance were, that dryer and gasifier increase specific steam consumptions, with low-pressure steam consumption. With the product gas option, the availability of bark boiler fuels is lower, and so the capacity of bark boiler decreases, which leads to lower steam generation, and lower specific power generation in each product gas case. The dryer and gasifier increase the power

consumption, but in these cases, the greater effect is due to the decreased steam and power generation. As a result, the amount of surplus power available will also decrease.

The main conclusions on the effects of lignin separation for lime kiln use are, that the steam balance is affected by reduced steam consumption and steam generation in recovery boiler and increased evaporation plant steam consumption. Although the power consumption is decreased in recovery boiler and cooling water pumping departments, the effects of increased evaporation capacity, lignin separation, and drying, increases the specific power consumption higher, than in the base case. From the effects of higher specific power consumption and decreased total steam generation in the recovery boiler, the specific power generation and so, available surplus power decreases, compared to the base case.

In terms of operating costs, each alternative fuel case generates savings compared to the heavy fuel oil base case. The saved operating costs by replacing the heavy fuel oil are higher than the lost revenue from lost power sales and other increased operating costs. With northern softwood cases, the OPEX difference is even bigger, than with southern hardwood cases, because of the savings from emission trading and carbon taxation, when using heavy fuel oil.

In terms of investment costs, the alternative fuel options require additional investments compared to a traditional pulp mill. The required investments were estimated by AFRY cost accounting. The cases were examined in cases of upgrade and new plant. In summary, the product gas is a viable option in softwood and eucalyptus cases. With lignin separation, the high investment for the LignoBoost plant decreases economic competitiveness in this thesis, but the effects of possible additional pulp production should be studied further, to solve the effects on economic profitability.

In a conclusion, based on these cases and calculations, alternative fuels can be found profitable, compared to fossil fuel use in the lime kiln. However, the cases and calculations are estimates and include certain assumptions, which means that it should be stated that the practical suitability and economic consideration of the corresponding plants should be evaluated further. However, the initial values used in the calculations are reliable and based on experience, due to which the results in energy balance consideration can be considered comparable.

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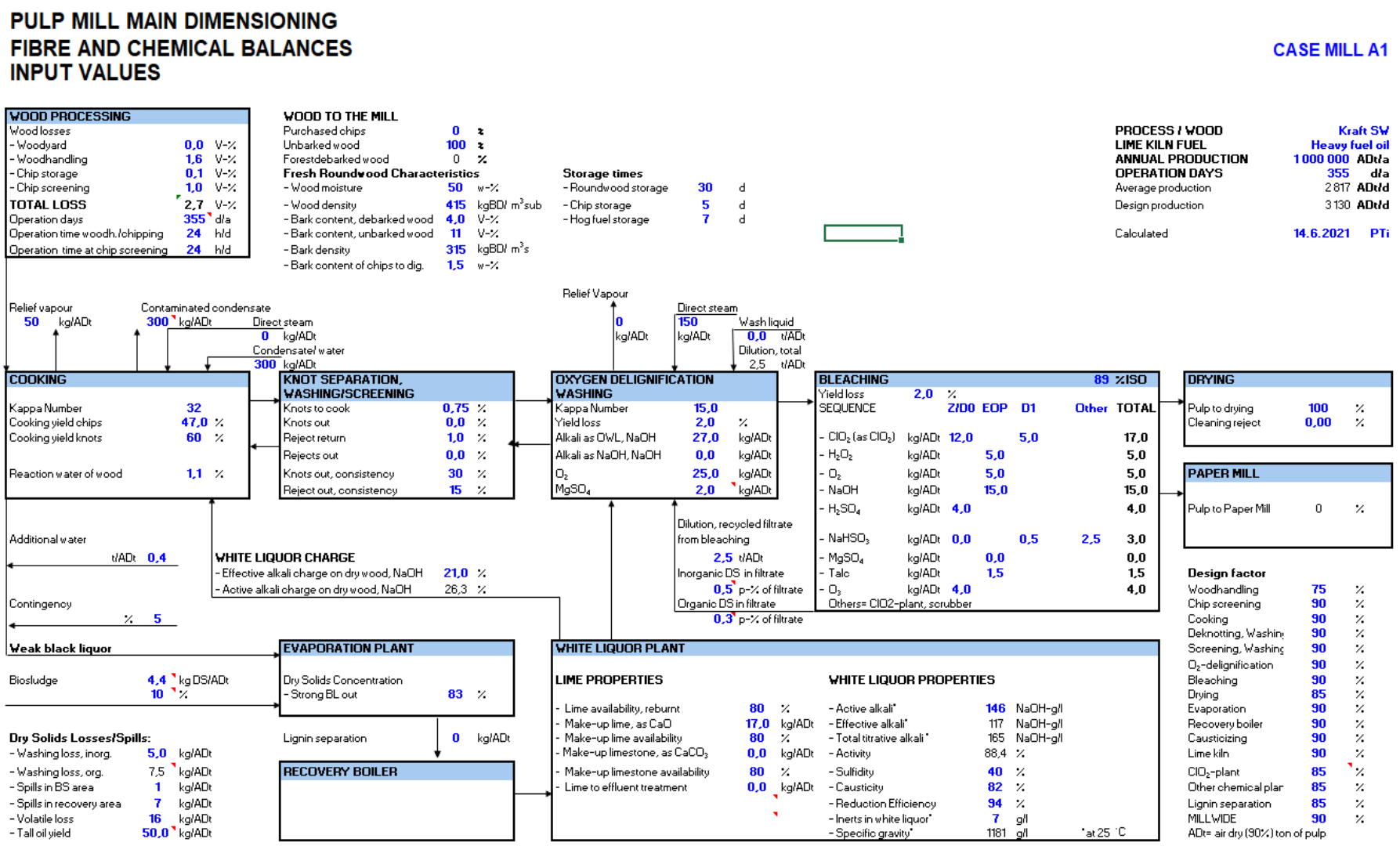
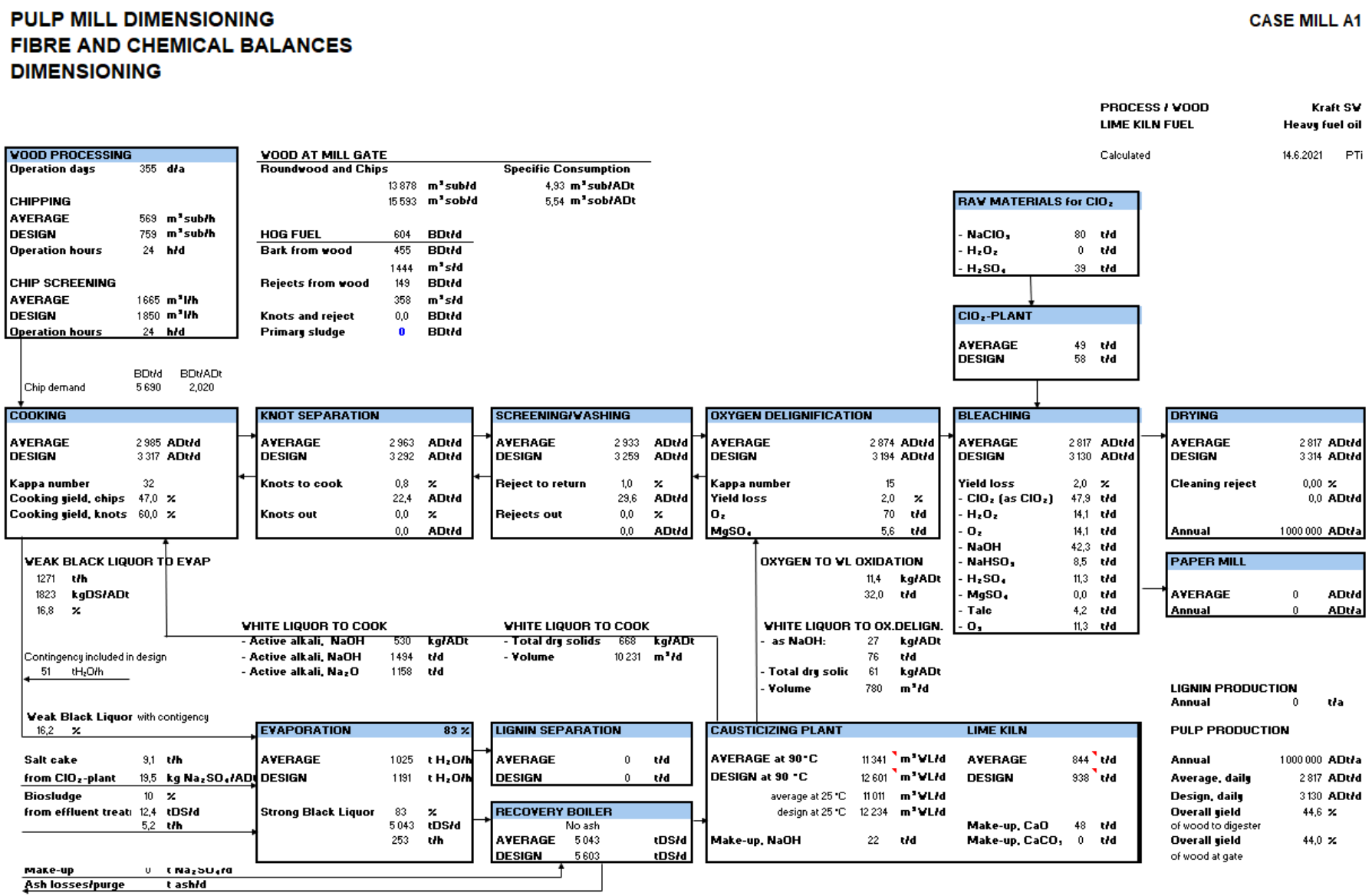
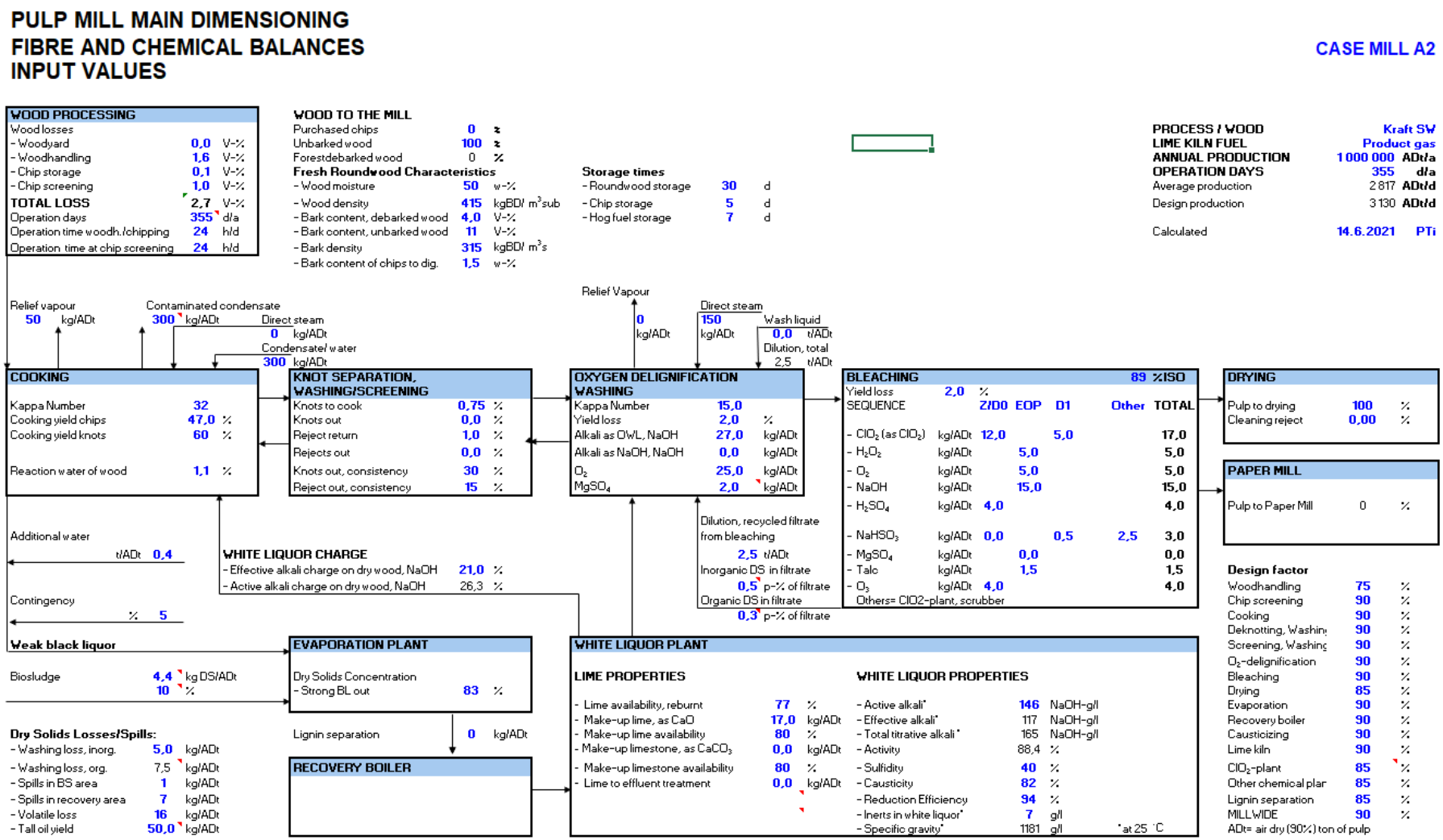
Appendix I: Fibre- and chemical balance Mill A1 - input



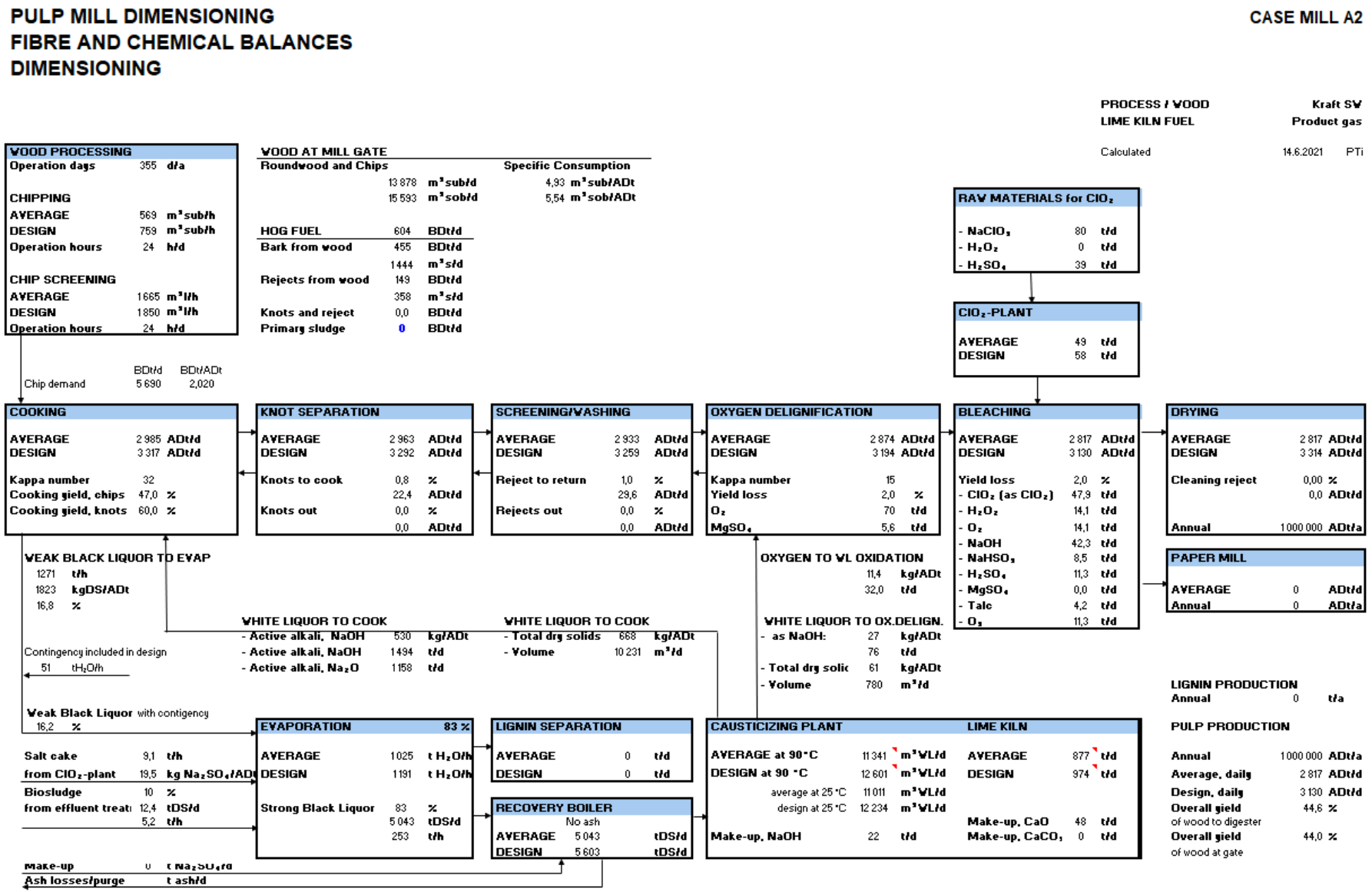
Appendix II: Fibre- and chemical balance Mill A1 - results



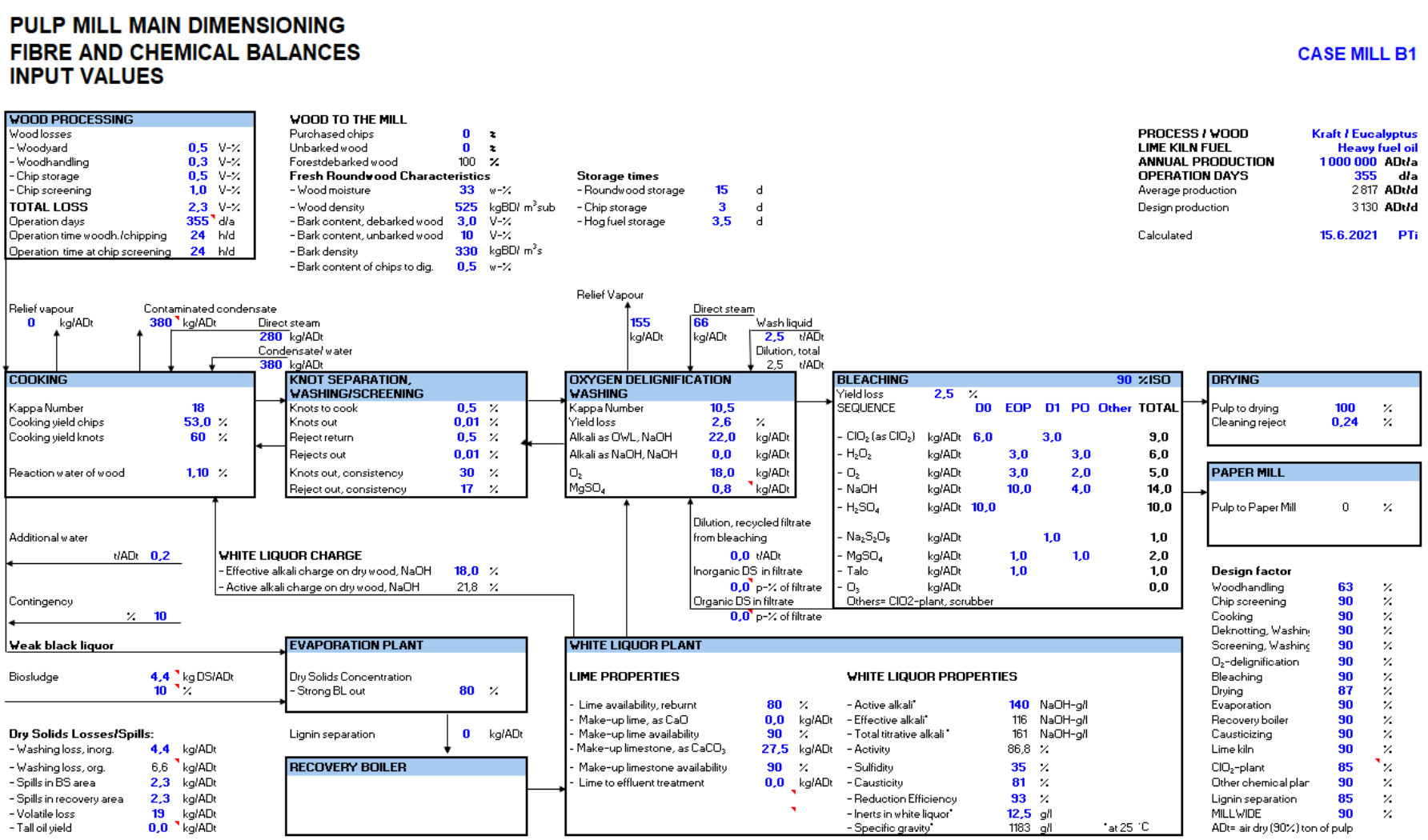
Appendix III: Fibre- and chemical balance Mill A2 - input



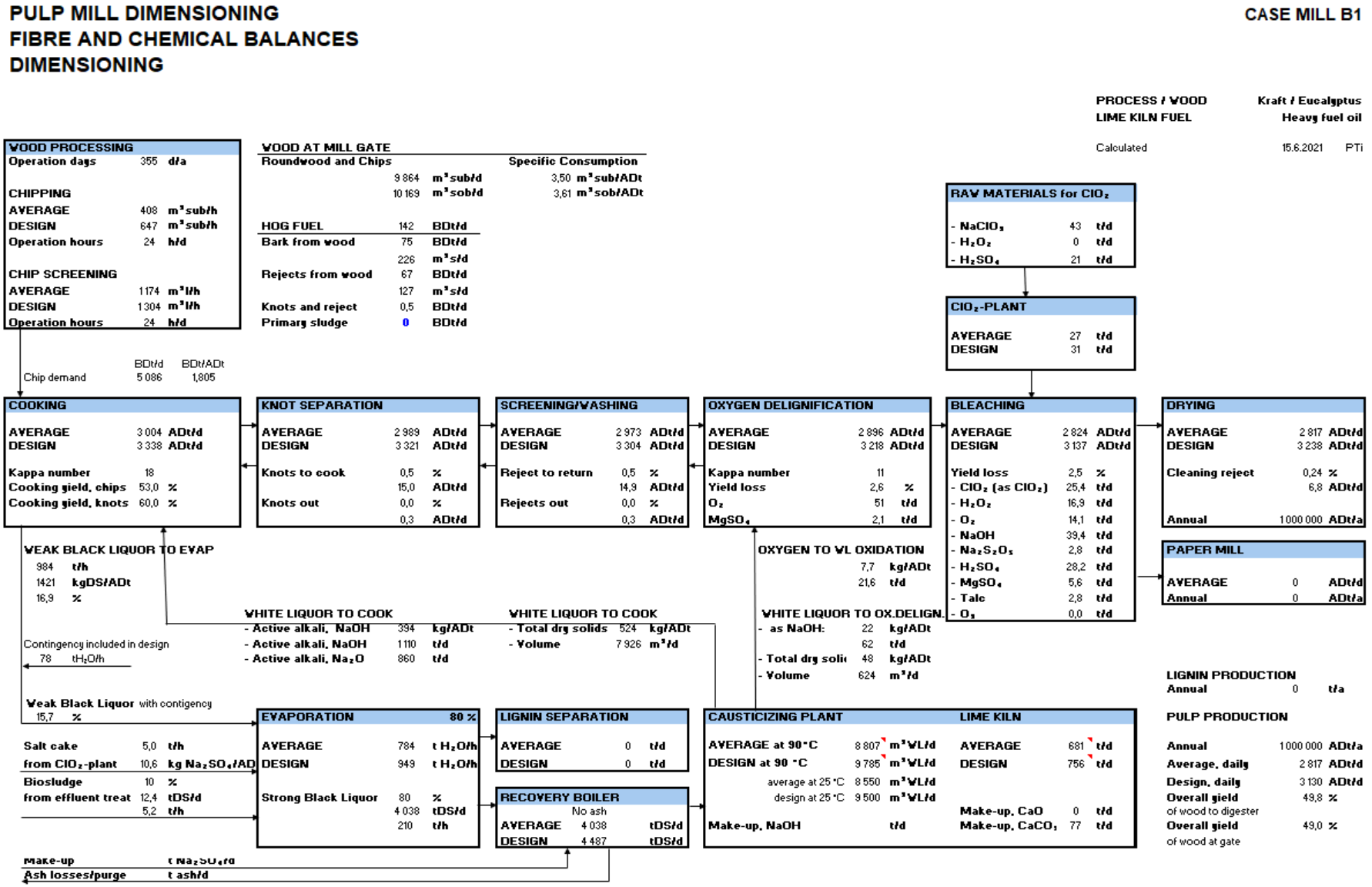
Appendix IV: Fibre- and chemical balance Mill A2 - results



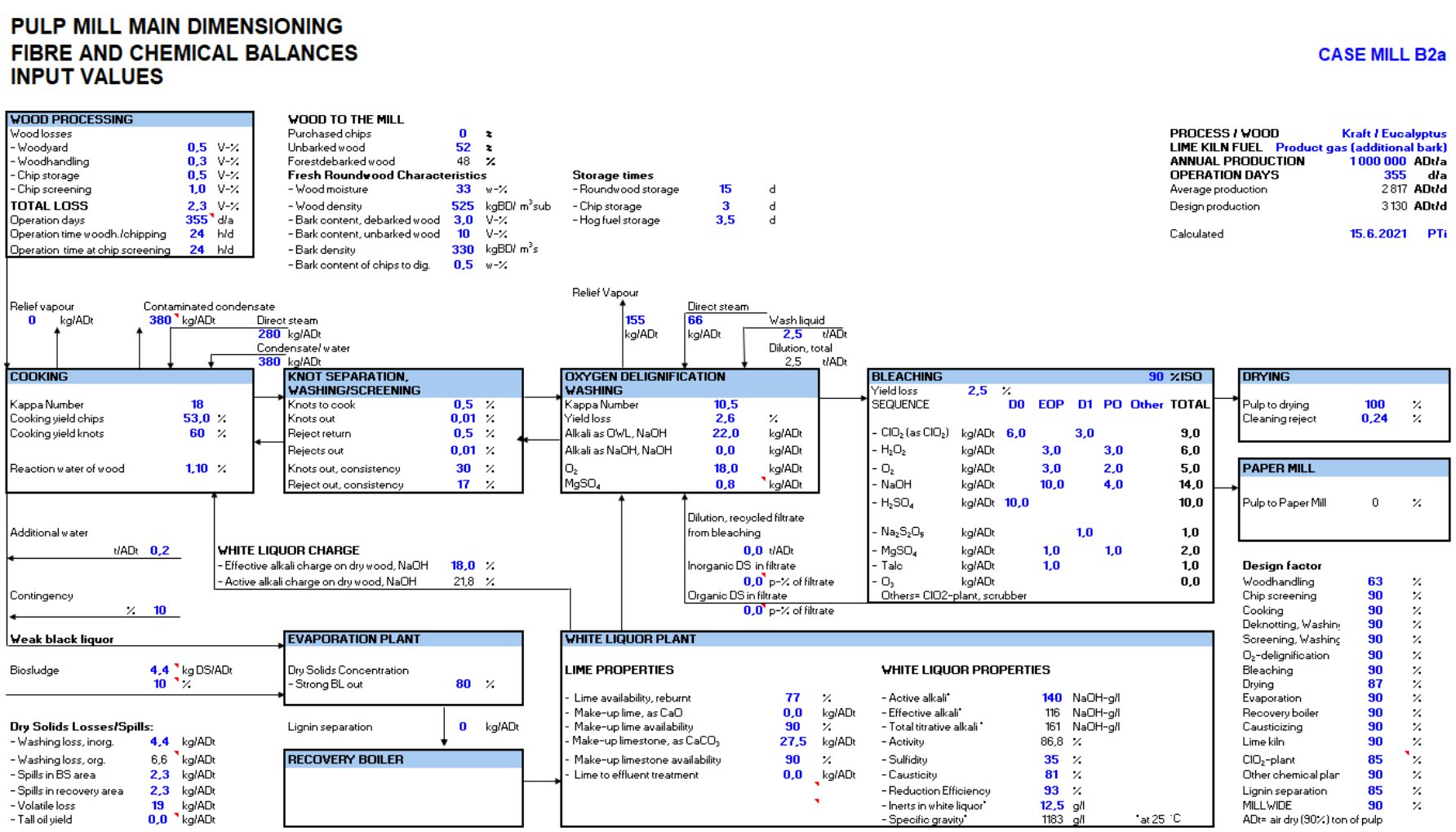
Appendix V: Fibre- and chemical balance Mill B1 - input



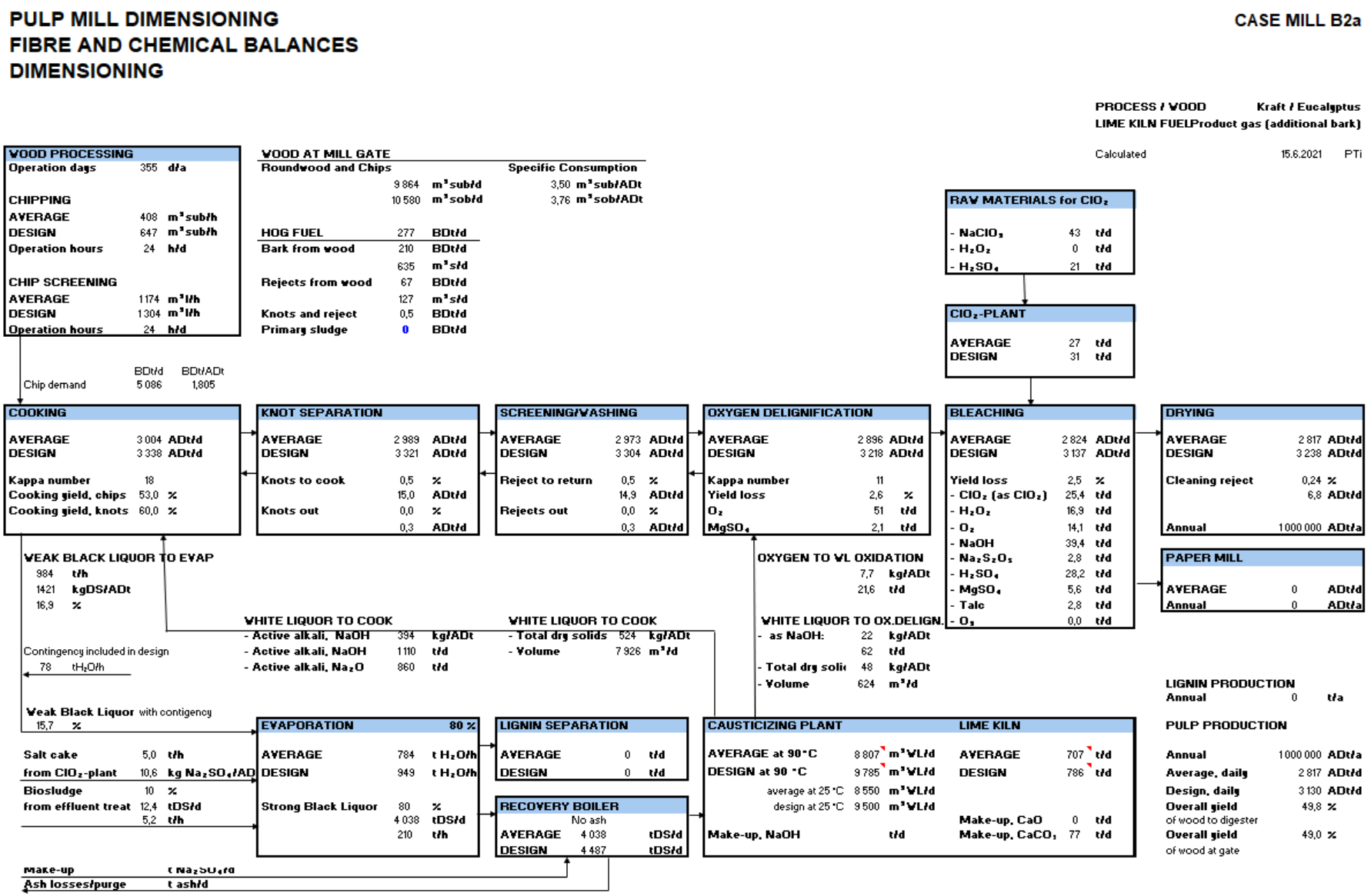
Appendix VI: Fibre- and chemical balance Mill B1 - results



Appendix VII: Fibre- and chemical balance Mill B2a - input



Appendix VIII: Fibre- and chemical balance Mill B2a - results

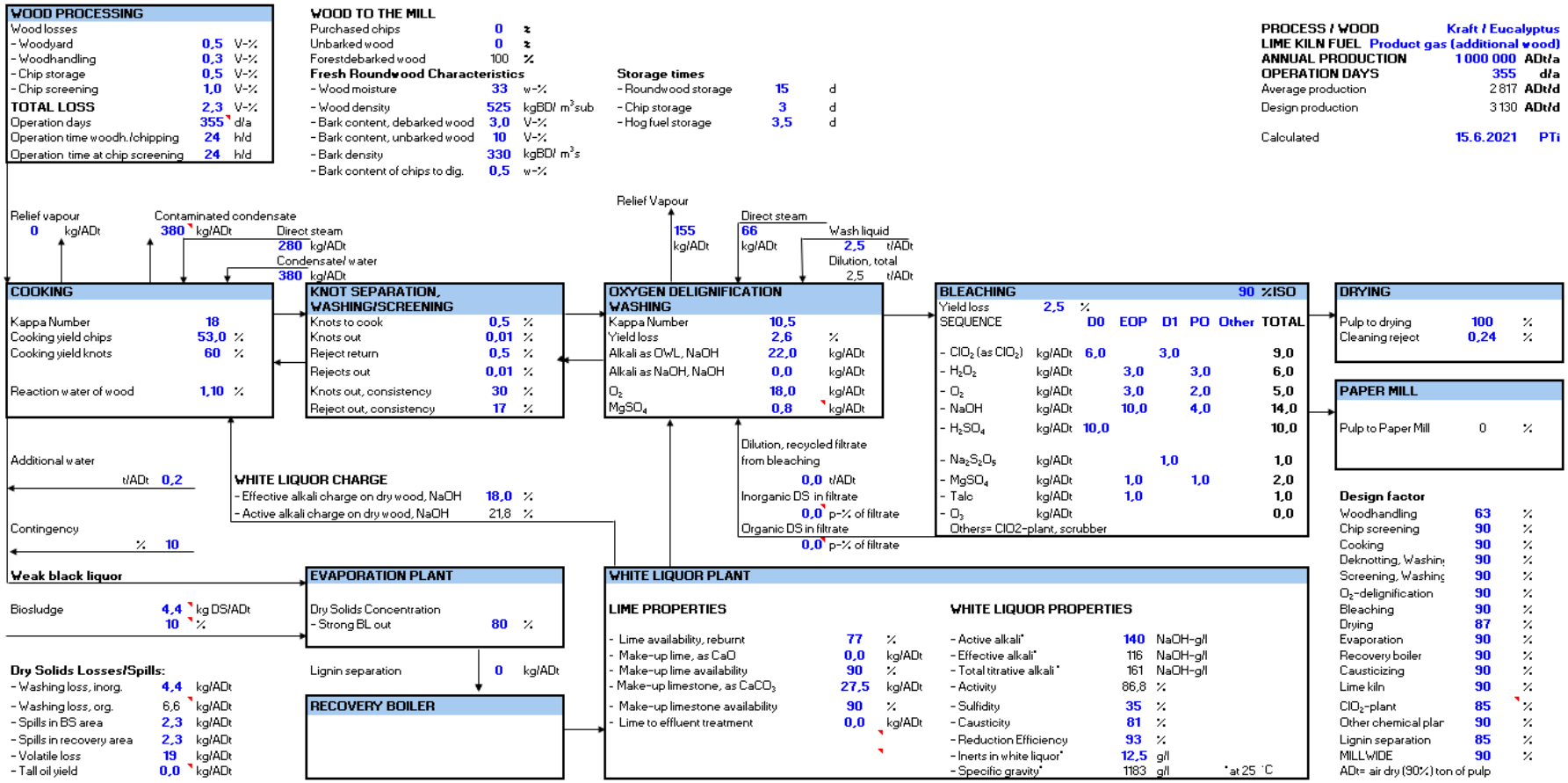


Appendix IX: Fibre- and chemical balance Mill B2b - input

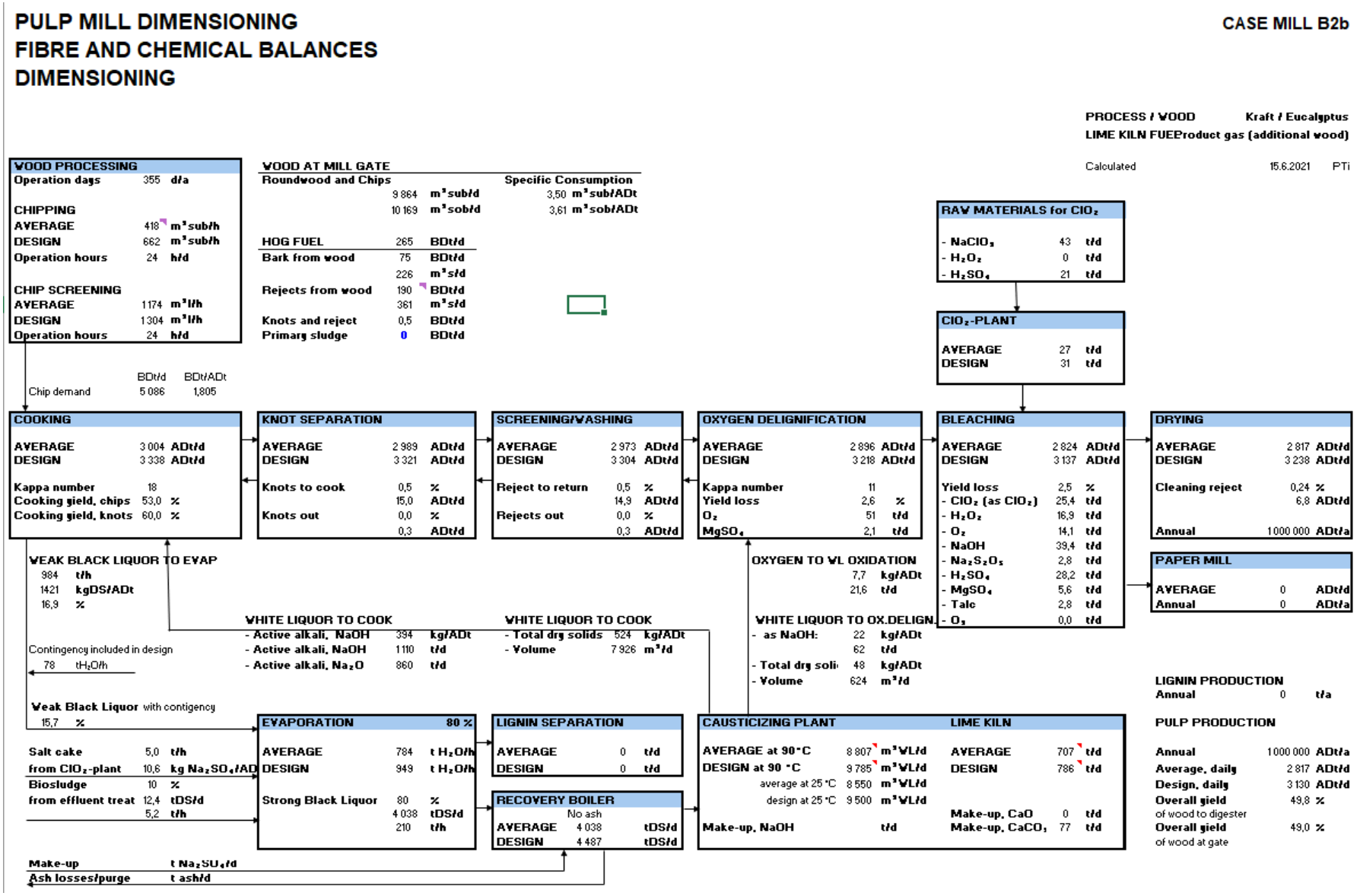
PULP MILL MAIN DIMENSIONING
FIBRE AND CHEMICAL BALANCES
INPUT VALUES

CASE MILL B2b

PROCESS / WOOD	**Kraft / Eucalyptus**	
LIME KILN FUEL	**Product gas (additional wood)**	
ANNUAL PRODUCTION	**1 000 000**	**ADt/a**
OPERATION DAYS	**355**	**d/a**
Average production	2 817	**ADt/d**
Design production	3 130	**ADt/d**
Calculated	**15.6.2021**	**PTi**



Appendix X: Fibre- and chemical balance Mill B2b - results



Appendix XI: Fibre- and chemical balance Mill B3 - input

PULP MILL MAIN DIMENSIONING
FIBRE AND CHEMICAL BALANCES
INPUT VALUES

CASE MILL B3

PROCESS / WOOD	**Kraft / Eucalyptus**	
LIME KILN FUEL	**Lignin**	
ANNUAL PRODUCTION	**1 000 000**	**ADt/a**
OPERATION DAYS	**355**	**d/a**
Average production	2 817	**ADt/d**
Design production	3 130	**ADt/d**
Calculated	**15.6.2021**	**PTi**

WOOD PROCESSING

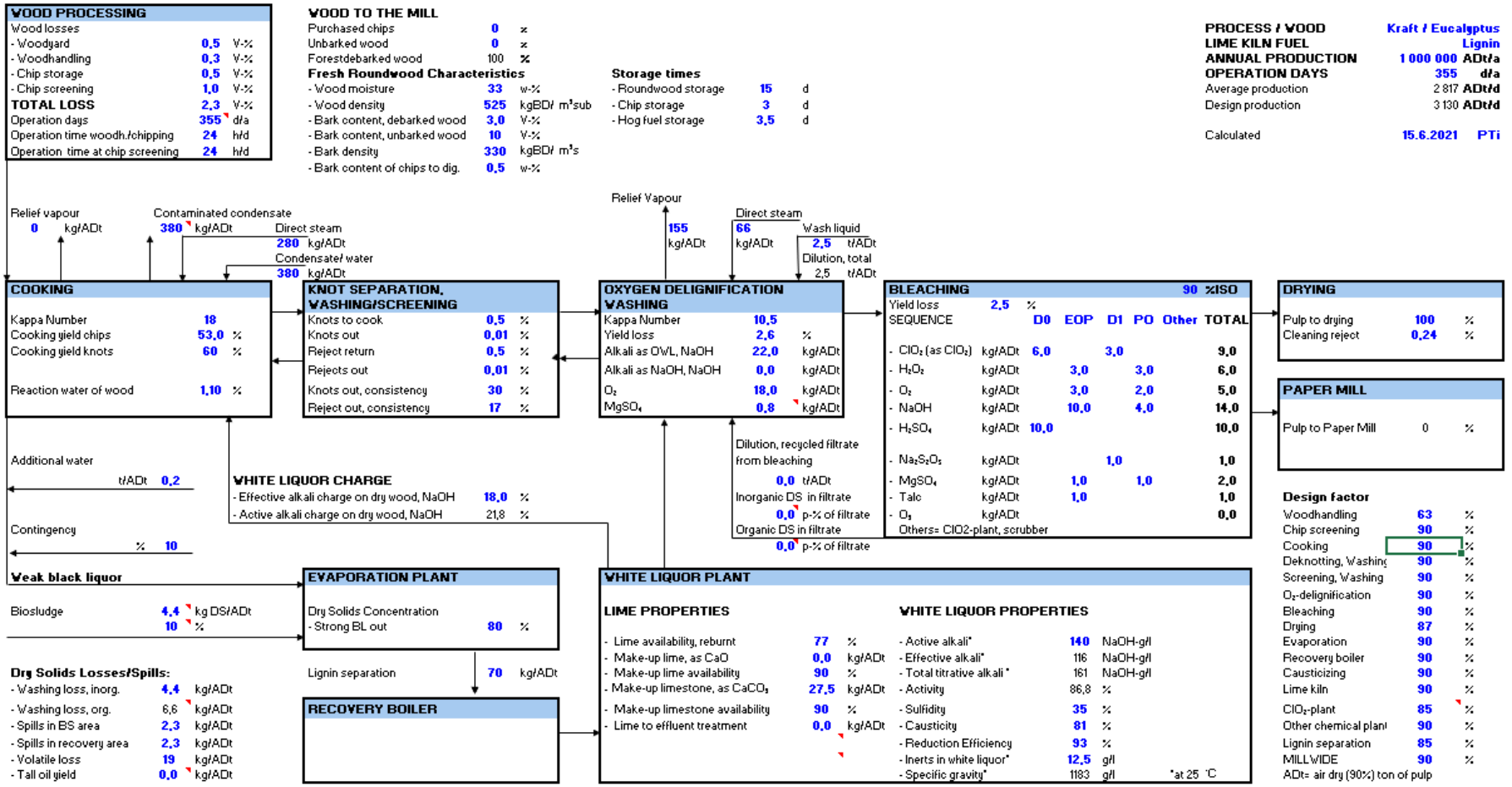
Wood losses		
- Woodyard	0,5	V-%
- Woodhandling	0,3	V-%
- Chip storage	0,5	V-%
- Chip screening	1,0	V-%
TOTAL LOSS	2,3	V-%
Operation days	355	d/a
Operation time woodh./chipping	24	h/d
Operation time at chip screening	24	h/d

WOOD TO THE MILL

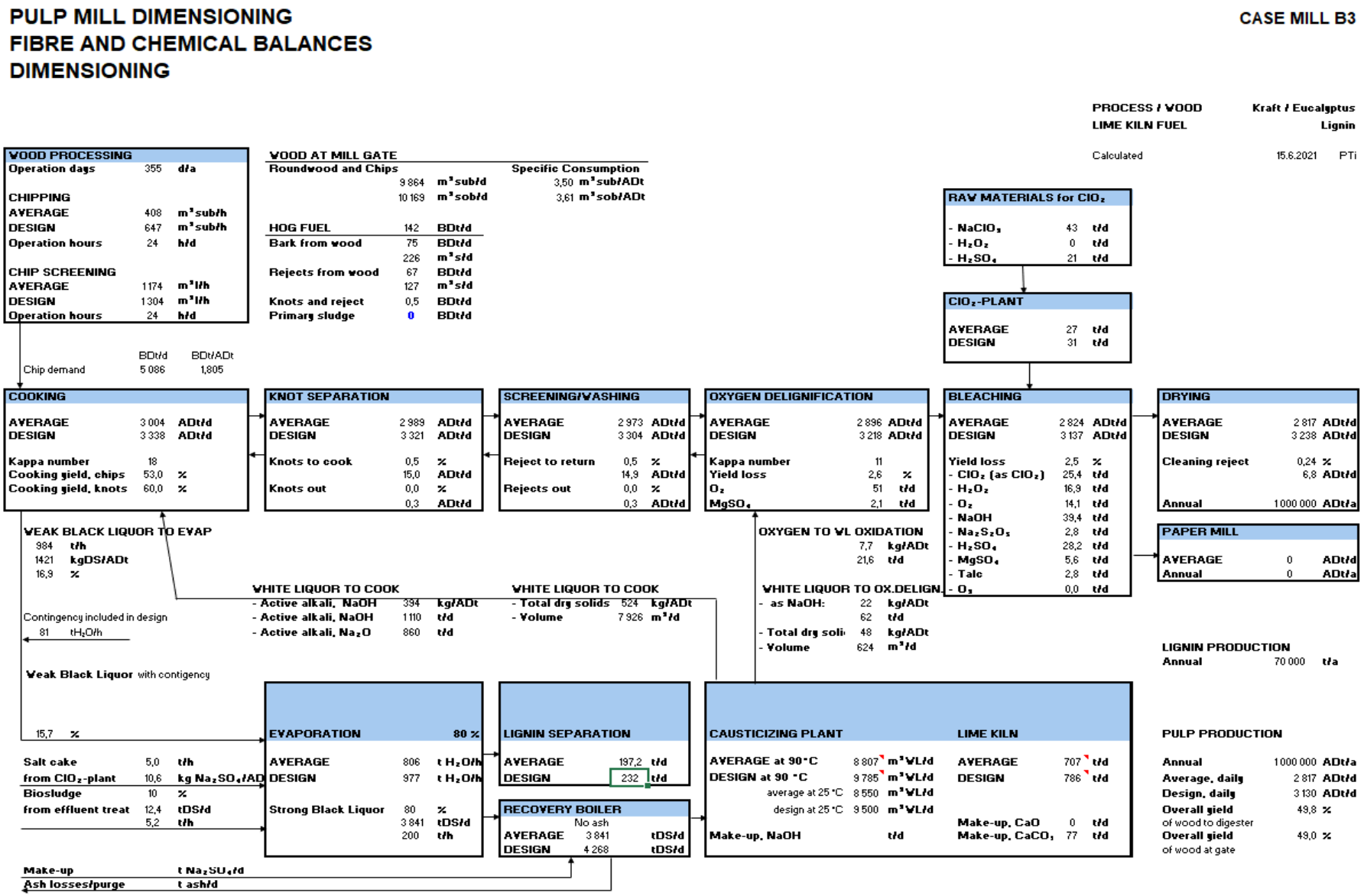
Purchased chips	0	%
Unbarked wood	0	%
Forestdebarked wood	100	%
Fresh Roundwood Characteristics		
- Wood moisture	33	w-%
- Wood density	525	kgBD/ m³sub
- Bark content, debarked wood	3,0	V-%
- Bark content, unbarked wood	10	V-%
- Bark density	330	kgBD/ m³s
- Bark content of chips to dig.	0,5	w-%

Storage times

- Roundwood storage	15	d
- Chip storage	3	d
- Hog fuel storage	3,5	d



Appendix XII: Fibre- and chemical balance: Mill B3 - results



Appendix XIII: Energy balance - Summary

PROJECT:	Master's thesis Paavo Tiitta	19.7.2021

SUMMARY

CASE		Mill A1	Mill A2a	Mill A2b	Mill B1	Mill B2a	Mill B2b	Mill B3
Wood		**SW**	**SW**	**SW**	**HW (Euca)**	**HW (Euca)**	**HW (Euca)**	**HW (Euca)**
Lime kiln fuel		**HFO**	**Product gas**	**Product gas**	**HFO**	**Product gas**	**Product gas**	**Lignin**
Bark boiler in operation		**yes**	**yes** **bark boiler**	**no** **bark is sold**	**yes**	**no additional bark**	**no additional wood**	**yes**
Specific steam consumption								
- Woodhandling	GJ/ADt	0,0	0,0	0,0	0,0	0,0	0,0	0,0
- Fibreline	GJ/ADt	4,8	4,8	4,8	4,8	4,8	4,8	4,8
- Recovery line	GJ/ADt	6,4	6,7	6,7	5,1	5,2	5,2	5,0
- Chemicals	GJ/ADt	0,4	0,4	0,4	0,1	0,1	0,1	0,1
- Miscellaneous	GJ/ADt	0,2	0,2	0,2	0,2	0,2	0,2	0,2
Total	**GJ/ADt**	**11,8**	**12,1**	**12,1**	**10,2**	**10,3**	**10,3**	**10,1**
Steam generation								
- Recovery boiler (HP)	kg/s	229,4	229,4	229,4	189,3	189,3	189,3	174,7
- Bark boiler (HP->MP1)	kg/s	36,1	19,6	0,0	9,2	0,0	0,0	9,2
- CNCG boiler (MP1)	kg/s	0,0	0,0	0,0	0,0	0,0	0,0	0,0
- Desuperheating	kg/s	1,8	1,8	1,8	1,6	1,6	1,6	1,5
Total	**kg/s**	**267**	**251**	**231**	**200**	**191**	**191**	**185**
Steam consumption								
- MP2, 29 bar	**kg/s**	**10,1**	**10,1**	**10,1**	**8,0**	**8,0**	**8,0**	**7,6**
- RB sootblowing	kg/s	10,1	10,1	10,1	8,0	8,0	8,0	7,6
- RB preheating	kg/s	0,0	0,0	0,0	0,0	0,0	0,0	0,0
- MP1, 11 bar	**kg/s**	**52,1**	**52,1**	**52,1**	**44,0**	**44,0**	**44,0**	**42,7**
- Fibreline	kg/s	19,3	19,3	19,3	15,9	15,9	15,9	15,9
- Recovery line	kg/s	31,5	31,5	31,5	27,1	27,1	27,1	25,7
- Miscellaneous	kg/s	1,3	1,3	1,3	1,0	1,0	1,0	1,0
- LP 5,0 bar	**kg/s**	**110,3**	**114,5**	**113,1**	**95,7**	**96,5**	**96,4**	**96,1**
- Woodhandling	kg/s	0,0	0,0	0,0	0,0	0,0	0,0	0,0
- Fibreline	kg/s	42,9	42,9	42,9	46,5	46,5	46,5	46,5
- Recovery line	kg/s	61,0	65,2	63,7	46,7	47,5	47,4	47,0
- Chemicals	kg/s	5,1	5,1	5,1	1,2	1,2	1,2	1,2
- Miscellaneous	kg/s	1,4	1,4	1,4	1,4	1,4	1,4	1,4
- Turbine condenser 0,03/0,07 bar(a)	**kg/s**	**94,2**	**74,0**	**55,8**	**52,1**	**42,2**	**42,2**	**38,3**
Total	**kg/s**	**270**	**253**	**233**	**202**	**193**	**193**	**187**

CASE		Mill A1	Mill A2a	Mill A2b	Mill B1	Mill B2a	Mill B2b	Mill B3
Wood		**SW**	**SW**	**SW**	**HW (Euca)**	**HW (Euca)**	**HW (Euca)**	**HW (Euca)**
Lime kiln fuel		**HFO**	**Product gas**	**Product gas**	**HFO**	**Product gas**	**Product gas**	**Lignin**
Bark boiler in operation		**yes**	**yes**	**no** **bark is sold**	**yes**	**no additional bark**	**no additional wood**	**yes**
Specific electricity consumption								
- Pulp mill	**kWh/ADt**	**655**	**643**	**625**	**558**	**553**	**553**	**563**
- Woodhandling	kWh/ADt	47,1	47,1	47,1	24,9	24,9	25,5	24,9
- Fibreline	kWh/ADt	298,4	298,4	298,4	298,1	298,1	298,1	298,1
- Recovery line	kWh/ADt	207,6	201,4	187,9	152,5	150,0	149,8	149,3
- Water/Chemicals	kWh/ADt	85,0	81,7	78,8	69,6	68,0	68,0	79,1
- Cooling tower	kWh/ADt	10,0	8,3	6,8	6,1	5,3	5,3	5,1
- Miscellaneous	kWh/ADt	6,4	6,4	6,4	6,4	6,4	6,4	6,4
Specific electricity generation								
- Turbogenerator	kWh/ADt	1704	1534	1355	1188	1099	1099	1056
Surplus pulp mill	**kWh/ADt**	**1049**	**890**	**729**	**631**	**546**	**546**	**494**
Surplus to grid	**kWh/ADt**	**1049**	**890**	**729**	**631**	**546**	**546**	**494**
Electricity consumption								
- Pulp mill	**MW**	**76,8**	**75,5**	**73,4**	**65,4**	**64,9**	**64,9**	**66,1**
- Woodhandling	MW	5,5	5,5	5,5	2,9	2,9	3,0	2,9
- Fibreline	MW	35,0	35,0	35,0	35,0	35,0	35,0	35,0
- Recovery line	MW	24,4	23,6	22,1	17,9	17,6	17,6	17,5
- Water/Chemicals	MW	10,0	9,6	9,2	8,2	8,0	8,0	9,3
- Cooling tower	MW	1,2	1,0	0,8	0,7	0,6	0,6	0,6
- Miscellaneous	MW	0,7	0,7	0,7	0,7	0,7	0,7	0,7
Total	**MW**	**76,8**	**75,5**	**73,4**	**65,4**	**64,9**	**64,9**	**66,1**
Generation								
- Turbogenerator	MW	200,0	180,0	159,0	139,5	129,0	129,0	124,0
Surplus to grid	**MW**	**123,2**	**104,5**	**85,6**	**74,1**	**64,1**	**64,1**	**57,9**