

# DROP-IN FUELS FROM BLACK LIQUOR PART STREAMS

## – BRIDGING THE GAP BETWEEN SHORT- AND LONG-TERM TECHNOLOGY TRACKS

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## PREFACE

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## SUMMARY

Reducing greenhouse gas (GHG) emissions in the transport sector requires both short and long-term interventions. An important short-to-medium term intervention is the wider deployment of drop-in biofuels that are functionally equivalent to fossil transport fuels and thus compatible with today's vehicle fleet and petroleum refining infrastructure. In Sweden, there is currently a great interest in drop-in biofuels with good GHG performance, due to the introduction of the reduction obligation for petrol and diesel fuels in 2018.

We have evaluated different pathways that can be used to produce drop-in petrol and diesel blends from part-streams of black liquor (BL), and, at the same time, debottleneck recovery boiler-limited pulp mills. The focus has been on production of liquid drop-in biofuels that can be blended with fossil fuels, with particular attention to petrol components. We evaluated five different technology pathways based on two main routes:

- 1) Lignin separation from BL and lignin liquefaction, followed by hydrotreatment to petrol and diesel, with hydrogen produced via either natural gas reforming (1a) or water electrolysis (1b),
- 2) BL gasification followed by methanol synthesis and MTG (methanol-to-gasoline) (2a). Addition of hydrogen from electrolysis to the generated syngas (2b) or of pyrolysis oil to the gasifier (2c) were also considered, for increased production scale.

In addition, we studied the impact of mill energy profile on integrated energy balances and biofuel production costs by examining three different pulp mill configurations as integration sites. The mills had contrasting energy profiles, with one being a state-of-the-art market pulp mill with an energy surplus used to produce electricity, one an integrated pulp and paper mill reliant on energy import for meeting the internal demand, and one a relatively energy-balanced market pulp mill.

Our results show that drop-in biofuels can be produced from BL part-streams with production costs of around 80 EUR/MWh (ca. 7-8 SEK/l), thereby equalling or bettering the economic performance of comparable forest residue-based fuels. The best performing pathways in the lignin separation-hydrotreatment (1a) and BL gasification-catalytic synthesis (2a) routes were found to have similar production costs. Both of those routes could thus constitute potentially attractive options for recovery boiler-limited pulp mills looking to increase pulp capacity and broaden product portfolios through comparatively modest investments. With cases 2b and 2c, we also wanted to test the hypothesis that secondary feedstocks such as hydrogen and pyrolysis oil can improve the economic performance of small-scale debottlenecking units based on BL gasification. This was, however, not proven, and is highly sensitive to future developments in the price of pyrolysis oil and electricity, as well as the capital cost of PEM electrolyzers.

While the gasification-catalytic synthesis route has a higher current technology readiness level on average, the gap is expected to shrink notably in the near future as key process steps in the separation-hydrotreatment route undergo planned demonstration in industrial conditions. However, our lignin upgrading results are subject to large uncertainties, being based on lab testing of a reference lignin in the absence of representative industrial data.

Further, the use of natural gas as hydrogen source represents the cheaper option for lignin hydro-treatment (1a), but is accompanied by GHG savings that are smaller relative to those for other pathways. This can be disadvantageous under the current Swedish quota obligation scheme for drop-in biofuels that is designed to reward biofuels with high GHG emission reductions. At the same time, the large capital cost of PEM electrolyzers makes it unlikely that the renewable-hydrogen pathway will be economically competitive in the near future, irrespective of future electricity price developments. Future availability of cheaper electrolyzers and reliable experimental data on lignin hydro-treatment are likely to impact the trade-off between fossil and renewable hydrogen in the medium term.

Substantial synergies can be obtained by implementing the technologies investigated in this project. The “dual service” that both lignin separation and gasification of a BL part stream offer gives a reduction in the overall investment cost if the pulp capacity increase is implemented together with the biofuel production. In order to assess the value of increased recovery capacity, we estimated the alternative investment cost that would be associated with a recovery boiler rebuild. This alternative cost was subtracted from the total biofuel investment cost, as it corresponds to the alternative for obtaining the same recovery capacity. This makes estimation of investments costs required in other parts of the process, e.g. the pulp line, unnecessary.

If the synergy is allocated to the biofuel production, the production costs can be reduced by significant amounts (up to 23%). If the synergy is instead allocated to the pulp production, pulp production costs can be reduced by up to 64-82 EUR/ADt pulp, which corresponds to an increased gross margin of 35-70% for the increased production volume, irrespective of biofuel production pathway. This, however, requires that the alternative cost of rebuilding the existing recovery boiler for a capacity increase is at the higher end of the investigated alternatives.

From the pulp mill’s perspective, we want to highlight two main aspects that are critical to consider in the choice between recovery boiler retrofit and BL-based biofuel production:

- 1) What would be the cost of the recovery boiler rebuild?
- 2) What is the current energy situation in the pulp mill?

The integration of biofuel production from BL part-streams results in significant impacts on the mill’s energy balance, especially in the steam system. Mills that are currently operating with an energy surplus that is exported as electricity therefore have a significant advantage as integration sites. Mills that operate their biomass or power boilers at or near capacity may have to invest in extra capacity to meet the increased demand from biofuel integration. Also other aspects of the mill’s operation will be affected by the introduction of BL-based biofuel production. The lignin separation-hydrotreatment route is less suitable for pulp mills already restricted in the evaporation section, while the gasification-catalytic synthesis route is less suitable for pulp mills already restricted in the lime kiln.

The introduction of an additional process step (lignin separation or gasification) in the recovery cycle naturally increases the risk of operational disruptions. However, the risk of impact on pulp production is considered to be limited because there is a buffer capacity in the form of tanks for both black liquor and green liquor. This allows the mill to continue the pulp production at full capacity for a limited time, as long as the majority of the recovery, which is done in the recovery boiler, is in

operation. In the event of longer operational disruptions in the new processes, the pulp production will be affected. This, however, only applies to a part of the production, corresponding to the part of the black liquor that goes to lignin separation or gasification.

The possibility to debottleneck the recovery boiler was highlighted as an important driver in a workshop on the topic of drivers for and barriers against implementation of BL part stream-based biofuel production. Conversely, barriers related to the industrial integration, e.g., the technical risk of close integration of new technologies and the need to time the potential investment into an “investment window of opportunity” when the mill or refinery is shut down for maintenance, were also highlighted.

Finally, it can be concluded that since the demand for forest residue-based drop-in alternatives that can replace both petrol and diesel is expected to grow in the future, the complementary deployment of lignin separation-hydrotreatment and BL gasification-catalytic synthesis can be a strategically interesting option for achieving deep reductions in transport GHG emissions. In order to make this happen though, challenges not only related to technology aspects, but also to lacking key actors (and associated resources), unclear roles, and weak network structures, must also be overcome.





## SAMMANFATTNING

För att minska transportsektorns utsläpp av växthusgaser krävs både kort- och långsiktiga insatser. På kort- till medellång sikt är det viktigt med drop-in-bränslen som är likvärdiga med fossila drivmedel och därmed kompatibla med dagens fordonsflotta och raffinaderiinfrastruktur. I Sverige är intresset för drop-in-bränslen med god växthusgasprestanda stort, på grund av reduktionsplikten för bensin- och dieselbränslen som infördes 2018.

I detta projekt har vi utvärderat olika teknikspår för biodrivmedelsproduktion från svartlutsdelströmmar, för att på så sätt samtidigt kunna avlasta sodapannan i massabruk som är begränsade av återvinningskapaciteten. Fokus har varit på produktion av flytande drop-in-bränslen som kan blandas med fossila drivmedel, i synnerhet bensinkomponenter. Vi utvärderade fem olika teknikspår baserade på två huvudspår:

- 1) Separation och förvätskning av lignin från svartlut, följt av vätgasbehandling till bensin och diesel, med vätgas producerat genom antingen naturgasreformering (1a) eller vatten-elektrolys (1b),
- 2) Svartlutsförgasning följt av metanolsyntes och bensinsyntes via MTG (methanol-to-gasoline) (2a). Tillsats av vätgas från elektrolys till syntesgasen (2b) eller av pyrolysolja till förgasaren (2c) beaktades också, för ökad produktionsskala.

Vi studerade också hur massabrukets energiprofil påverkar de integrerade energibalanserna och produktionskostnaderna för biodrivmedel genom att inkludera tre olika massabruk. Bruken hade olika energiprofiler, där ett är ett state-of-the-art avsalubruk med energiöverskott som används för att producera el, ett är ett integrerat massa- och pappersbruk som är beroende av energiimport för att möta det interna behovet, och ett är ett relativt energibalanserat avsalubruk.

Våra resultat visar att svartlutsdelströmmar kan användas för att producera drop-in-bränslen med produktionskostnader på runt 80 EUR/MWh (cirka 7-8 kr/l), vilket är likvärdigt med eller till och med bättre än den ekonomiska prestandan för jämförbara drivmedel från skogsrester. De bäst presterande teknikspåren för ligninseparationsspåret (1a) och svartlutsförgasningsspåret (2a) visade sig ha liknande produktionskostnader. Båda spåren kan därmed utgöra attraktiva alternativ för sodapannebegränsade massabruk som både vill öka sin massaproduktionskapacitet och bredda sin produktportfölj. Med fall 2b och 2c ville vi också testa hypotesen att tillsats av en ytterligare insatsråvara i form av vätgas eller pyrolysolja kan förbättra den ekonomiska prestandan för småskaliga svartlutsförgasare för sodapanneavlastning. Denna hypotes kunde dock inte bevisas, och resultaten är också mycket känsliga för den framtida utvecklingen av pris på pyrolysolja och el, samt kapitalkostnaden för PEM-elektrolysörer.

Även om förgasningsspåret har i genomsnitt högre teknikmognadsnivå i dagsläget, förväntas klyftan mellan teknikspåren krympa väsentligt inom en snar framtid i takt med att flera viktiga processsteg i ligninseparationsspåret genomgår planerad demonstration under industriella förhållanden. Våra resultat för ligninuppgraderingen är emellertid osäkra då de, i brist på representativa industriella data, baserats på laboratorietester av ett referenslignin.

Det är vidare betydligt billigare att använda naturgas (1a) än vattenelektrolys (1b) för framställning av vätgas för ligninuppgradering. Naturgasspåret ger däremot inte lika betydande minskningar av

växthusgasutsläpp som de andra teknikspåren. Detta kan vara ofördelaktigt under det nuvarande svenska kvotpliktssystemet för biodrivmedel, eftersom detta är utformat för att premiera biodrivmedel med hög växthusgasprestanda. Samtidigt gör de höga kapitalkostnaderna för PEM-elektrolysörer det osannolikt att elektrolysspåret kommer att kunna bli ekonomiskt konkurrenskraftigt på kort sikt, oavsett framtida utveckling av elpriset. På längre sikt kan däremot kostnadsminskningar för elektrolysörer och mer tillförlitliga experimentella data för vätgasbehandling av lignin komma att påverka avvägningen mellan fossil och förnybar vätgas.

Genom att implementera de tekniker som undersökts i detta projekt kan betydande synergieffekter uppnås. Den ”dubbla tjänst” som både ligninseparation och förgasning av en svartlutsdelström erbjuder ger en minskning av de totala investeringskostnaderna, om massaproduktionskapaciteten ökas samtidigt som biodrivmedelsproduktion introduceras. För att kunna skatta värdet av utökad återvinningskapacitet gjorde vi en uppskattning av den alternativa investeringskostnaden för en ombyggnad av den existerande sodapannan. Denna alternativa kostnad subtraherades från den totala investeringskostnaden för biodrivmedelsproduktion, eftersom den motsvarar alternativet för att erhålla samma återvinningskapacitet. På detta sätt blir det inte nödvändigt att uppskatta erforderliga investeringskostnader i andra delar av processen, t.ex. i fiberlinjen.

Om synergin allokeras till biodrivmedelsproduktionen kan produktionskostnaderna för drivmedel minskas betydligt (upp till 23%). Om synergin istället allokeras till massaproduktionen, kan massaproduktionskostnaderna sänkas med upp till 64-82 EUR per ton massa (ADt), vilket motsvarar en ökad bruttomarginal på 35-70% för den ökade produktionsvolymen, oavsett teknikspår för biodrivmedelsproduktionen. Detta kräver dock att alternativkostnaden för ombyggnad av befintlig sodapanna är i den högre änden av de undersökta alternativen.

Ur massabrukets perspektiv vill vi lyfta fram tre huvudaspekter som är viktiga att beakta i valet mellan sodapanneombyggnad och svartlutsbaserad biodrivmedelsproduktion:

- 1) Vad skulle ombyggnad av sodapannan kosta?
- 2) Vad är den nuvarande energisituationen i massabruket?

Integrering av biodrivmedelsproduktion från svartlutsdelströmmar får betydande påverkan på brukets energibalans, särskilt för ångsystemet. Bruk som för närvarande har ett energiöverskott som exporteras som el har därför en betydande fördel vad gäller integrationsmöjligheter. Bruk som redan nu driftar sin biomassapanna nära kapacitetsgränsen kan behöva investera i extra kapacitet för att möta den ökade efterfrågan från biodrivmedelsproduktionen. Introduktionen av svartlutsbaserad biodrivmedelsproduktion kommer också påverka andra aspekter av brukets verksamhet. Ligninseparationsspåret är mindre lämpat för massabruk som redan är begränsade i industnningen, medan svartlutsförgasningsspåret är mindre lämpat för bruk som redan är begränsade i mesaoombränningen.

Införandet av ytterligare ett processteg (ligninseparation eller förgasning) i återvinningscykeln ökar naturligtvis statistiskt risken för driftstörningar. Risken för påverkan på massaproduktionen anses dock vara begränsad eftersom det finns en buffertkapacitet i form av tankar för både svartlut och grönlut. Detta tillåter bruket att fortsätta massaproduktionen med full kapacitet under en begränsad tid, så länge som huvuddelen av återvinningen, som görs i sodapannan, är i drift. Vid längre driftstörningar i de nya processerna kommer massaproduktionen påverkas men det är viktigt att påpeka

att detta bara gäller en del av produktionen, motsvarande den del av svartluten som går till ligninseparation eller förgasning.

Möjligheten att avlasta sodapannan framhölls också som en viktig drivkraft i en workshop relaterad till drivkrafter för och hinder mot implementering av biodrivmedelsproduktion baserad på svartlutsdelströmmar. Omvänt framhölls också hinder relaterade till den industriella integreringen, t.ex. tekniska risker förknippade med nära integrering av ny teknik, och behovet av att en möjlig investering måste anpassas i tid till ett ”investeringsfönster” i samband med att massabruket eller raffinaderiet är stängt för underhåll.

Slutligen drar vi slutsatsen att eftersom efterfrågan på skogsrestsbaserade drop-in-alternativ som ersättning för både bensin och diesel förväntas växa i framtiden, kan både ligninseparations- och svartlutsförgasningsspåret utgöra strategiskt intressanta alternativ för betydande minskningar av transportsektorns växthusgasutsläpp. För att detta ska hända krävs dock att inte bara tekniska utmaningar utan också hinder i form av brist på nyckelaktörer (och relaterade resurser), oklara roller och svaga nätverksstrukturer övervinns.

## NOMENCLATURE/ABBREVIATIONS

Abbreviation	Definition
BL	Black liquor
BLG	Black liquor gasification
ESP	Electrostatic precipitator
GHG	Greenhouse gas
HHV	Higher heating value
HVO	Hydrogenated vegetable oil
IP	Intermediate pressure (steam)
LHV	Lower heating value
LLGO	Light light gas oil
LP	Low pressure (steam)
MeOH	Methanol
MFSP	Minimum fuel selling price
MP	Medium pressure (steam)
MTG	Methanol-to-gasoline
O&M	Operation and maintenance
PEM	Polymer electrolyte membrane (electrolysis)
PO	Pyrolysis oil
RED	Renewable energy directive
RTD	Raw tall diesel
SKKP	Smurfit Kappa Kraftliner Piteå
TCI	Total capital investment
TIS	Technological Innovation System
TRL	Technology readiness level
VGO	Vacuum gasoil

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# 1 INTRODUCTION

Reducing greenhouse gas (GHG) emissions in the transport sector requires both short and long-term interventions [1]. An important short-to-medium term (ca 5-10 years) intervention is the wider deployment of drop-in biofuels [2–5] that are functionally equivalent to fossil transport fuels and thus compatible with today’s vehicle fleet and petroleum refining infrastructure [6,7]. In Sweden, there is currently a great interest in drop-in biofuels with good GHG performance, due to the introduction of the reduction obligation for petrol and diesel fuels in 2018 [8]. For diesel, there are already well-established alternatives with relatively high GHG mitigation potential, in particular HVO from different waste feedstocks. For petrol, the alternatives are significantly fewer, as is evident in the initial reduction requirements (19.3% from diesel, compared to only 2.6% from petrol).

Efforts are currently underway to develop drop-in biofuels from lignocellulosic forest residue-based feedstocks [9–12]. One such feedstock is kraft black liquor (BL), which is a lignin-rich by-product of chemical pulping. A small fraction of BL in the form of crude tall oil is already used in biofuel (HVO) production but the maximum potential of this fraction is very limited [13]. Significantly larger potential can be found in lignin, which is the largest component in BL, and which can be upgraded to drop-in petrol and diesel blends by different pathways that can be classified into two principle production routes.

In the (*lignin*) *separation-hydrotreatment route*, around one-fifth of the lignin in the BL produced at a given mill is separated and dispatched to a petroleum refinery for hydrotreatment. Techno-economic assessments grounded on knowledge from laboratory-scale tests have found the route to be profitable over a wide range of plant sizes, but refinery-integrated processes were coarsely modelled and data quality was adversely affected by low technology readiness [14–16].

Entrained-flow gasification of BL and the subsequent upgrading of syngas to biofuels constitutes the (*BL*) *gasification-catalytic synthesis route*. BL can be co-gasified with similar forest residue-based feedstock such as pyrolysis oil (PO) to increase the amount of biofuels that can be produced from the fixed quantity of BL available at an individual mill [17]. A similar capacity increase can be obtained by co-processing syngas with electrolysis hydrogen in a gasification-electricity hybrid pathway [18–20]. Whether the increase in biofuels yield can also translate into better economic performance has not been investigated for drop-in biofuels.

The economics of drop-in biofuels can be enhanced by co-locating and integrating production with petroleum refineries, thereby providing valuable access to hydrogen [6,21]. The availability of cheap, ideally renewable, hydrogen is a key challenge for future development of drop-in biofuels from oxygen-rich lignocellulosic biomass feedstocks [21]. The lack of, and need for, representative experimental data on the hydrogen consumption and hydrocarbon product yields of different drop-in biomass-based feeds has been highlighted recently [22].

In chemical pulping, the lignin in BL is typically combusted in a recovery boiler, which at some pulp and paper mills can be a bottleneck to capacity expansion. Redirecting some of the lignin to a biofuel plant, based on either separation-hydrotreatment or gasification-catalytic synthesis, frees capacity in the recovery boiler, which can be used to increase the production of pulp, assuming no other capacity constraints are in effect. The increased pulping capacity can, in conjunction with the

sale of produced biofuels, translate into significant additional revenue. It has been shown that relatively small BL gasification units, which complement rather than replace recovery boilers, may potentially also be economically competitive provided they are integrated with a pulp mill boasting a large energy surplus [16]. Such units are associated with lower technical risk and smaller investment size, two factors that have been identified as barriers to commercial deployment [23]. Concepts based on a stepwise change of existing processes and systems are also more likely to attract investment capital than more large-scale adjustments, such as total replacement of the recovery boiler [24].

## 1.1 PROJECT OBJECTIVES

The overall aim of the project is to evaluate and compare different biofuel production pathways that can be used to produce drop-in petrol and diesel blends from part-streams of BL and thereby debottleneck pulp production capacity at recovery boiler-limited pulp mills. Focus is on production of liquid drop-in biofuels that can be blended with fossil fuels, with particular attention to petrol components. Different technology pathways are evaluated based on the two main routes (*lignin*) *separation-hydrotreatment* and *BL gasification-catalytic synthesis*, respectively. In addition, the impact of mill energy profile on integrated energy balance and biofuel production cost is studied by examining three different pulp mill configurations with contrasting energy profiles as integration sites.

Specific objectives of this report are to:

- 1) Evaluate the five technology pathways and quantify their performance regarding economics, energy efficiency, greenhouse gas emission performance, and technology maturity (TRL).
- 2) Evaluate the technical and economic effects for pulp mills, including the effects of changed pulp production capacity and changed mass and energy balances, and develop a practical basis for the pulp industry to estimate the value of increasing the pulp mill recovery capacity through the implementation of pulp mill integrated biofuel production from black liquor part-streams.
- 3) Provide an overview of general driving forces for and key barriers (technical, economic, organizational, etc.) to actual implementation of the studied technology pathways, which includes and builds on relevant industrial players' perceptions of different drivers and barriers.

## 1.2 REPORT OUTLINE

This report is divided into three main chapters – one per objective. Each chapter contains a description of methods and data used, a summary of the results related to the specific objective, and a brief summary of the main take-aways. Overall conclusions and recommendations are given in the report's final chapter.



### 1.3 DROP-IN BIOFUEL PATHWAYS

Two of the studied pathways belong to the *separation-hydrotreatment route*. These are designed to investigate enviro-economic trade-offs between the use of renewable and fossil hydrogen in refinery-integrated co-processing of fossil and biomass-based feeds. Three pathways belong to the *gasification-catalytic synthesis route*. These are chosen in order to test the hypothesis that supplementing BL with secondary feedstocks such as PO and hydrogen can improve the economic case for recovery boiler debottlenecking plants.

Table 1 lists the pathways evaluated in this study, classified by production route. Each pathway can be broken down into two stages. In the first stage, a part-stream of BL from the pulp mill is upgraded to an intermediate product: stabilised methanol in the gasification-catalytic synthesis route, and a stabilised lignin mixture in the separation-hydrotreatment route. In the second stage, the intermediate product is upgraded to transport fuel blends at the petroleum refinery. The pathways are described in more detail in Appendix A.

**Table 1. Examined biofuel pathways.**

	Pathway	Feedstock(s)	Production Route	Product(s)
1a <sup>a</sup>	Lignin	Black liquor (lignin)	Separation-Hydrotreatment	Petrol & Diesel
1b <sup>b</sup>	Lignin+electrolysis	Black liquor (lignin)	Separation-Hydrotreatment	Petrol & Diesel
2a <sup>c</sup>	Black liquor gasification (BLG)	Black liquor	Gasification-Catalytic Synthesis	Petrol & LPG
2b <sup>d</sup>	BLG+electrolysis	Electricity (hydrogen) & black liquor	Gasification-Catalytic Synthesis	Petrol & LPG
2c <sup>e</sup>	BLG+pyrolysis oil	Pyrolysis oil & black liquor	Gasification-Catalytic Synthesis	Petrol & LPG

<sup>a</sup> Lignin extracted from kraft BL by membrane separation is stabilized, purified and sent to an oil refinery for hydrotreatment and upgrading to petrol and diesel blendstock.

<sup>b</sup> Identical to 1a except that the hydrogen used for hydrotreatment is produced by water (PEM) electrolysis.

<sup>c</sup> Syngas from entrained-flow gasification of kraft BL is upgraded to drop-in petrol via the MTG (methanol-to-gasoline) process at a petroleum refinery.

<sup>d</sup> Syngas from entrained-flow gasification of kraft BL is mixed with hydrogen from water (PEM) electrolysis and upgraded first to methanol and then to petrol.

<sup>e</sup> A variant of 2a that uses blends of kraft BL and imported PO as feedstock.

## 2 BIOFUEL PRODUCTION PERFORMANCE EVALUATION

This chapter relates to the following project objective:

1. Evaluate the five technology pathways and quantify their performance regarding economics, energy efficiency, greenhouse gas emission performance, and technology maturity (TRL).

### 2.1 METHODS AND DATA

The text presented here is a condensed version of the methodology described in [25].

#### 2.1.1 Study design

Three pulp mill configurations with different production capacities and energy requirements were used as integration sites for production of biofuel intermediates (see Appendix B). *Model Mill* is a simulation model representing a state-of-the-art market pulp mill with an energy surplus. *SKKP* (Smurfit Kappa Kraftliner Piteå) is a Swedish integrated pulp and paper mill reliant on energy import for meeting the internal demand. *Södra* (Södra Cell Mörrum) is a Swedish market pulp mill that is able to meet its own energy needs without fuel import but does not generate a noteworthy energy surplus.

The final upgrading to transport biofuels is co-located and integrated with a refinery modelled on Preem Oil Refinery, Lysekil, where petrol, diesel, LPG and various grades of fuel oil are produced. The Lysekil refinery has a crude oil refining capacity of 11.4 Mt per year with a gross energy demand in excess of 400 MW<sub>th</sub>, which is supplied by internally produced energy gases and externally purchased natural gas. The hydrogen supplied for process use in the refinery is principally produced by steam reforming of natural gas.

We applied a scenario in which the pulping capacity is increased by 18.5% at each mill. This was achieved by directing 18.5% of the BL dry solids flow to the biofuel plant, while keeping the thermal load on the recovery boiler the same as under normal operation. The amount of energy extracted from the recovery boiler for use as input to biofuel production was therefore the same for all pathways at a given mill. However, the presence of secondary feedstocks meant that both 2b (electricity) and 2c (PO) had a higher total feedstock input than 2a. Table 2 summarises the feedstock inputs for the studied pathways.

**Table 2. Feedstock inputs to biofuel production units for each mill.**

	Units	Model Mill	SKKP	Södra
Recovery boiler capacity	tons dry solids BL/d	3760	2000	2700
BL to biofuel plant <sup>a</sup>	tons dry solids/d	695	370	500
BL to biofuel plant <sup>b</sup>	MW HHV dry basis	103.7	56.3	70.8
Lignin in BL to biofuel plant <sup>c</sup>	tons dry solids/d	348	189	238
Lignin in BL to biofuel plant <sup>d</sup>	MW HHV dry basis	103.7	56.3	70.8
PO to biofuel plant <sup>e</sup>	tons dry solids/d	160	92.7	115
PO to biofuel plant <sup>f</sup>	MW HHV dry basis	43.1	24.9	30.8
Hydrogen to biofuel plant	MW HHV	58.4	24.5	37.3

<sup>a</sup> Applicable to gasification pathways (2a, 2b, 2c). Equals 18.5% of the BL input to the recovery boiler at each mill.

<sup>b</sup> HHVs (d.b.) for BLs from Södra, SKKP and Ref Mill are 12.2 MJ/kg, 13.1 MJ/kg and 12.9 MJ/kg, respectively.

<sup>c</sup> Application to lignin pathways (1a, 1b).

<sup>d</sup> HHV (d.b.) for lignin is 27.8 MJ/kg [26].

<sup>e</sup> The fraction of PO in the PO/BL blend evaluated in 2c was set at 20 wt.%, as this is the highest fraction that has been verified in pilot-scale gasification experiments [27].

<sup>f</sup> Based on a PO HHV (d.b.) of 23.2 MJ/kg [28].

### 2.1.2 Process modelling

Integrated material and energy balance models were produced for each pathway using best available data supplemented by expert input. This section describes the central modelling choices and assumptions, with details given in [25].

#### The separation-hydrotreatment route

The material and energy balances for the separation-hydrotreatment route were based on data provided by SunCarbon AB and Preem AB, respectively. The weak BL fed to the membrane separation was taken from the evaporator train after the removal of fatty acids and extractives. Based on results from pilot-scale membrane separation experiments, the retentate stream was modelled as containing 80% of the lignin and 100% of the non-lignin organic compounds in the weak BL feed. Heat treatment, purification and stabilization of the retentate stream was carried out using IP (intermediate pressure) and LP (low pressure) steam. Lignin was delivered to the refinery in a 50/50 lignin/VGO mixture, which was further diluted with VGO to reduce the oxygen content for processing in the two-step iso-cracker.

Lignin hydrotreatment was modelled on data from lab-scale tests of a reference lignin, which, unfortunately, came from a different source than the lignin used for modelling the separation stage and using other carriers (LLGO, light light gas oil, and RTD, raw tall diesel). The hydrogen consumption and yields from lignin were calculated through a subtraction approach. The yield structure for the hydrocracking of the heavy ends from the lignin hydrodeoxygenation was based on VGO yield structure, as hydrocracking data for lignin was not available. Potential shifts in fossil product distributions as a consequence of co-processing were ignored.

This convoluted approach reflects the present deficiency of knowledge on lignin co-processing and hydrotreatment in industrially relevant conditions (see Section 2.2.2 for further discussion). Results are therefore subject to large uncertainties and should be interpreted with caution.

### The gasification-catalytic synthesis route

The gasification-catalytic synthesis route was based on the BLG technology developed by Chemrec AB [29]. Energy and mass balance data was generated by modelling using SIMGAS (a MATLAB-based tool for thermodynamic equilibrium calculations) [30,31] and Aspen Plus™ 8.4. The modelled process consists of an oxygen-blown entrained-flow reactor pressurized at 30 bar, followed by a water-gas-shift reactor, acid gas removal (amine wash), methanol synthesis, and partial distillation (“stabilization”). The yield structure for MTG products was taken from [32] who, in turn, based their work on [33,34]. Hydrogen (pathway 2b) was produced by PEM electrolysis of water with an electricity-to-product efficiency of 80% on a HHV basis [35]. The oxygen produced as a by-product of electrolysis was sent to the gasifier.

### Material and energy integration with pulp mill and refinery

In lignin separation-hydrotreatment, the inorganic constituents of the permeate stream and the hemicelluloses in the lean BL were returned to the mill evaporation unit. The sulphuric acid containing wash water stream from the lignin purification unit was sent to the mill and mixed with lean BL. The resulting increase in the amount of sulphur in the mill recovery cycle was mitigated by increasing the purge rate of electrostatic precipitator (ESP) ash from the recovery boiler. Additional sodium hydroxide was added to the recovery cycle to make up for the resulting loss of sodium that left together with sulphur. Other points of integration included the supply of IP and LP steam, and the combustion of sulphur-rich gases released in the lignin separation step.

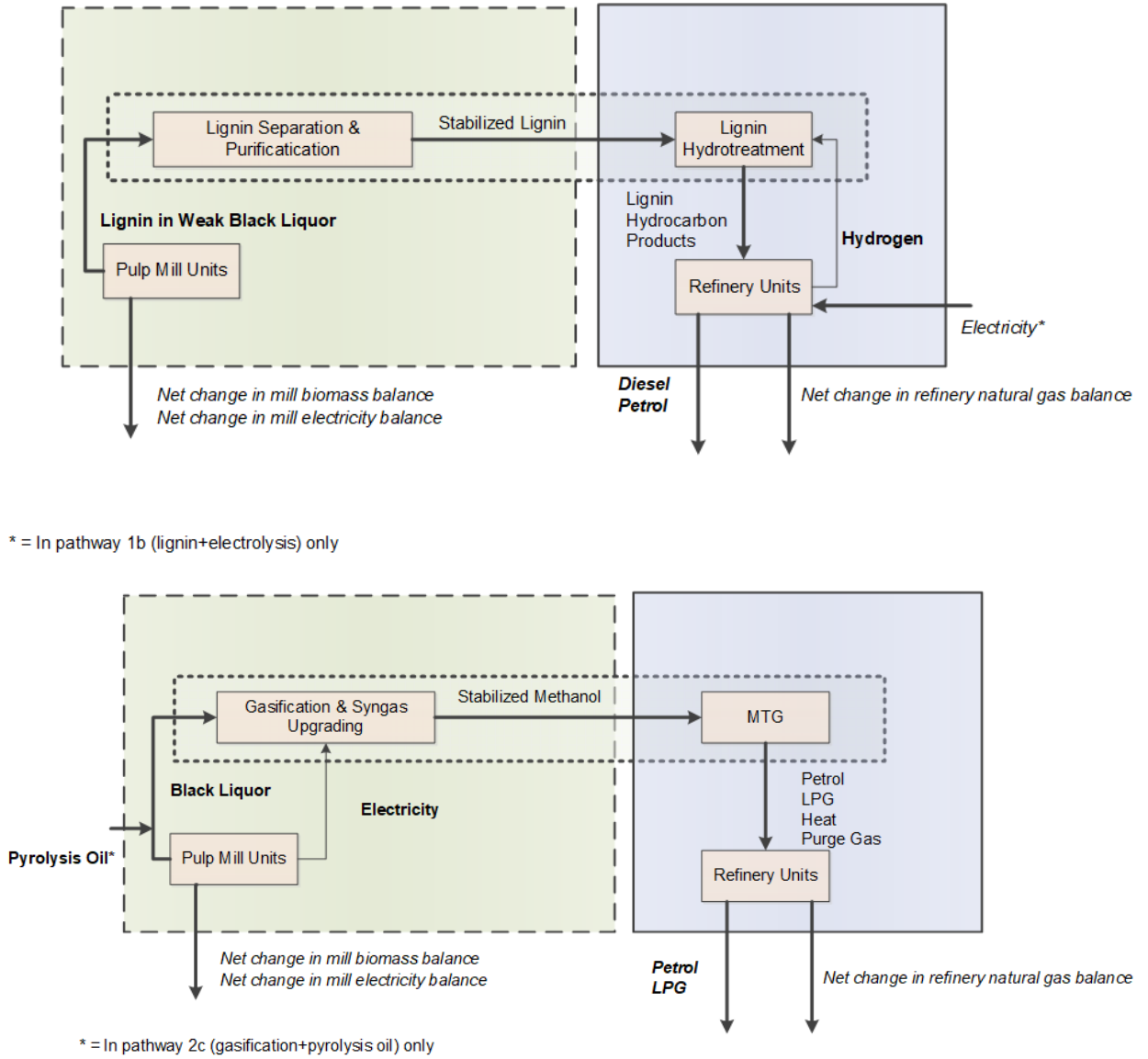
In gasification-catalytic synthesis, the hydrogen sulphide separated in the amine wash was returned to the recovery boiler to avoid affecting the overall mill chemical balance. Purge gas from methanol synthesis was fired in the mill lime kiln. Electricity was imported from the mill. LP and IP steam were exported from the biofuel units to the mill.

### Material and energy integration with petroleum refinery

All of the purge gas and 25% of the heat recovered from the MTG reactors were assumed to replace an equivalent amount of energy from natural gas. The hydrogen used for the hydrotreatment of lignin was produced at the refinery by steam reforming of natural gas in 1a, and by PEM electrolysis 1b. Heat and fuel gases from lignin hydrotreatment replaced the natural gas used for steam generation at the refinery in a ratio of 1:1 on an energy basis. The final upgrading of biofuel blend components was carried out at the refinery.

## **2.1.3 Energy efficiency assessment**

Two different measures of efficiency were used to quantify energy performance, as illustrated in Figure 1.



**Figure 1. Simplified schematic of system boundaries and cross-boundary flows in (i) lignin separation and hydrotreatment (top), (ii) gasification-catalytic synthesis (bottom) routes. The biofuel production process is delineated by dotted lines. The pulp mill and petroleum refinery are delineated by dashed and solid lines, respectively. Streams labelled in bold are used for calculating system efficiency. Streams labelled in italics are used to calculate expanded system efficiency. See text for definitions.**

A system efficiency  $\eta_{\text{system}}$  that considers all primary energy inputs to biofuel production units is defined in Eq. 1, where  $E_{\text{Primary-Inputs}}$  corresponds to the energetic value of all primary energy inputs.

$$\eta_{\text{system}} = E_{\text{Product}} / E_{\text{Primary-Inputs}} \quad (\text{Eq. 1})$$

The system boundary in eq. 1 is drawn around the biofuel production process only, as shown in Figure 1. To account for the changes in the overall energy balance of the mill and the refinery resulting from the integration of biofuel production, an expanded system efficiency  $\eta_{\text{system-exp}}$  is defined in eq. 2 where  $E_{\text{Integrated-Energy-Inputs}}$  is the net energetic value of integrated energy inputs/products.

$$\eta_{\text{system-exp}} = E_{\text{Product}} / E_{\text{Integrated-Energy-Inputs}} \quad (\text{Eq. 2})$$

The primary energy inputs and integrated energy inputs/products used for efficiency calculations are listed in Table 3. As per the definition of  $\eta_{\text{system-exp}}$  the feedstock for both lignin and gasification pathways is not BL or BL lignin, but the net change in mill biomass and electricity balance. Similarly, in addition to biofuel products, the net change in refinery natural gas balance constitutes a secondary product. Note that the natural gas reformer supplying hydrogen in 1a is outside the expanded system boundary, while the electrolyser used for hydrogen production in 1b is within.

**Table 3. Inputs and outputs for efficiency calculations.**

	Primary Energy Inputs [ $\eta_{\text{system}}$ ]	Integrated Energy Inputs/Products [ $\eta_{\text{system-exp}}$ ]	Products [ $\eta_{\text{system}}$ ] [ $\eta_{\text{system-exp}}$ ]
1a	Lignin in weak black liquor <sup>a</sup> , hydrogen	Hydrogen; net change in: mill biomass balance, mill electricity balance, refinery natural gas balance	Petrol, diesel
1b	Lignin in weak black liquor <sup>a</sup> , electricity (hydrogen)	Electricity (for hydrogen); net change in: mill biomass balance, mill electricity balance, refinery natural gas balance	Petrol, diesel
2a	BL, electricity (utilities) <sup>b</sup>	Net change in: mill biomass balance, mill electricity balance, refinery natural gas balance	Petrol, LPG
2b	BL, electricity, hydrogen <sup>b</sup>	Net change in: mill biomass balance, mill electricity balance, refinery natural gas balance	Petrol, LPG
2c	BL, PO, electricity	Pyrolysis oil; net change in: mill biomass balance, mill electricity balance, refinery natural gas balance	Petrol, LPG

<sup>a</sup> Calculated as the difference in lignin content between weak black liquor and lignin-deficient liquor

<sup>b</sup> The relatively small quantities of electricity and hydrogen used in the MTG unit are ignored.

#### 2.1.4 Economic evaluation

We evaluated the economic performance by calculating a minimum fuel selling price (MFSP) using the discounted cash flow rate of return method. The total capital investment (TCI) estimate for each pathway was in large part based on commercial nth-plant (mature technology) cost estimates at different levels of granularity, and scaled to the relevant size. The cost composition and scaling exponents of individual process units can be found in the Supplementary Material to [25]. The PEM electrolyser installed investment cost was set at 1500 EUR/kW<sub>e</sub> in line with the findings of a comprehensive recent cost review [36]. The contingency for cost escalation was fixed at 30% of fixed capital investment. To reflect arrangements likely applied in practice and to achieve better economies-of-scale, MTG units were sized to provide five times the capacity of the mill biofuel units. A similar arrangement was employed for the hydrotreatment unit in the lignin pathways, which was sized to treat 761 tons of liquefied lignin per day corresponding to two or three pulp mill equivalents, depending on the mill.

TCI was annualized using a capital recovery factor of 0.102, which was calculated assuming a real discount rate of 8% over a plant lifetime of 20 years. O&M expenditure was fixed at 4% of TCI inclusive contingency. A plant availability factor of 90% was used to calculate annual biofuel production. As nth-plant technology has been assumed, an availability factor commensurate with that of a recovery boiler was deemed appropriate. The prices of energy carriers and other materials that fall under operating expenditure (OPEX) are given in Table 4. Gasifier oxygen was assumed to be imported except in 2b, where the majority of the demand was met by oxygen from the electrolyser. It was assumed, optimistically, that the replacement of the PEM stack due to performance degradation is carried out once over the economic lifetime of the electrolyser, with the cost set at 60% of installed equipment cost and included in OPEX [35].

**Table 4. Energy and material prices.**

Bark	[EUR/MWh]	14.6	Average price for Swedish forest industry by-products in 2017.
Forestry Residue	[EUR/MWh]	19.1	Average Swedish price in 2017. Transport costs are included. Excludes taxes. Woodchips from both coniferous and deciduous harvesting residues.
Electricity - Imported	[EUR/MWh]	33.5	Electricity price for industrial customers. Includes production cost, network cost and electricity tax but not VAT. Excludes the cost of electricity certificates (7 EUR/MWh). [37]
Electricity - Exported	[EUR/MWh]	31.8	Average spot price for south-central Sweden in 2017. Does not include electricity certificate, energy tax, VAT, network cost or markup. [38]
Oxygen	[EUR/kg]	0.07	Taken from Andersson et al. [39]
Carbon dioxide	[EUR/kg]	0.10	RISE in-house estimate
Sodium Hydroxide	[EUR/kg]	0.50	RISE in-house estimate
Sulfuric Acid	[EUR/kg]	0.10	RISE in-house estimate
Solid Waste (ESP ash disposal)	[EUR/kg]	0.10	RISE in-house estimate
Additive (Lignin Product)	[EUR/kg]	6.00	SunCarbon AB in-house estimate
Zinc Oxide	[EUR/kg]	2.5	Average spot price for zinc in 2017.
Catalyst (Syngas Upgrading)	[EUR/kg methanol]	0.0011	Taken from Jafri et al. [16]
Pyrolysis Oil	[EUR/MWh]	76.0	Average of the low scenario and high scenario in "Building up the future - Cost of Biofuel" by Maniatis et al. [40]
Natural Gas	[EUR/MWh]	45.3	Average price for Swedish class I5 customers in July-December 2018. Includes the cost of natural gas, net, tax (after reduction for repayment of energy and carbon tax). Excludes general sales tax.
Hydrogen	[EUR/MWh]	54.3	Calculated as 3.564 times the price of natural gas produced using steam-methane reforming, on a mass basis. The multiple represents the average of a low and a high estimate in a previous study [16]. For steam reforming with PSA, efficiency 75-80%. IRR = 10%
Diesel Gross Margin	[EUR/MWh]	0.135	Average margin in 2016 [41]
Petrol Gross Margin	[EUR/MWh]	0.201	Average margin in 2016 [41]

### 2.1.5 GHG footprint assessment

GHG footprints were estimated using a simplified approach based on the Renewable Energy Directive (RED) guidelines [42]. Since RED prohibits the allocation of GHG emissions to heat even when used a co-product, such as in the case of natural gas replacement at the refinery, GHG footprints were also calculated using the principle of system expansion. The allocation of emissions on the basis of system expansion is advocated in ISO 14044 to account for the benefits associated with the replacement of fossil products and services [43]. To quantify such benefits, an alternative estimation of GHG footprints was also carried out, which assumed that the heat released from the hydrotreatment of lignin and from the upgrading of methanol to petrol can substitute for natural gas at the refinery on a 1:1 basis. The emission factors used as inputs are listed in Table 5.



**Table 5. Emission factors used as inputs to GHG footprint estimations.**

Input	GHG footprint [gCO <sub>2</sub> eq/MJ LHV]	Comments [Source]
Forest residues	2.2	Emissions associated with “typical” technology and transport distance. 1a (lignin) and 1b (lignin+electrolysis) only [44].
Hydrogen	87.3	Emissions associated with steam methane reforming of natural gas (75g CO <sub>2</sub> eq/MJ, [45]) and natural gas supply (12.3 CO <sub>2</sub> eq/MJ, estimate) 1a (lignin+electrolysis) and 2b (BLG+electrolysis) only.
Electricity	13.1	Swedish electricity mix in accordance with Swedish Energy Agency recommendation. All pathways [46].
Natural gas	67.0	LCA emissions in combustion applications [47].
Petrol	93.5	Used as fossil petrol reference [48].
Diesel	95.5	Used as fossil diesel reference [48].

### 2.1.6 Technology maturity evaluation

The technology maturity of the examined pathways on the technology readiness level (TRL) scale using two different approaches with contrasting but complementary perspectives: (a) the weighted average approach, and (b) the weakest link approach. Process configurations were broken down into smaller steps. Each step was assigned a weighting based on importance and complexity and a TRL used based on the definitions provided by the European Commission [49] and the US Department of Energy [50]. In the weighted average approach, the weighted scores of all steps were added up to calculate the overall score. In the weakest link approach, the lowest score for a step with a weighting > 0.2 was used as the overall score. The approaches have been discussed in greater detail in a previous study [16].

## 2.2 TECHNO-ECONOMIC PERFORMANCE

The text presented here is an condensed version of the results from [25].

### 2.2.1 Energy efficiency

Table 6 shows the resulting energy balances from the modelled biofuel production, with detailed integrated balances given in Appendix C. All pathways integrated with a given mill use the same quantity of BL or BL lignin, which amounts to 104 MW, 71 MW and 56 MW for Model Mill, Södra and SKKP, respectively (see Table 2 for equivalent mass flows)

Diesel products, with nearly negligible amounts of petrol, dominate the final products from the lignin separation pathways (1a, 1b). The total consumption of hydrogen equals 0.148 kg per kg of biofuel product, which is significantly more than the expected theoretical consumption if hydrotreatment is assumed to be limited to hydrogenolytic depolymerisation and hydrodeoxygenation only. The excess can be explained by the noteworthy quantities of fuel gases that are also produced according to the experimental data used to assess the process performance. These gases were assumed to be combusted to furnish the refinery with renewable heat. As described in Section 2.1.2, the yield structure for lignin hydrotreatment was assembled from data embedded with large uncertainties. Experiments in industrial conditions with more representative lignin blends are planned, which may return a different product distribution.



In contrast, the dominant product in the BL gasification pathways (2a, 2b, 2c) is petrol with LPG as a secondary co-product. The use of electrolysis hydrogen (2b) or a 20 wt.% PO/BL blend (2c) as supplementary feedstock leads to an increase in biofuel yield by 75% and 56%, respectively.

The steam balance shows that the lignin separation route is a net importer of steam from the mill. Steam is consumed during several process steps co-located at the mill such as heat treatment, purification and stabilization. As there are no significant temperature gradients, the potential for heat recovery for steam generation is limited. Conversely, the biofuel production units in the BL gasification route generate a steam surplus since significant quantities of heat can be recovered during methanol synthesis and the initial cooling of syngas after the gasification reactor.

**Table 6. Energy balance for biofuel production units. Integration effects on overall mill and refinery energy balances are excluded.**

		Model Mill					Södra					SKKP				
Pathway		1a	1b	2a	2b	2c	1a	1b	2a	2b	2c	1a	1b	2a	2b	2c
<b>Inputs</b>																
<b>Biofuel Feedstock</b>																
Black liquor	MW <sub>th</sub> HHV			104	104	104			71	71	71			56	56	56
Black liquor lignin	MW <sub>th</sub> HHV	104	104				71	71				56	56			
Pyrolysis oil	MW <sub>th</sub> HHV					43					31					25
Electricity (hydrogen)	MW <sub>e</sub>				58					37					24	
<b>Others</b>																
Electricity (process) <sup>a</sup>	MW <sub>e</sub>		106	3.8	5.4	5.8		72	2.5	3.5	3.9		57	2.0	2.6	3.2
Hydrogen (process)	MW <sub>th</sub> HHV	84					58					46				
LP Steam <sup>b</sup>	MW <sub>th</sub>	53	53				36	36				29	29			
IP Steam <sup>b</sup>	MW <sub>th</sub>	4.4	4.4				3.0	3.0				2.4	2.4			
<b>Outputs</b>																
<b>Biofuel Products</b>																
Petrol	MW <sub>th</sub> HHV	1.4	1.4	43	75	67	1.0	1.0	28	48	45	0.8	0.8	22	35	36
Diesel	MW <sub>th</sub> HHV	84	84				58	58				46	46			
LPG	MW <sub>th</sub> HHV			5.0	8.7	7.8			3.2	5.6	5.2			2.6	4.1	4.2
<b>Others</b>																
Fuel gases <sup>c</sup>	MW <sub>th</sub> HHV	70	70				48	48				38	38			
MeOH purge <sup>d</sup>	MW <sub>th</sub> HHV			3.6	4.1	5.3			2.3	2.7	3.6			1.9	2.1	2.9
MTG purge <sup>c</sup>	MW <sub>th</sub> HHV			0.8	1.4	1.3			0.5	0.9	0.9			0.4	0.7	0.7
Heat <sup>c</sup>	MW <sub>th</sub>	32	32	2.9	5.0	4.5	22	22	1.8	3.2	3.0	17	17	1.5	2.4	2.4
LP Steam <sup>b</sup>	MW <sub>th</sub>			3.5	9.0	2.5			2.8	6.2	2.1			4.0	6.1	3.5
IP Steam <sup>b</sup>	MW <sub>th</sub>			2.9	5.0	4.9			1.8	9.4	3.2			1.7	6.9	3.0

<sup>a</sup> Includes electricity for hydrogen production by water electrolysis in 1b.

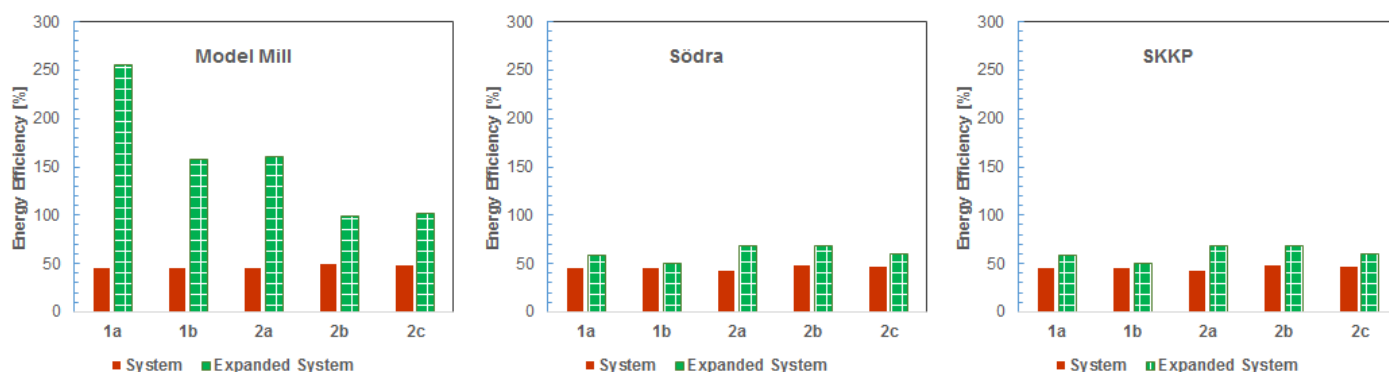
<sup>b</sup> LP and IP steam are exchanged at 3.5-4 bar(g) and 25-28 bar(g) depending on mill. There is a small misalignment (0.5-2 bar) between biofuel plant and mill steam levels, which would be fully harmonized in a physical plant.

<sup>c</sup> From lignin hydrotreatment, used to replace natural gas at the refinery.

<sup>d</sup> To lime kiln.

Figure 2 shows the resulting energy efficiencies. Detailed energy balance models can be found in the Supplementary Material of [25]

System efficiencies vary between 45% and 50%, which indicates that the energy performance of lignin separation-based and BL gasification-based pathways is relatively similar when evaluated on the basis of primary energy inputs. The incremental efficiency of biofuel production from the secondary feedstocks is 61% for BLG+electrolysis (2b) and BLG+pyrolysis oil (2c), which shows that secondary feedstocks can be converted to biofuels more efficiently than pure BL when taking into account only primary inputs to the biofuel production process.



**Figure 2. System (primary energy inputs) and expanded system (overall mill and refinery balance) efficiencies for all pathways.**

A somewhat different picture emerges when integration effects at the mill and refinery are also taken into consideration. Expanded system efficiencies are 99-256%, 51-68%, and 51-68% for Model Mill, Södra and SKKP, respectively. The efficiencies of the lignin (1a), lignin+electrolysis (1b) and BLG (2a) pathways exceed 100% when integrated with Model Mill. The main reason behind this non-intuitive result is that relatively inefficient electricity generation from BL is substituted with more efficient biofuel production. As described in Section 2.1.3, in the expanded system the feedstock is not BL but net change in electricity and biomass. These pathways therefore offer a more energy efficient means of using the energy surplus at the Model Mill.

The lignin pathway (1a) shows the highest efficiency. The only expanded energy input besides hydrogen is the reduction in the electricity surplus at the mill. In the lignin+electrolysis (1b) pathway, the electrolyser used for the production of hydrogen is included within the system boundary and the loss of energy during conversion is visible in the energy performance. Both lignin pathways, and to a lesser extent, the gasification pathways, benefit from the assumption that energy gases and the heat released during hydrotreatment of lignin and upgrading of methanol can substitute for fossil-derived energy at the refinery.

The BLG pathway (2a) also offers an efficient route for converting the pulp mill energy surplus into biofuels. It is a net exporter of steam to the mill and, unlike the lignin separation pathways; the only integrated input is reduction in mill electricity export. Energy losses in the process chain mean that the energy performance is similar to that of lignin+electrolysis (1b) and somewhat below that of lignin (1a). The use of a secondary feedstock together with BL (2b, 2c) does not lead to an increase in efficiency when integration effects are taken into account.

The expanded system performance of the mills that do not benefit from an energy surplus (Södra, SKKP), is significantly lower than that of the Model Mill. The higher steam demand of the lignin pathways relative to that of the gasification pathways means a reduced performance when integrated with mills that have a negative (SKKP) or near-zero (Södra) energy surplus.

### 2.2.2 Technology maturity

Technology readiness scores are collated in Figure 3, with scores and weights for the individual steps in each of the studied pathways given in the supplementary material of [25].

The TRL of the gasification-based pathways is notably higher than that of the lignin separation-based pathways, under both the weighted average approach and the weakest link approach. All process steps in the gasification-based pathway 2a have been demonstrated in pilot scale leading to a TRL of 7 [27,29,51]. The variants with PO co-gasification (2c) and hydrogen addition (2b) have a slightly lower TRL.

The most important individual steps in the lignin separation-based pathways have TRLs of 4-6. The weakest links from a TRL perspective are the formation of a pure, stable intermediate VGO/lignin mixture from the separated lignin, and the deoxygenation and cracking of this intermediate mixture. A TRL of 4 is assigned to both these steps, corresponding to lab scale validation, but it should be noted that work is currently in progress to validate both steps in pilot scale.

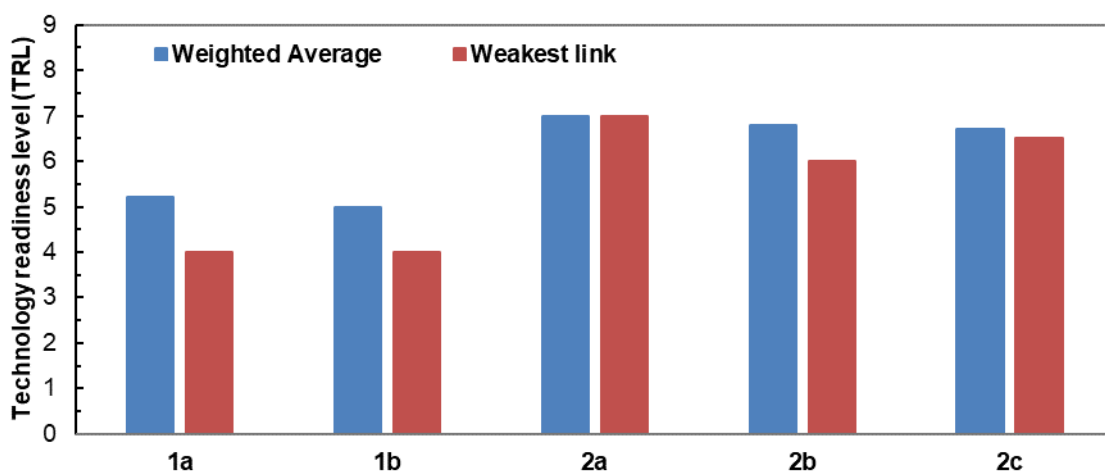
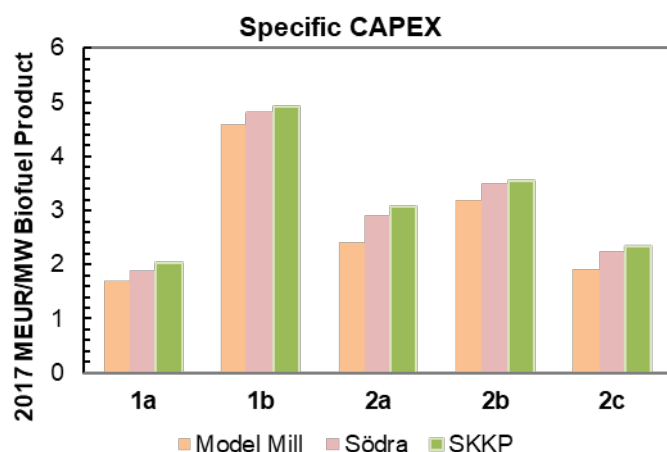


Figure 3. Technology readiness of examined pathways.

### 2.2.3 Economic performance

Capital costs per MW<sub>th</sub> of biofuels yield are shown in Figure 4. The small differences in specific CAPEX between different mills are attributable to economies-of-scale. The lignin pathway (1a) has the lowest specific CAPEX, followed by BLG+pyrolysis oil (2c) and BLG (2a). With the electrolyser unit as the dominant cost component, lignin+electrolysis (1b) has the highest specific CAPEX, more than twice that of 1a. The electrolyser also constitutes a significant cost factor in BLG+electrolysis (2b), although the difference relative to 2a is not as large as that between the two lignin pathways. It is conceivable that projected decreases in PEM electrolysis cost as the technology grows in maturity could lower the gap further.

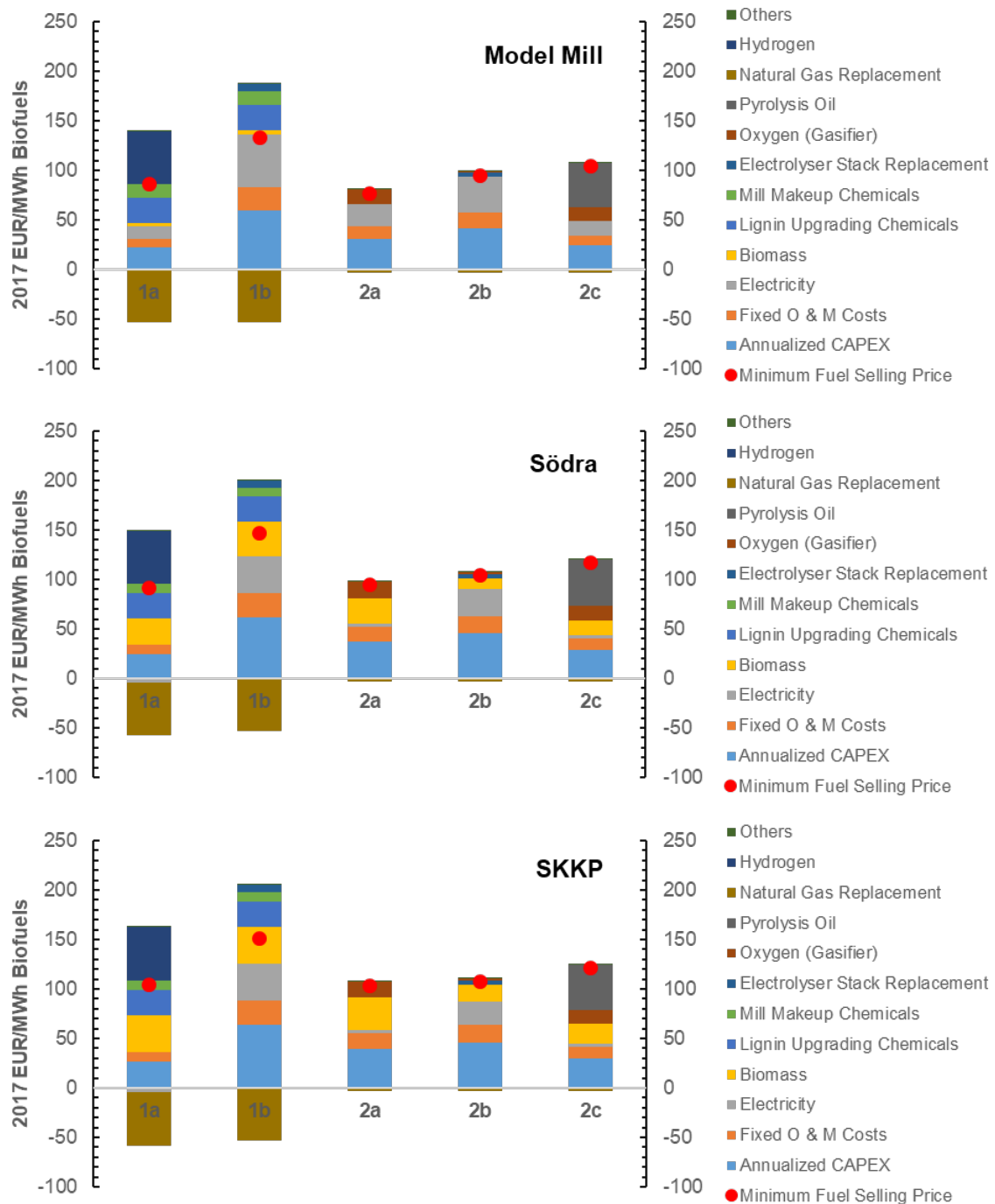


**Figure 4. Capital expenditure per MW of biofuel product (specific CAPEX).**

Biofuel production cost breakdowns and MFSPs for examined pathways are shown in Figure 5 for each of the three mill cases. Resulting MFSPs are 77-130 EUR/MWh for Model Mill, 91-150 EUR/MWh for Södra, and 100-150 EUR/MWh for SKKP, respectively. The MFSPs for the best cases are better than or comparable to the reported production costs for most drop-in alternatives from similar forest residue-based feedstocks, which range from 50 to 120 EUR/MWh for fast pyrolysis-based options, and 100 to 140 EUR/MWh for Fischer-Tropsch fuels [40]. The costs can also be compared to current drop-in biofuels on the market, for which production costs range from ca 65 EUR/MWh for palm oil/PFAD-based HVO, to over 100 EUR/MWh for tall oil-based HVO [52]. The import of biomass and electricity as a result of increased energy demand has a notably adverse impact on the economic performance of Södra and, particularly, of SKKP. The net cost of increased energy demand is significantly lower for Model Mill as a consequence of its energy surplus.

A comparison of examined pathways shows that 1a and 2a have the lowest total production costs in all mill cases. This indicates that both the gasification-catalytic synthesis route and the separation-hydrotreatment route can be used to produce biofuels from BL part-streams at comparable costs. 1b has the highest production costs, which is largely down to the high specific CAPEX of PEM electrolysis. The contrasting economic performance of the two lignin pathways (1a) and (1b) reflects the cost difference between fossil and renewable alternatives for lignin hydrotreatment.

The results appear to negate the hypothesis that the economic case for gasification-based biofuels from BL part-streams can be improved through the use of PO and hydrogen as secondary feedstocks. The specific CAPEX for 2c is lower than that for the other gasification-based alternatives but the costs associated with the purchase of PO, which is priced at 76 EUR/MWh, exceed the economic gain from increased biofuel yields. The economic viability of 2c is therefore dependent to a significant extent on the market price of PO. Similarly, the economic performance of 2b is markedly influenced by electrolyser investment cost with the cost difference compared to BLG (2a) being 4-18 EUR/MWh. A reduction in electrolysis cost by a third could put 2b on an equal footing with the best performing alternatives (1a, 2a).

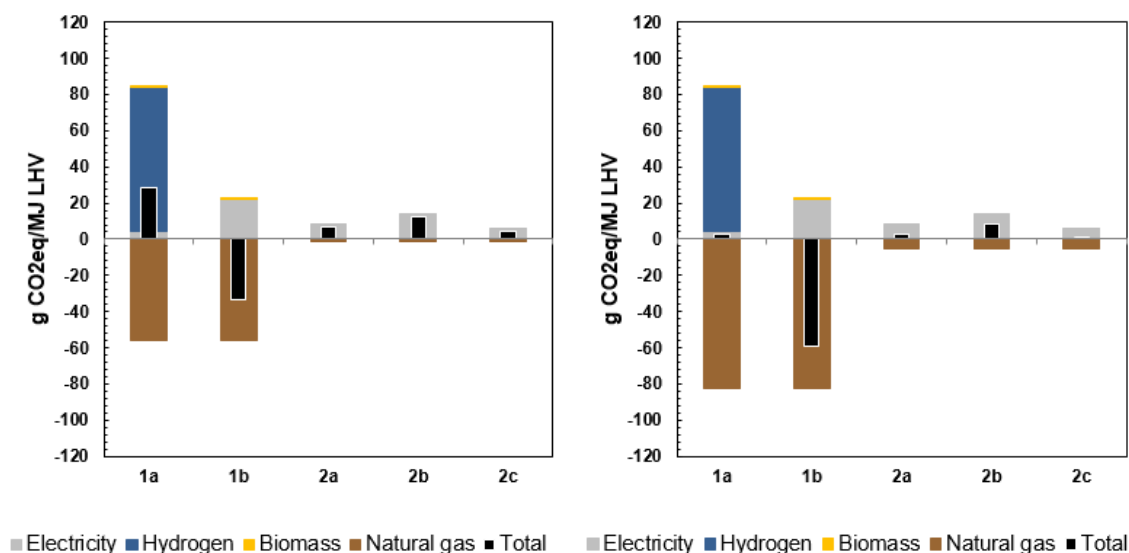


**Figure 5. Breakdown of biofuel production costs for examined pathways and mills. Lignin upgrading chemicals are carbon dioxide, sulphuric acid and stabilization additive. Mill makeup covers additional sodium hydroxide and ESP ash disposal costs. The savings from the replacement of natural gas with energy gases are included as a negative cost. Costs associated with methanol synthesis catalyst renewable, zinc oxide replacement and fossil production are aggregated under the label “Others”. A cost of 100 EUR/MWh corresponds to 9.5 SEK/l diesel and 8.6 SEK/l petrol.**

#### 2.2.4 GHG footprint

Results for the GHG footprint evaluation are given in Figure 6. Under the RED method, GHG emissions for the pathways range between 29 g CO<sub>2</sub>-eq/MJ for lignin (1a) and -33 g CO<sub>2</sub>-eq/MJ for

lignin+electrolysis (1b), with the gasification pathways falling in the middle. These numbers translate into GHG emission savings of 69-135% compared to fossil-based petrol and diesel references of 93.5 and 95.5 g CO<sub>2</sub>-eq/MJ, respectively.



**Figure 6. GHG footprint estimates for examined biofuel pathways derived using RED methodology (left) and an alternative approach based on system expansion (right). A Swedish electricity mix is assumed. Results shown are for Model Mill.**

It is evident from the results that the fossil hydrogen used for lignin hydrotreatment in 1a is by far the largest source of GHG emissions across all pathways. The recently adopted RED II requires new-built plants to deliver GHG savings of 65%. Without the significant mitigating effect of the energy gases replacing natural gas at the refinery, 1a falls short of the savings requirement. Energy gases are a product of lignin hydrotreatment, which has a highly uncertain yield structure based on current knowledge, as discussed previously. A shift in the product distribution towards petrol at the expense of energy gases could put 1a at the risk of failing to meet the RED II target, particularly if the hydrogen consumption remains unchanged. However, better experimental data is needed before a more definitive assessment can be attempted.

It can also be seen that the GHG footprint for 1a is significantly smaller when emissions are estimated using the principle of system expansion, which permits the crediting of emission savings associated with the replacement of natural gas with heat from lignin hydrotreatment. A comparison of 1a with 1b, which has net negative GHG emissions clearly demonstrates the extent of the reduction that can be achieved by switching to electrolysis hydrogen. Finally, all of the gasification-based pathways are able to satisfy the savings requirements in RED as the expanded system feedstocks, biomass and Swedish electricity, both have low GHG footprints.

## 2.3 SUMMARY OF TECHNO-ECONOMIC PERFORMANCE

The results of the economic evaluation show that black liquor part-streams can be used to produce drop-in biofuels with production costs ~80 EUR<sub>2017</sub>/MWh, thereby equalling or bettering the economic performance of comparable forest residue-based fuels [40]. The best performing pathways in the (lignin) separation-hydrotreatment and (black liquor) gasification-catalytic synthesis routes

were found to have broadly similar production costs. Both routes are therefore potentially attractive options for recovery boiler-limited pulp mills looking to increase pulp capacity and broaden product portfolios through comparatively modest investments. The gasification-catalytic synthesis route has a higher technology readiness level on average, but the gap is expected to shrink notably in the near future as key process steps in the separation-hydrotreatment route undergo planned demonstration in industrial conditions. However, our lignin upgrading results are subject to large uncertainties, being based on lab testing of a reference lignin in the absence of representative industrial data.

The use of natural gas as hydrogen source represents the cheaper option (1a) for lignin hydrotreatment by some margin, but is accompanied by GHG savings that are smaller relative to those for other pathways. This can be disadvantageous under the current Swedish quota obligation scheme for drop-in biofuels that is designed to reward biofuels with high GHG emission reductions. At the same time, the large capital cost of proton exchange membrane electrolyzers makes it unlikely that the renewable-hydrogen pathway (1b) will be economically competitive in the near future. The use of alkaline electrolyzers represents a potential option for cost reduction that has not been investigated in this study. Future availability of cheaper electrolyzers and reliable experimental data on lignin hydrotreatment are likely to impact the trade-off between fossil and renewable hydrogen in the medium term.

The hypothesis that secondary feedstocks such as hydrogen (2b) and pyrolysis oil (2c) can improve the economic performance of small-scale debottlenecking units based on black liquor gasification (2a) has not been proven. Results in this question are sensitive to future developments in the price of pyrolysis oil and the capital cost of electrolyzers.

Since the demand for forest residue-based drop-in alternatives that can replace both petrol and diesel is expected to grow globally, the complementary deployment of black liquor gasification-catalytic synthesis and lignin separation-hydrotreatment can be a strategically interesting option for achieving deep reductions in transport GHG emissions.



## 3 PULP MILL ASPECTS

This chapter relates to the following project objective:

2. Evaluate the technical and economic effects for pulp mills, including the effects of changed pulp production capacity and changed mass and energy balances, and develop a practical basis for the pulp industry to estimate the value of increasing the pulp mill recycling capacity through the implementation of pulp mill integrated biofuel production from black liquor (BL) part-streams.

### 3.1 TECHNICAL EFFECTS

The starting point of this project was that biofuel production from a part of the total BL volume can constitute an option for recovery boiler debottlenecking, for pulp mills aiming to increase their pulp production, but that currently are at the recovery capacity. As has been described, we applied a scenario in which the pulping capacity is increased by 18.5% at each mill, which was achieved by directing 18.5% of the BL dry solids flow to the biofuel plant, while keeping the thermal load on the recovery boiler the same as under normal operation. It is likely that removing a bottleneck in the recovery boiler will result in new bottlenecks elsewhere in the production. We have here assumed that the costs of resolving those bottlenecks are low enough that it will be relevant to implement the capacity increase, but we have considered explicitly only the part of the capacity increase that concerns how the recovery is affected.

The following sections focus on impacts of introducing BL-based biofuel production on the mill's energy system, with a short outline of other effects that also needs to be considered.

#### 3.1.1 Integration impacts on the mill's overall energy system

Figure 7 shows the results of integrating biofuel production on the mill's electricity and boiler bio-mass balances under the assumed 18.5% increase in pulp production capacity. A reference case with increased pulp capacity but no biofuels production is provided for comparison ("No bio-fuels"). The reference case presupposes that an increase in recovery boiler capacity of 18.5% can be realised by a capacity expansion retrofit<sup>1</sup>. It also assumes the bark available at the Model Mill is not fired in the power boiler but is made available on the biomass commodity market.

In the "No biofuels" case, the Model Mill operates with a significant electricity surplus, Södra exports only small amounts, while SKKP is a net importer of electricity. Steam from the recovery boiler can cover internal process demand at Model Mill and Södra, while additional biomass needs to be fired in the power boiler to meet the requirements of units such as the paper machines at SKKP. The integration of biofuels leads to an increase in energy demand at the Model Mill, which is met by reducing the electricity exports.

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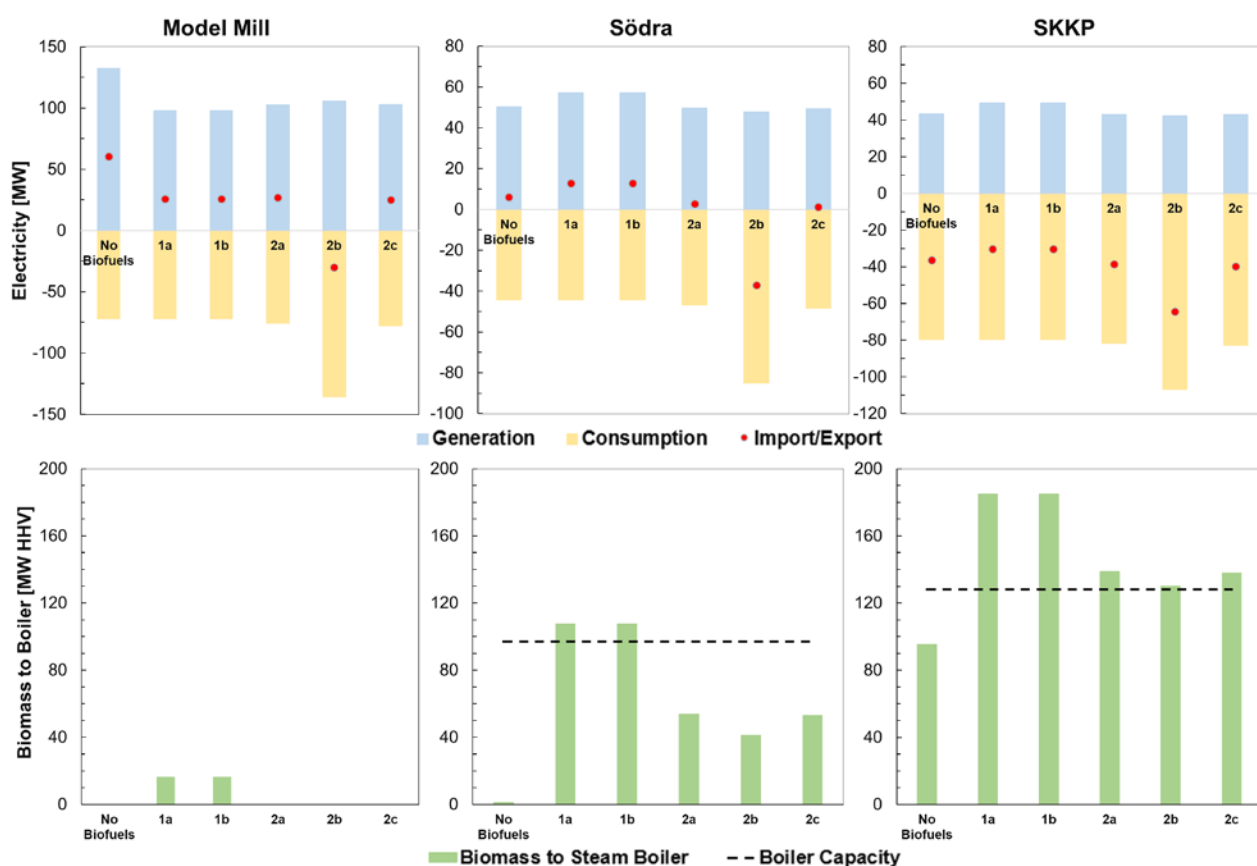
<sup>1</sup> This possibility (and its cost) is highly mill and boiler specific and will be dependent on the boiler design, age etc. as discussed in section 3.2.1 below.



In the lignin pathways, wash water from lignin purification is returned to the mill's evaporator plant, leading to an increase in steam consumption. This, in conjunction with the steam requirements of various lignin processing steps, means that additional biomass needs to be fired in the power boiler, even for Model Mill. For Södra and SKKP, the increase in steam demand in the lignin pathways is large enough that the bark-fired biomass boiler operates at or above the limit of its capacity. This implies that investments in bark/biomass boiler capacity expansion could be required if the mills were to consider using the lignin separation route for increasing pulp production. Since the lignin separation route is under active commercial development, the specific steam consumption may come down in the near future as optimisation increases with improvements in technology maturity.

The electricity balance is in particular affected by the electrolyser in the gasification+electrolysis pathway (2b), where all mills need to import electricity from the grid, even Model Mill which otherwise operates with a significant electricity surplus.

BLG green liquor has a higher concentration of carbonate ions compared with recovery boiler green liquor. The specific energy requirement of the causticizing process in which carbonate is calcined is accordingly also 40-42% greater, which translates into an additional lime kiln energy requirement of 7.5-7.8%, which was met by purge gas from the methanol synthesis.



**Figure 7.** Effect of biofuel production on the balance of electricity (top row) and biomass boiler (bottom row) at the studied pulp mills. See Table 1 for an outline of pathways. Note that the y-axis scales differ among top-row plots.

### 3.1.2 Other integration related aspects to consider

#### The separation-hydrotreatment route

As has been described, the sulphuric acid containing wash water stream from the lignin purification unit was considered to be sent to the mill and mixed with lean BL. The resulting increase in the amount of sulphur in the mill recovery cycle was mitigated by increasing the purge rate of ESP ash from the recovery boiler. Additional sodium hydroxide was added to the recovery cycle to make up for the resulting loss of sodium that left together with sulphur.

The permeate from the membrane separation, containing approximately 80% of the inorganic constituents in the lean BL as well as hemicelluloses, was considered to be returned to the mill evaporation unit. As discussed in the previous section, this leads to an increase in steam consumption in the evaporation train, as the permeate is more lean than the original weak liquor. We used an experience number for a modern seven-effect evaporation plant to estimate the steam consumption, which may be overoptimistic for less modern mills. Another aspect that needs to be considered is that the higher concentration of inorganics may increase the risk of scaling in the evaporation. It may then be necessary to run the evaporation to a lower dry solids content in the strong black liquor, which in turn affects the combustion properties in the recovery boiler, leading to lower efficiency.

Development work is currently ongoing to find a solution to instead send the wash water to the waste-water treatment plant, which would reduce the steam consumption for evaporation, and probably also the make-up chemical demand. However, the possibility to remove the relevant organic contaminants in a standard waste-water treatment plant and the impact on the mill chemical balance has not been quantified and this option is not considered in the present project.

It is also important to consider the balance between LP, MP and IP steam, as significantly more LP than MP or IP steam is used in this route. This has been considered in the modelling, and thus contributes to the final energy balances (discussed above, with details in the Supplementary material of [25]). Conversely, significant amounts of cooling water exit the process at around 35 °C, and has not been considered in the modelling. This may, for example, affect environmental permits.

#### The gasification-catalytic synthesis route

While the final green liquor which is returned to the pulp mill is similar to the green liquor from the recovery boiler, there are some differences that needs to be considered.

The gasification removes sulphur to the gas phase, from where it is separated as hydrogen sulphide in the amine wash. We have here considered that the hydrogen sulphide is returned to the recovery boiler to avoid affecting the overall mill chemical balance. The sulphur could also be purged, thus reducing the need for sodium make-up, and used to produce elemental sulphur or sulphuric acid. This is, however, in general not motivated economically.

Due to the resulting higher carbonate content in the green liquor, the lime kiln load increases, as discussed in the previous section. The additional energy demand was considered to be met by firing purge gas from the methanol synthesis in the lime kiln. The availability of methanol purge gas rela-

tive to the increase in lime kiln load is presented in the Supplementary material of [25]). The practicality and cost of implementing purge gas combustion at the studied mills, as well as lime kiln capacity limitations, has not been examined here.

Regarding the effects on the steam system, the process delivers significant amounts of excess LP and IP steam to the pulp mill, which leads to less steam passing through the backpressure turbine and consequently lower electricity production. There is also relatively large amounts of excess hot water at around 90 °C, which has not been accounted for here, but which could, e.g., be used for make-up water preheating or be exported to a district heating network, if present. The condensate return is affected to some extent, as direct steam is used in the water gas shift unit. We have, however, not performed any process water optimisation calculations within this work. As in the case of lignin separation, significant amounts of cooling water are also needed, which has not been accounted for here.

## 3.2 ECONOMIC EFFECTS

The lignin separation and black liquor gasification processes provides two services: 1) offloading of the normal recovery equipment, and 2) the first step in a biofuel production process. This means that a synergy is realised if both a pulp production increase and biofuel production is implemented in the mill. We here try to estimate the size of this synergy.

### 3.2.1 Methodological approach for estimating the recovery service credit

The pulp production gross margin is often estimated as 100-200 EUR/ADt averaged over a market cycle. Chemical and energy recovery is responsible for about one third of the total investment cost in a pulp mill [53,54]. Hence, one approach to estimating the value of recovery capacity is to use one third of this margin estimate. This would then correspond to approximately 20-40 EUR/tDS BL. However, the pulp gross margin is highly variable both over time and between mills, which makes this approach problematic. Moreover, the gross margin for individual mills is business sensitive information that is not publicly available.

An alternative approach to indirectly estimating the value of the recovery capacity is to estimate an alternative investment cost for the increased recovery capacity provided by the BL gasifier or lignin separation. This alternative investment cost would typically represent a recovery boiler rebuild. The alternative cost can then be subtracted from the total biofuel investment cost, since it would correspond to the alternative for obtaining the same recovery capacity. This is a highly relevant means of comparing for a mill that wants to make an investment in increased pulp production capacity, and wants to answer the question of whether or not to combine this with biofuel production. It also has the advantage that it makes estimation of investments costs required in other parts of the process, e.g. the pulp line, unnecessary.

The cost associated with a recovery boiler capacity expansion is highly boiler specific, depending on boiler size, type, age as well as any previous modifications, and on whether the boiler has been prepared for an expansion or not. The specific cost can be both higher and lower than the specific cost for the corresponding capacity in a new recovery boiler, but such a cost can nonetheless give a rough estimate that is relevant, even if it is highly approximate.

**Table 7. Cost estimates for new recovery boiler investments.**

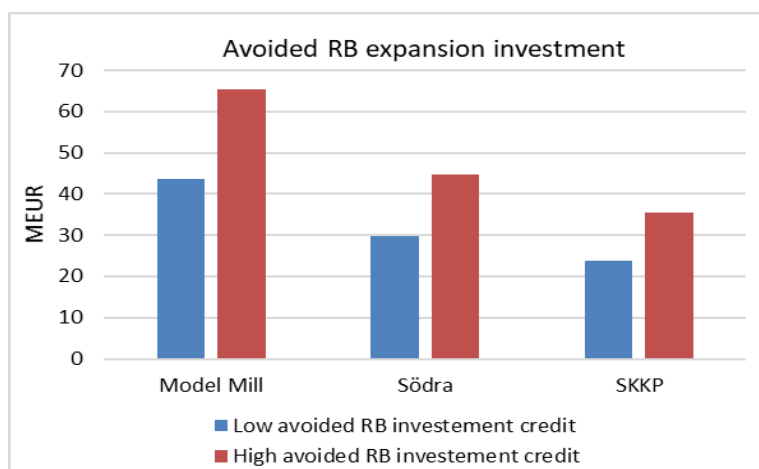
	Capacity (tDS/d)	Capacity (MW BL)	Investment (MSEK)	Specific investment (MSEK/tDS,d)	Specific investment (MSEK/MW BL)	Ref
Iggesund 2010	2500	405	2300	0.92	5.7	[55]
Husum 2020	4300	697	3000	0.70	4.3	[56]
Obbola 2007	1000	162	825	0.83	5.1	[57]
Reference mill	2800	463	1890	0.68	4.1	[54]

Table 7 gives publicly available data of recent recovery boiler investments or investment cost estimates. It shows that the specific investment cost is ca. 0.7-0.9 MSEK/tDS,d or 4-6 MSEK/MW BL. The investments in the table includes full project costs for recovery boiler and steam turbine. The latter is included since the OPEX used for the comparison includes changes in electricity production in the turbine. It can be noted that using 10 SEK/EUR, 350 d/y and an annuity factor of 0.15, the recovery capital cost is 29-39 EUR/tDS BL, which is in good agreement with the above-mentioned 20-40 EUR/tDS BL estimate based on pulp margin and investment cost fraction.

No assumptions about reduced O&M costs have been made. This means, in essence, that the O&M costs are not considered to be subject to synergies. The O&M cost for the combined pulp capacity expansion and biofuel production are simply the sum of the O&M of the two individual projects.

### 3.2.2 Impact on investment cost

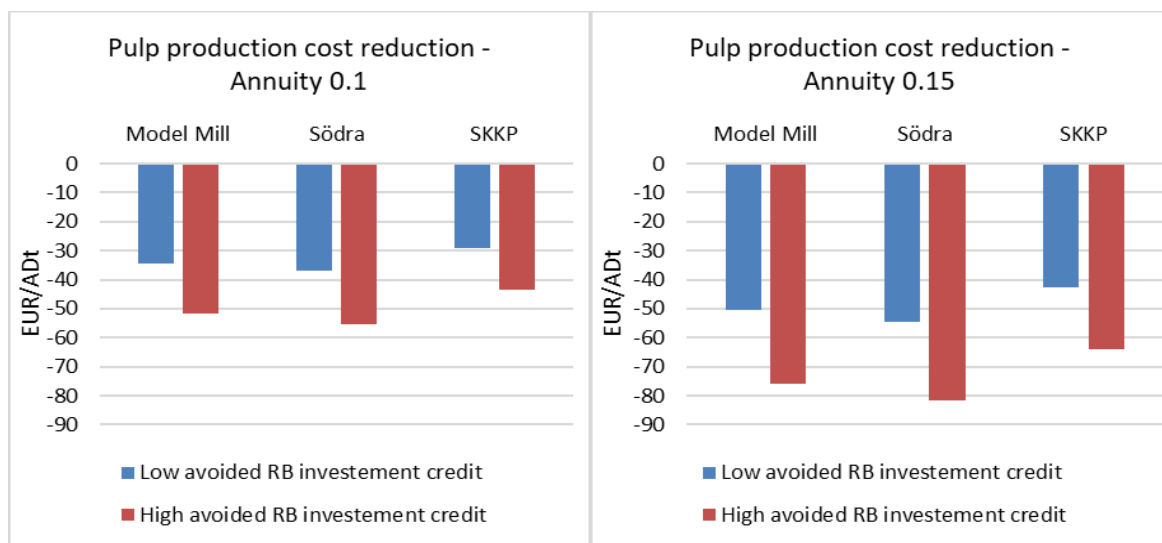
The avoided investment in the recovery boiler capacity expansion is applied as an estimate of the size of the above-mentioned synergy. Figure 8 shows this avoided investment for our three studied pulp mills, and for a low and high investment cost estimation, respectively. The differences between mills is directly dependent on the pulp production capacity of the mills, since a production increase of 18.5% is assumed in all cases and the same specific investment costs are used. The avoided investment can be arbitrarily allocated to either the pulp production increase or the biofuel production, since it is an effect of implementing both at the same time. In the following sections, we show both allocation possibilities. It is important to note that the synergy cannot be allocated fully to both options at the same time. Hence, the allocations here are mutually exclusive, but it is of course also possible to allocate the synergy partly to each option in any proportion.



**Figure 8. Avoided investment cost in recovery boiler expansion when biofuel production is implemented with pulp production capacity expansion.**

### 3.2.3 Allocation of synergy to increased pulp production

An allocation of the synergy to the increase pulp production gives results as shown in Figure 9 for the three examined mills, for two different assumptions on cost of capital: “Annuity 0.1” corresponds to 8% over 20 years while “Annuity 0.15” corresponds to 14% over 20 years. The low investment credit assumption and low cost of capital gives production cost reductions of 29-37 EUR/ADt, corresponding to an increased gross margin of approximately 15-30% for the increased production volume (based on the 100-200 EUR/ADt margin assumption discussed above). For the high investment credit assumption and high cost of capital, the corresponding cost reduction is 64-82 EUR/ADt or 35-70% increased gross margin. These numbers are independent of the biofuel production technology selected since the avoided recovery boiler expansion capacity is the same in all cases and this is allocated purely to increased pulp production.

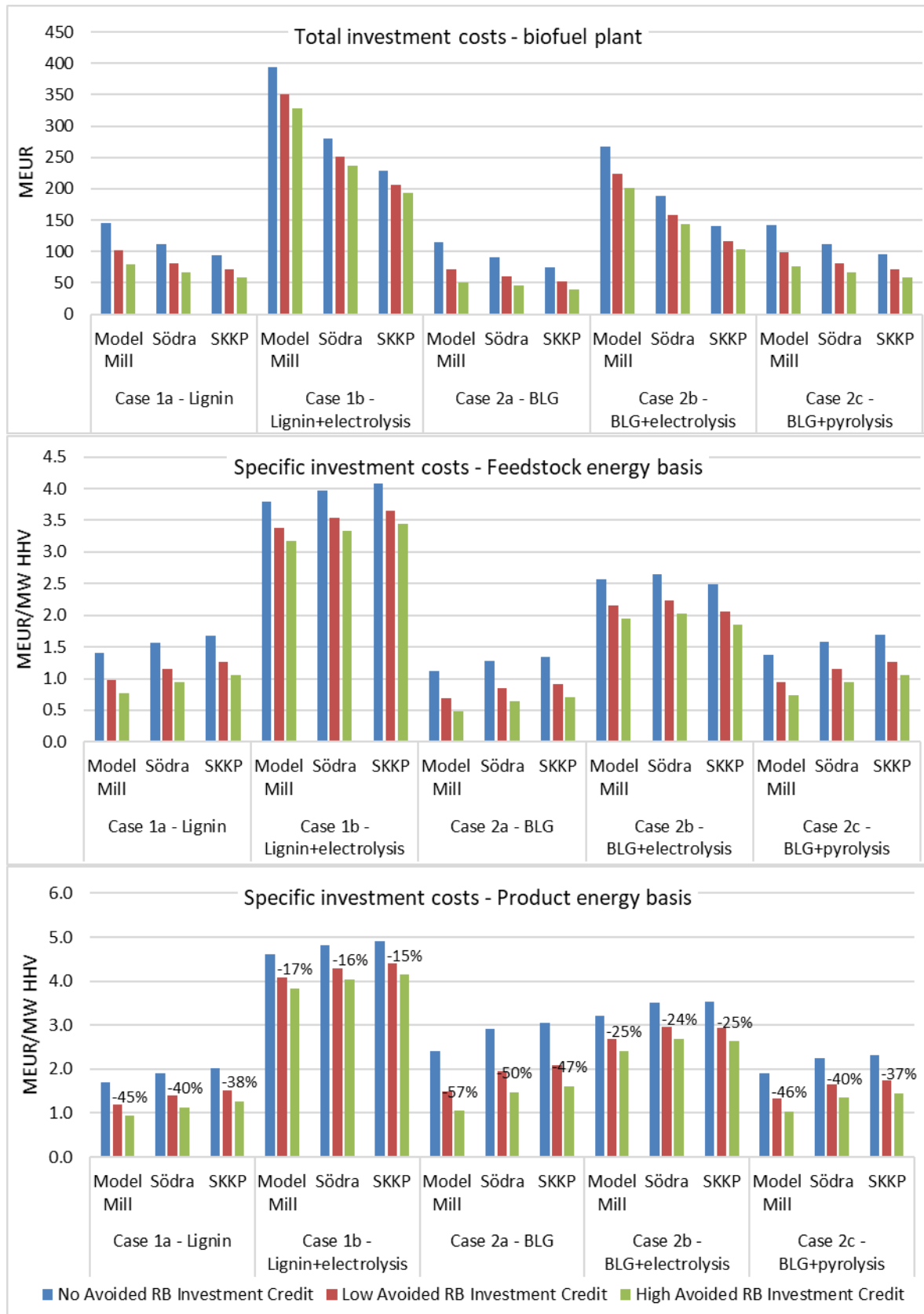


**Figure 9. Impact on pulp production cost for the increased pulp production capacity when the synergy (avoided investment cost in recovery boiler expansion) is allocated to increased pulp production.**

### 3.2.4 Allocation of synergy to biofuel production

If the synergy is instead allocated to the biofuel production, i.e., essentially assuming that the pulp production expansion would have been done anyway, the impact on biofuel production cost is no longer independent of the biofuel production technology used, since biofuel yields compared to BL/lignin feedstock vary. Figure 10 shows the resulting investment costs for the five studied biofuel production pathways, for our three studied pulp mills, with as well as without the credit for the avoided recovery boiler investment.

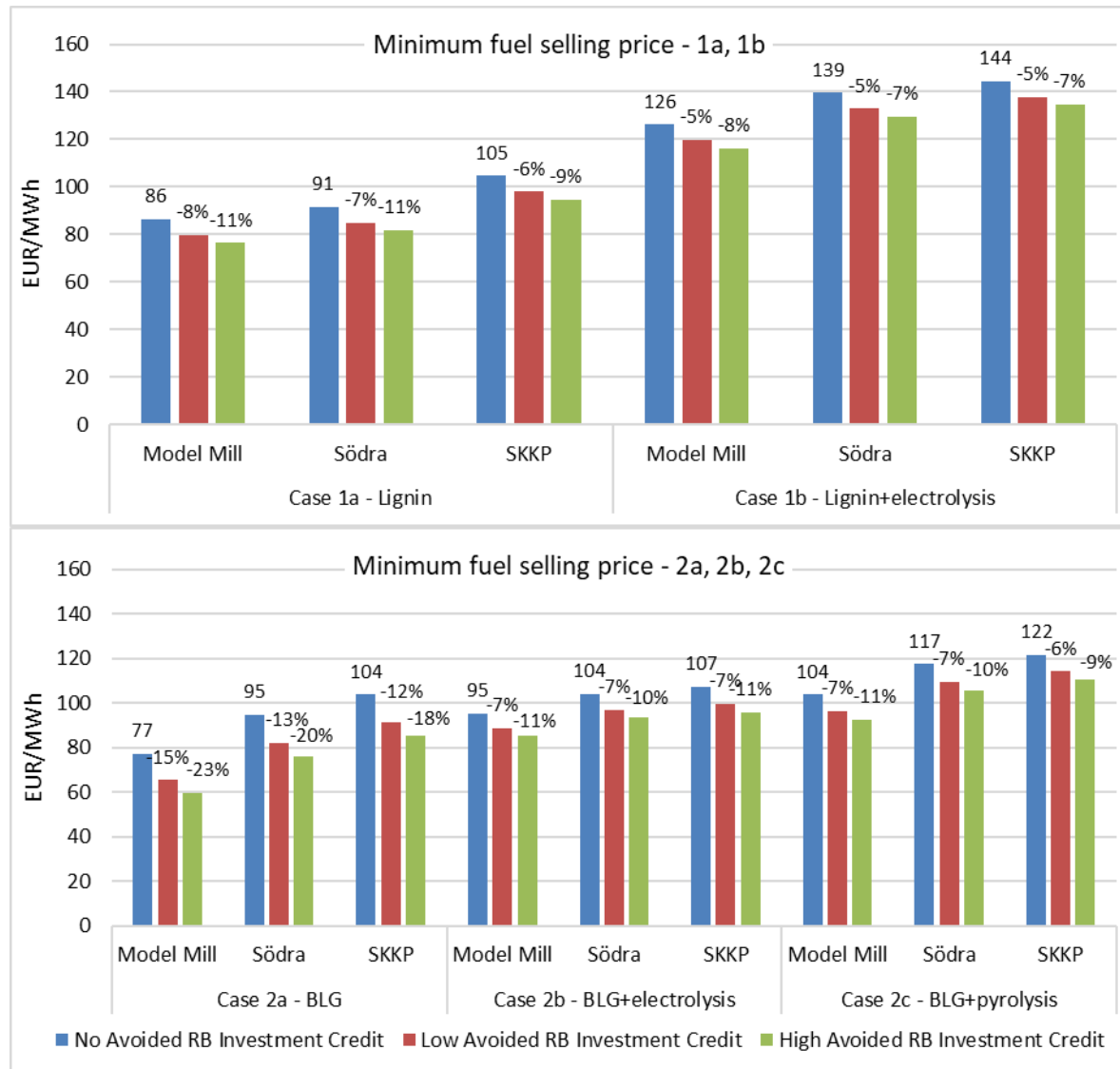
As noted above, the absolute reduction in investment cost is always the same for a given mill (Figure 8). The resulting relative change in investment cost from the recovery service credit, when allocated to the biofuel production, is thus largest for the cases with lower investment cost (1a, 2a, 2c). Case 2a shows a decrease of 32-38% and 47-57% in investment cost for the low and high recovery service cost credit, respectively. The smallest relative decrease is seen for case 1b (10-17%) due to its high total investment.



**Figure 10. Impact on biofuel production investment costs for the increased pulp production capacity when the synergy (avoided investment cost in recovery boiler expansion) is allocated to the biofuel production. Top: total CAPEX, middle: CAPEX per MW feedstock input, bottom: CAPEX per MW biofuel product.**

Figure 11 shows the resulting impact on the minimum fuel selling price (MFSP) for the five studied biofuel pathways, assuming a capital cost corresponding to the 0.1 annuity factor of the original MFSP calculations.

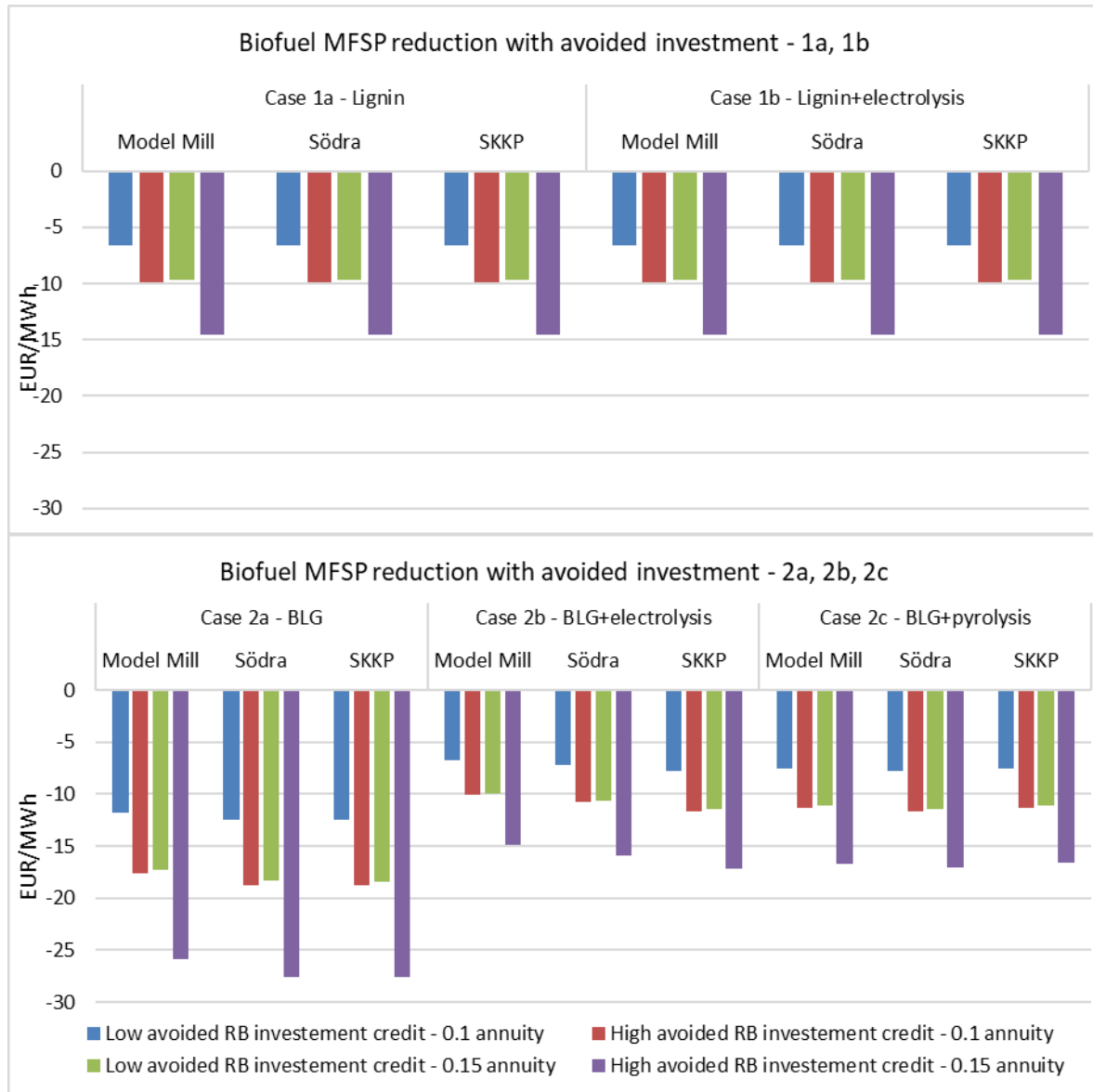
The largest effect on MFSP can be found for pathway 2a with reductions of 15% and 23% for the low and high recovery service cost credit, respectively. The MFSP obtained for pathway 2a at the Model Mill with the credit considered (60-65 EUR/MWh) must be considered very low for a forest based second generation biofuel. Similarly, pathway 2a also shows low MFSPs with the recovery service credit considered at the Södra and SKKP mills (76-91 EUR /MWh).



**Figure 11. Impact on the MFSP of the avoided investment cost for the increased pulp production capacity when the synergy (avoided investment cost in recovery boiler expansion) is allocated to the biofuel production, for the five studied biofuel production pathways.**

Figure 12 shows absolute reductions in biofuel MFSP for different avoided investment cost scenarios (low/high avoided investment and low/high cost of capital), similar to the results in Figure 9. For the lignin separation pathways 1a and 1b, the reduction in MFSP amounts to 7-14 EUR/MWh and similar for gasification with additional feedstock (pathways 2b and 2c, 7-17 EUR/MWh). For

the BLG pathway 2a, the effect is however larger with reductions of 12-19 EUR/MWh for the low annuity scenario, and 17-28 EUR/MWh for the high annuity scenario. The reason for the larger effect for 2a is the lower biofuel production capacity for this pathway, which is partly caused by the fact that it does not use any additional feedstock such as hydrogen (1a, 1b, 2b) or pyrolysis oil (2c).



**Figure 12. Impact on the MFSP of the avoided investment cost for the increased pulp production capacity when the synergy (avoided investment cost in recovery boiler expansion) is allocated to the biofuel production, under different scenario for investment credit and annuity.**

### 3.3 SUMMARY OF PULP MILL CONSIDERATIONS

A key assumption has been that a pulp mill that wants to achieve a pulp production capacity increase, but that has a bottleneck in the current recovery capacity, has two options for achieving the capacity increase; (1) a recovery boiler rebuild for increased recovery capacity, or (2) debottlenecking through using part of the total BL volume for biofuel production. While the first option has the



advantage of sticking to only the core business of the pulp mill, the second option generates a synergy between relieving the existing recovery equipment and creating a new revenue stream for the pulp mill from the produced biofuel.

From the pulp mill perspective, two aspects critical to consider in this choice are: *What would be the cost of the recovery boiler rebuild?* and *What is the current energy situation in the pulp mill?* With a low cost of the recovery boiler rebuild (exemplified here by the low investment credit assumption and low cost of capital), pulp production costs can be reduced by around 29-37 EUR/ADt, which corresponds to an increased gross margin of approximately 15-30% for the increased production volume. With a higher rebuild cost (exemplified by the high investment credit assumption and high cost of capital), the corresponding cost reduction can reach 64-82 EUR/ADt or 35-70% increased gross margin. These numbers are independent of the biofuel production technology selected since the avoided recovery boiler expansion capacity is the same for all biofuel production pathways.

Regarding the second aspect, the integration of biofuel production from BL part-streams results in significant impacts on the mill's energy balance, in particular in the steam system. Mills that are currently operating with an energy surplus that is exported as electricity therefore have a significant advantage as integration sites. Mills that operate their biomass or power boilers at or near capacity may have to invest in extra capacity to meet the increased demand from biofuel integration. For a pulp mill looking to expand pulping capacity through the production of drop-in biofuels from black liquor, integration impacts that require more detailed investigation include the impact on evaporator loads, relevant for lignin pathways, and lime kilns, pertinent for gasification pathways.

The introduction of an additional process step (lignin separation or gasification) in the recovery cycle naturally increases the risk of operational disruptions. However, the risk of impact on pulp production is considered to be limited because there is a buffer capacity in the form of tanks for both black liquor and green liquor. This allows the mill to continue the pulp production at full capacity for a limited time, as long as the majority of the recovery, which is done in the recovery boiler, is in operation. In the event of longer operational disruptions in the new processes, the pulp production will be affected, but it is important to point out that this only applies to a part of the production, corresponding to the part of the black liquor that goes to lignin separation or gasification.

## 4 DRIVERS FOR AND BARRIERS TO IMPLEMENTATION OF THE STUDIED TECHNOLOGY PATHWAYS

This chapter relates to the following project objective:

3. Provide an overview of general driving forces for and key barriers (technical, economic, organizational, etc.) to actual implementation of the studied technology pathways, which includes and builds on relevant industrial players' perceptions of different drivers and barriers.

### 4.1 METHODOLOGICAL APPROACH

Previous research [58] highlights several system strengths, such as long-term research funding, significant research infrastructure, and strong actor networks, that have contributed to developing the Swedish biorefinery technological innovation system. However, significant system weaknesses which inhibit the development have also been pointed out, such as weak coordination among ministries, lack of industrial absorptive capacity, and unclear roles. To address the system weaknesses by building on the system strengths, previous research suggests a combination of four policy measures: (a) the implementation of a deployment policy for creating domestic niche markets, (b) improved policy timing and more structured coordination among different governmental agencies, (c) the provision of stronger incentives for mature industries to invest in R&D and improve their absorptive capacity; and (d) improved organisation and financing of existing research infrastructure [58].

While a full Technological Innovation System (TIS) analysis has been outside the scope of this project, we have employed a limited study focused on barriers to and drivers for actual implementation.

In the format of a workshop, the project partners and additional associated actors identified, discussed and aligned their view on key drivers and barriers (technical, economic, organisational, etc.) against actual implementation of the technology pathways studied in the project. The purpose of the identification of key drivers and barriers was to supplement the techno-economic analysis performed in the project with general aspects (mainly non-technical) which can be decisive for the feasibility of fuel production based on black liquor part-streams.

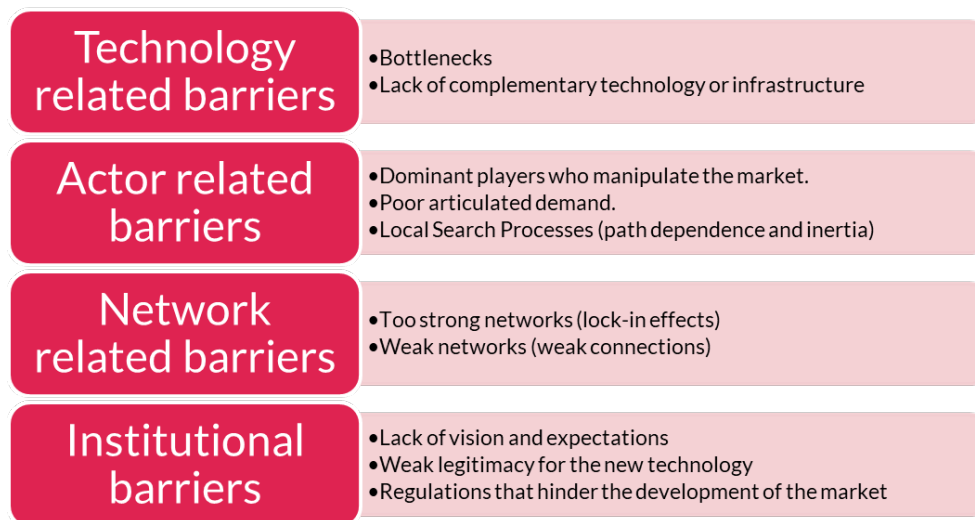
The implementation of the workshop was inspired by TIS analysis. TIS is a framework to analyse (e.g.) barriers and drivers for technological development and diffusion. Reasons for applying a TIS perspective are basically that the framework allows for an analysis of the links between technology, actors / organizations, networks and institutions (laws and regulations), and that it facilitates an enhanced understanding of the emergence, development, obstacles and dissemination of new knowledge (and technology). During the start of the workshop, the participants were given a brief introduction to TIS and some core concepts.

In a TIS analysis, care is taken to investigate the structure (Structural components) and function of the system with the purpose of identifying system weaknesses. Identified system weaknesses can either be influenced by the actors in the system themselves, or might require specific policy attention. Such an analysis is rather time consuming and demands significant resources to be performed.

Thus, for the aim and purpose in this project we performed a very light version using the structural components as a basis for discussion. The structural components are:

- Technology (codified knowledge, artefacts, ...)
- Actors (individuals, businesses, universities, other organisations)
- Networks (learning networks, political networks, social networks, ...)
- Institutions (laws and regulations, norms and values, standards, routines, ...)

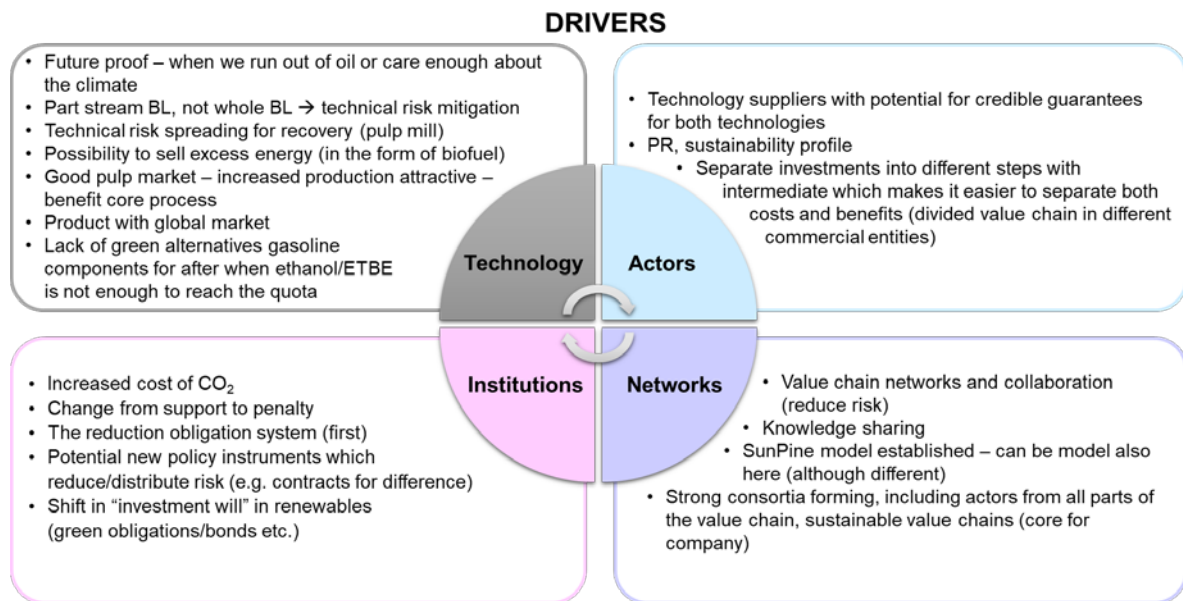
The structural components can in different ways inhibit innovation. Figure 13 shows some examples of barriers related to the different structural components.



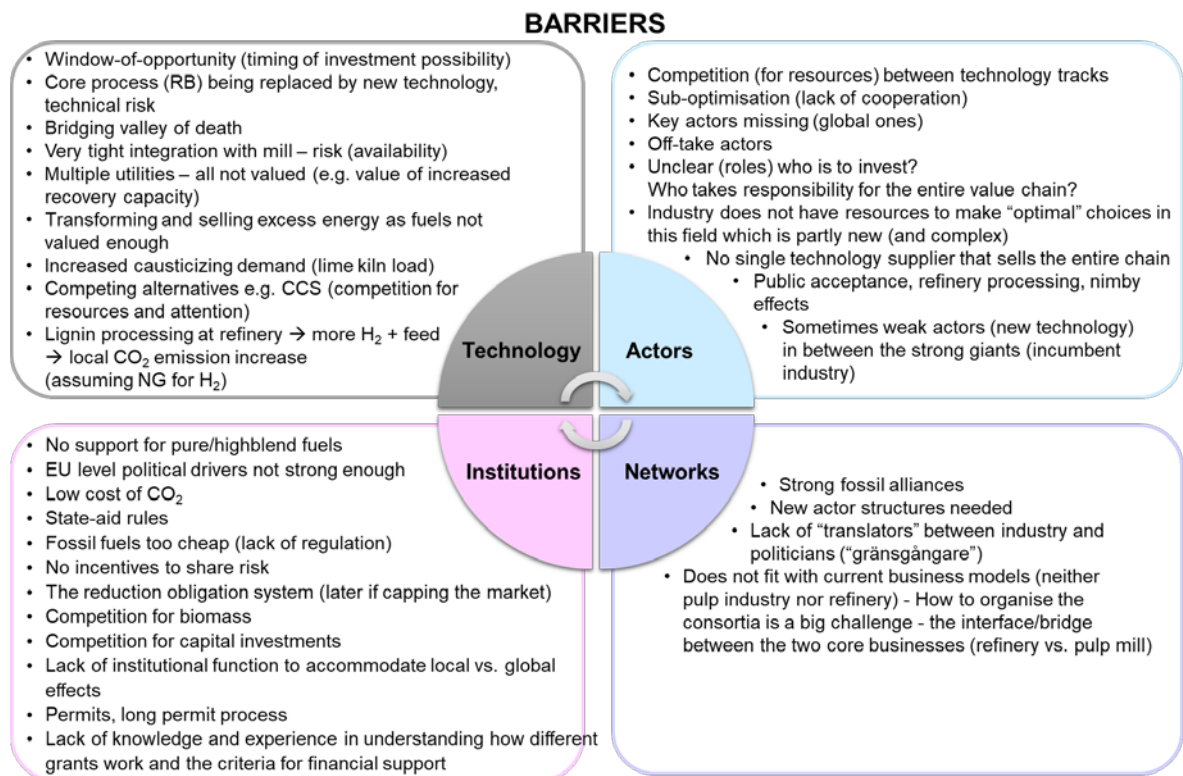
**Figure 13. Examples of barriers related to different structural components.**

## 4.2 WORKSHOP FINDINGS

The workshop findings are summarised in Figure 14 and Figure 15. The structure follows the one in the discussions related to the structural components: first drivers (strengths) are presented and thereafter a summary is given for identified barriers (weaknesses).



**Figure 14. Drivers for implementation of biofuel production from BL part-streams.**



**Figure 15. Barriers to implementation of biofuel production from BL part-streams.**

### 4.3 SUMMARY OF DRIVERS AND BARRIERS

All in all, it can be concluded that the workshop findings regarding barriers and drivers correlate rather well with the system strengths and weaknesses identified for the more general Swedish bio-

refinery TIS by [58]. This is especially true for the barriers and drivers associated with the structural components' institutions and networks. However, some of the drivers are more case specific focusing on the benefits possible due to integration with the pulp mill and/or the refinery. Especially the possibility to debottleneck the recovery boiler (and enabling a “cheaper” production increase) was highlighted. Yet, the integration with the pulp mill and/or refinery is also associated with barriers, where, e.g., the technical risk of close integration of new technologies and the need to time the potential investment into an “investment window of opportunity” when the mill or refinery is shut down for maintenance, were highlighted. Finally, in accordance with the more in-depth study by [58], the lack of key actors (and associated resources) and the challenge of unclear roles and weak network structures were emphasised.

## 5 CONCLUSIONS

This work has shown that BL part-streams can be used to produce drop-in biofuels with production costs ~80 EUR<sub>2017</sub>/MWh (ca 7-8 SEK/l), thereby equalling or bettering the economic performance of comparable forest residue-based fuels. Both studied routes could constitute potentially attractive options for recovery boiler-limited pulp mills looking to both increase their pulping capacity and broaden their product portfolios. Hydrogen for lignin hydrotreatment is significantly cheaper if produced from natural gas than via electrolysis, but offers lower GHG savings compared to the other pathways. Future availability of cheaper electrolyzers and reliable experimental data on lignin hydrotreatment are likely to impact the trade-off between fossil and renewable hydrogen.

Substantial synergies can be obtained by implementing the investigated technology concepts project. The “dual service” that both concepts offer gives a reduction in the overall investment cost if the pulp capacity increase is implemented together with the biofuel production. If this synergy is allocated to the biofuel production, the production costs can be reduced by significant amounts (up to 23%). If it instead allocated to the pulp production, pulp production costs can be reduced by up to 64-82 EUR/ADt pulp, which corresponds to an increased gross margin of 35-70% for the increased production volume, irrespective of biofuel production pathway. This, however, requires that the alternative cost of rebuilding the existing recovery boiler for a capacity increase would be in the higher end of the investigated alternatives.

From the pulp mill’s perspective, two main aspects are critical to consider in the choice between recovery boiler retrofit and black liquor-based biofuel production:

- 1) What would be the cost of the recovery boiler rebuild?
- 2) What is the current energy situation in the pulp mill?

The integration of biofuel production from BL part-streams results in significant impacts on the mill’s energy balance. Mills that are currently operating with an energy surplus have a significant advantage as integration sites, while mills that operate their power boilers at or near capacity may have to invest in extra capacity to meet the increased demand from biofuel integration. The lignin separation-hydrotreatment route is less suitable for pulp mills already restricted in the evaporation section, due to the increased evaporation load, while the gasification-catalytic synthesis route is less suitable for pulp mills already restricted in the lime kiln, due to the increased causticizing load.

While the gasification-catalytic synthesis route has a higher current technology readiness level on average, the gap is expected to shrink notably in the near future as key process steps in the separation-hydrotreatment route undergo planned demonstration in industrial conditions. However, our lignin upgrading results are subject to large uncertainties, being based on lab testing of a reference lignin in the absence of representative industrial data.

Finally, it can be concluded that since the demand for forest residue-based drop-in alternatives that can replace both petrol and diesel is expected to grow in the future, the complementary deployment of black liquor gasification-catalytic synthesis and lignin separation-hydrotreatment can be a strategically interesting option for achieving deep reductions in transport GHG emissions. In order to make this happen though, challenges in the form of lacking key actors (and associated resources), unclear roles and weak network structures must also be overcome.

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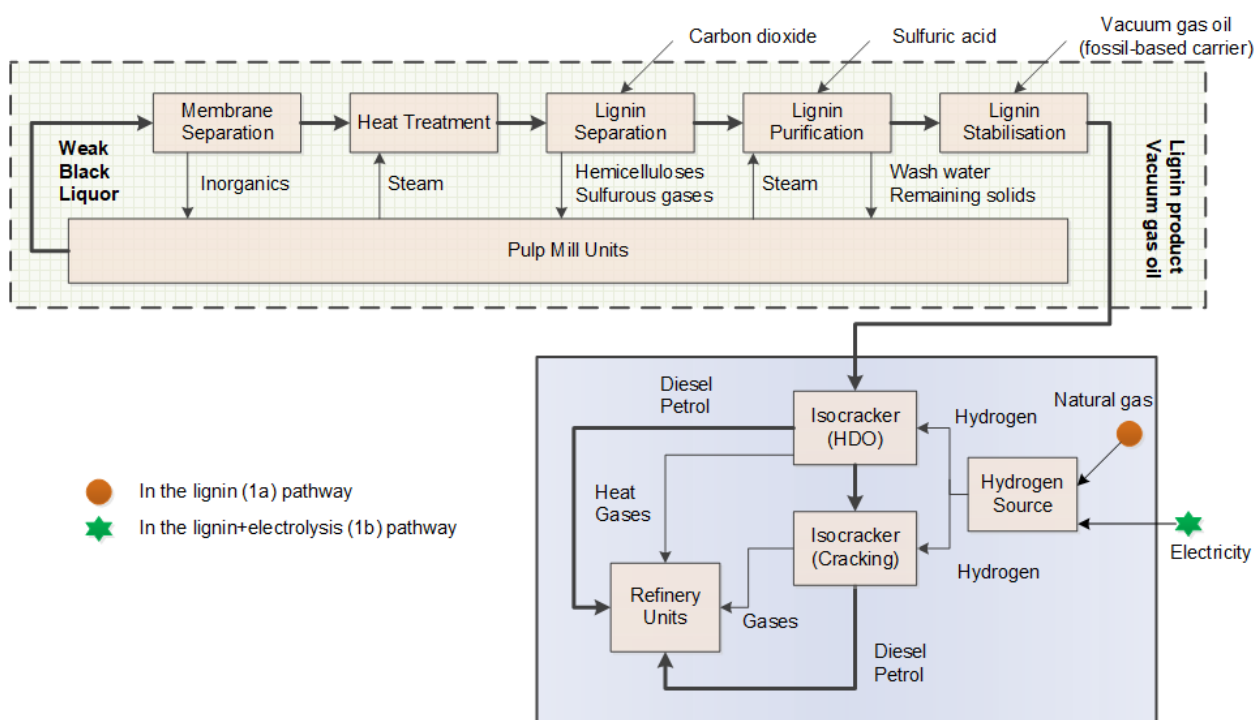
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## APPENDIX A – BIOFUEL PRODUCTION PATHWAYS

### THE SEPARATION-HYDROTREATMENT ROUTE

Lignin is separated from kraft BL, purified, stabilised and sent to a petroleum refinery where it is hydrotreated to remove oxygen and upgraded to diesel and petrol blends, as illustrated schematically in Figure A- 1. The process configuration for the pulp mill-integrated steps is based on technology developed by Sun Carbon AB [9]. The maximum amount of lignin that can be separated from BL without adventuring recovery boiler operation is mill specific, but is typically limited to ~20%. The two pathways in this route are identical in all aspects other than the choice of hydrogen supply source: steam reforming of natural gas in the lignin pathway (1a) and polymer electrolyte membrane (PEM) electrolysis of water in the lignin+electrolysis pathway (1b).



**Figure A- 1. Simplified process schematic for the lignin separation-hydrotreatment route. All process steps except the production of hydrogen are shared between pathways 1a and 1b.**

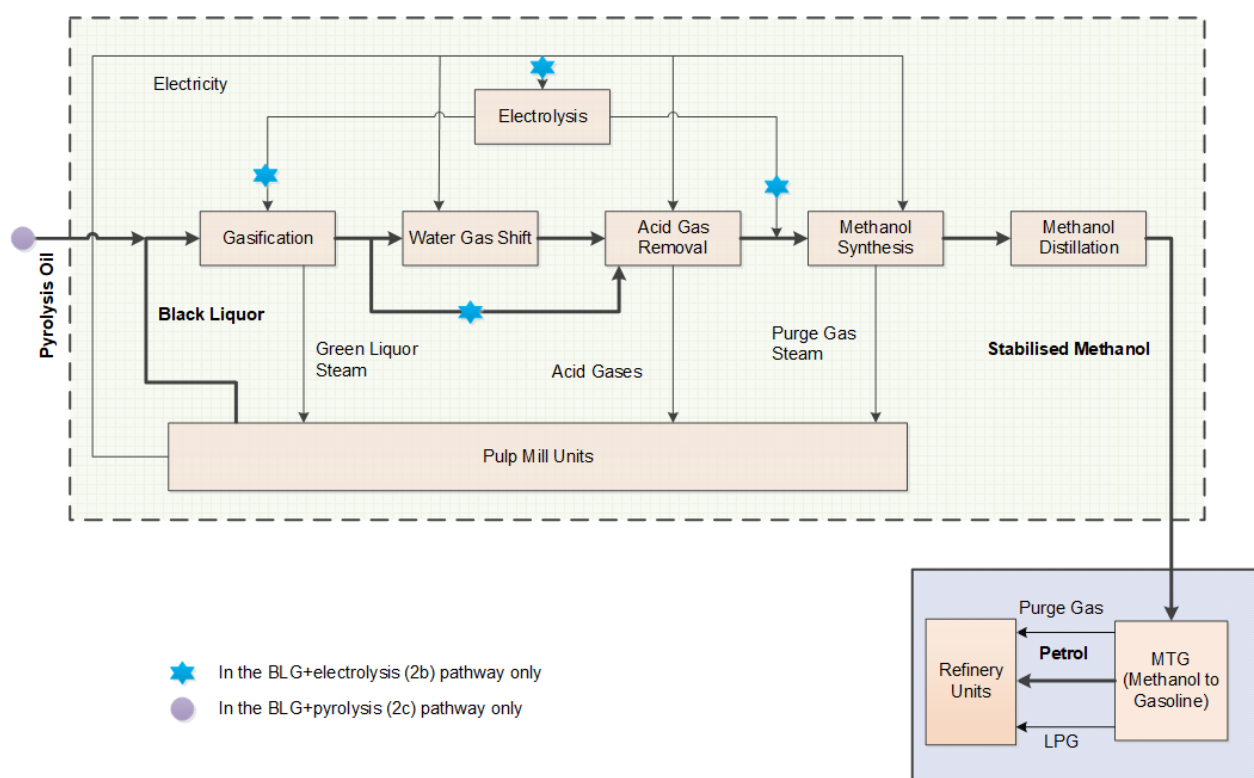
A stream of weak BL from the mill evaporation unit is split by membrane separation into a lignin and hemicellulose-rich retentate stream and an alkali-rich permeate stream. The alkali compounds in the permeate are returned to the evaporator train, while the retentate is subjected to indirect steam heating, which initiates an alkali-catalysed breakdown of the organic fraction. The hemicelluloses are also returned to the mill recovery cycle, while the depolymerised lignin fragments are acidulated with carbon dioxide, which reduces the pH and leads to the formation of a liquid lignin phase by protonation. The multiphase mixture is washed with sulphuric acid to leach out remaining impurities. An important part is the provision of lignin in a form that can be conveyed to the refinery and co-processed with crude oil derivatives. An organic solvent is used to generate a stable 50/50 mixture of lignin and a fossil carrier oil, vacuum gas oil (VGO), that can be transported and processed at the petroleum refinery.

The lignin/VGO mixture is hydrotreated in a two-step iso-cracker. In the first step, oxygen and sulphur are removed by treating the feed with hydrogen at elevated temperature and pressure in the presence of a catalyst. The bulk of the oxygen is removed by hydrodeoxygenation reactions. Partial saturation of aromatics also takes place. In the second step, the hydrocarbon products undergo cracking, isomerization and further saturation. The heavy ends are cracked into smaller components in the diesel and petrol range, which can be blended with similar fossil products.

## THE GASIFICATION-CATALYTIC SYNTHESIS ROUTE

Pure BL or blends of BL and PO are gasified to produce syngas that, optionally combined with hydrogen from water electrolysis, is used as feedstock for the production of drop-in petrol via a methanol intermediate, as shown in Figure A- 2. The gasification step common to all pathways is based on the BL gasification (BLG) technology developed by Chemrec AB [29]. For 2b and 2c, a second feedstock is used to increase biofuel production from a given volume of BL. For 2c, up to 25% PO can be mixed into the BL without any other changes to the gasification technology [59].

Concentrated BL is gasified in a pressurized refractory-lined reactor using oxygen as oxidizing and atomizing agent to produce a clean syngas made up primarily of carbon monoxide, hydrogen and carbon dioxide. Small quantities of hydrogen sulphide and methane are also present. The catalytic effect of the alkali in BL ensures nearly complete carbon conversion at relatively low reactor temperatures [60,61]. The alkaline pulping chemicals in the BL form a molten slag, which is dissolved in water and returned to the mill for re-use in the kraft cycle.



**Figure A- 2. Simplified process schematic for the black liquor gasification-catalytic synthesis route. There is no water gas shift in pathway 2b, syngas is sent for acid gas removal directly.**

After leaving the reactor, syngas is cooled down to 30-40 °C in two steps. The next treatment step differs between 2a and 2c on the one hand and 2b on the other. In 2a and 2c, the composition of the

cooled syngas is adjusted in a water-gas shift reactor after which carbon dioxide and hydrogen sulphide are removed by scrubbing with an amine solution. In 2b, there is no water gas shift unit. The stoichiometric requirement for methanol synthesis is satisfied by mixing syngas from the amine scrubber with hydrogen, which is produced by PEM electrolysis of water. Since amine scrubbers cannot reliably reduce the concentration of hydrogen sulphide to sub-ppm<sub>v</sub> levels, syngas is passed through a zinc oxide bed to reduce the concentration of sulfur to below 0.1 ppm. The conditioned syngas is catalytically upgraded to crude methanol, which is partially distilled to produce water-containing “stabilized” methanol. Petrol blendstock is synthesized from stabilized methanol via the methanol-to-gasoline (MTG) process at a petroleum refinery, to take advantage of existing infrastructure [51,62]. Small quantities of LPG are also obtained as a by-product. Other gaseous by-products from the synthesis loop are combusted for energy recovery.

## APPENDIX B – PULP MILL INPUT DATA

Table B- 1 summarises key pulp mill data under operation without integrated biofuel production.

**Table B- 1. Selected production and energy data for examined kraft mills under normal operation.**

	Units	Model Mill <sup>a</sup>	SKKP <sup>b</sup>	Södra <sup>c</sup>
<b>Production Data</b>				
Mill Type	-	Market Pulp	Pulp & Paper	Market Pulp
Pulp Production Capacity	tons/day	2000	1268	1288 <sup>d</sup>
<b>Electricity Data</b>				
Generation (Back Pressure Turbine)	MW <sub>el</sub>	70	35	40
Generation (Condensing Turbine)	MW <sub>el</sub>	73	-	-
Internal Consumption	MW <sub>el</sub>	61	67	39
Import from Grid	MW <sub>el</sub>	0	32	0
Export to Grid	MW <sub>el</sub>	83	0	1
<b>Steam Data</b>				
High Pressure Steam from Recovery Boiler	MW <sub>th</sub>	438	192	260
High Pressure Steam from Biomass Boiler	MW <sub>th</sub>	66	65 <sup>e</sup>	1
<b>Other Data</b>				
BL to Recovery Boiler	MW <sub>th</sub>	561	240	325
Feedstock to Lime Kiln	MW <sub>th</sub>	34	21	22
Feedstock to Auxiliary (Biomass/Power) Boiler	MW <sub>th</sub>	107	74	26-96
Process Steam Levels	bar(a)	13/10/4.5	27/11/3	25/14/11

<sup>a</sup> Recovery boiler produces high pressure (HP) steam at 101 bar(a). Falling bark is combusted in the lime kiln and the surplus can be fired in a power boiler to produce additional HP steam. The mill is equipped with a condensing turbine. Surplus electricity is exported to the grid.

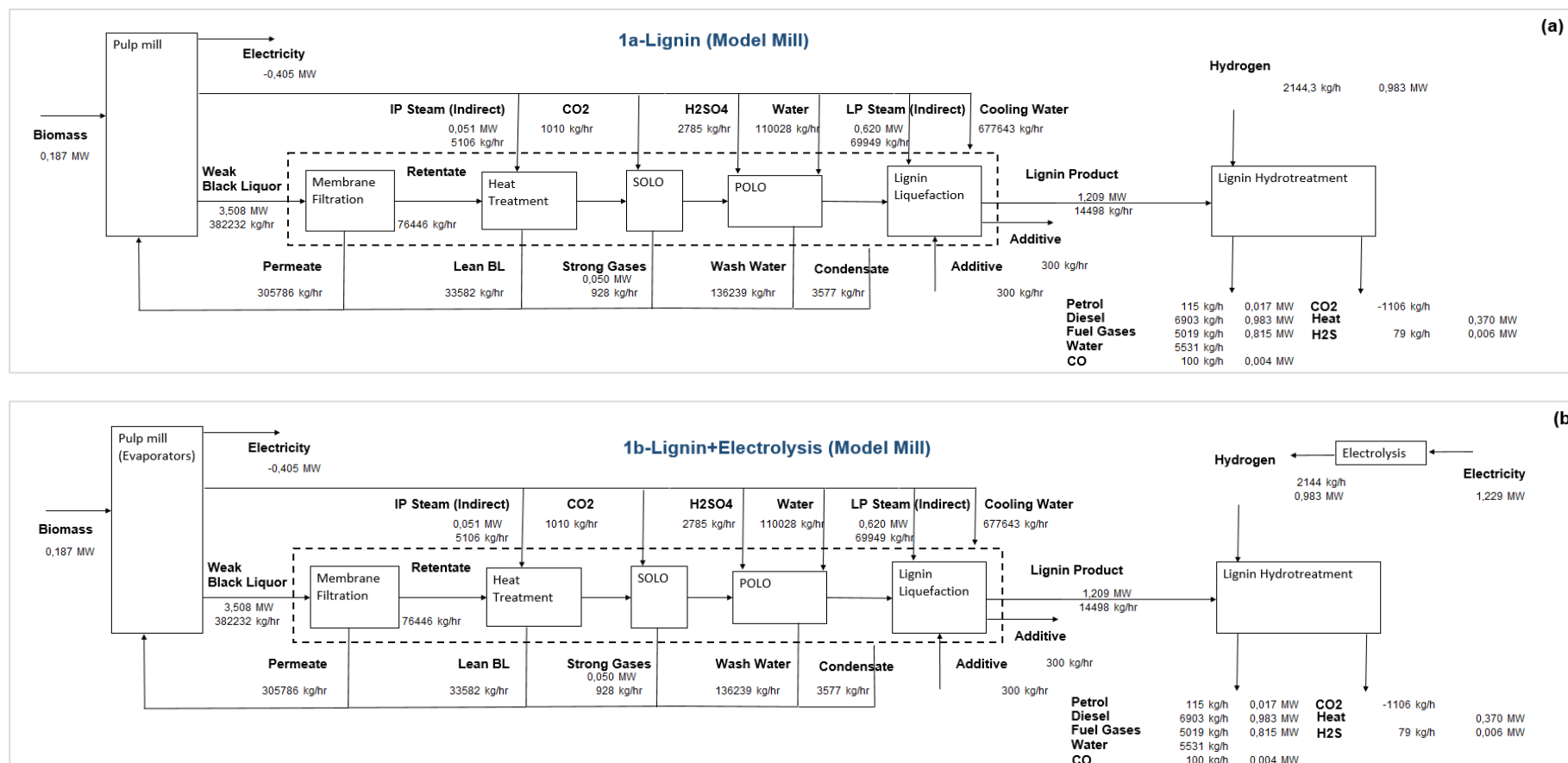
<sup>b</sup> Based on energy data for 2017. Recovery boiler produces HP steam at 58 bar(a). It is complemented by a multi-fuel biomass boiler that is fired mainly with bark from the debarking line, which produces HP steam at 120 bar(a). Two additional supplementary boilers are also present. Electricity is generated by two back pressure turbines. The lime kiln is fired with a combination of sawdust, pellets and bio-oil. The mill is a net importer of both electricity and biomass.

<sup>c</sup> Based on energy data for September 2018. Recovery boiler produces HP steam at 58 bar(a). The lime kiln is fired with tall oil residue. The mill is equipped with an auxiliary biomass boiler, but the steam output from the recovery boiler is normally sufficient to satisfy the process demand for steam and electricity. The mill has a lack of capacity in the existing turbines and no condensing turbine, and can thus not convert surplus steam to electricity.

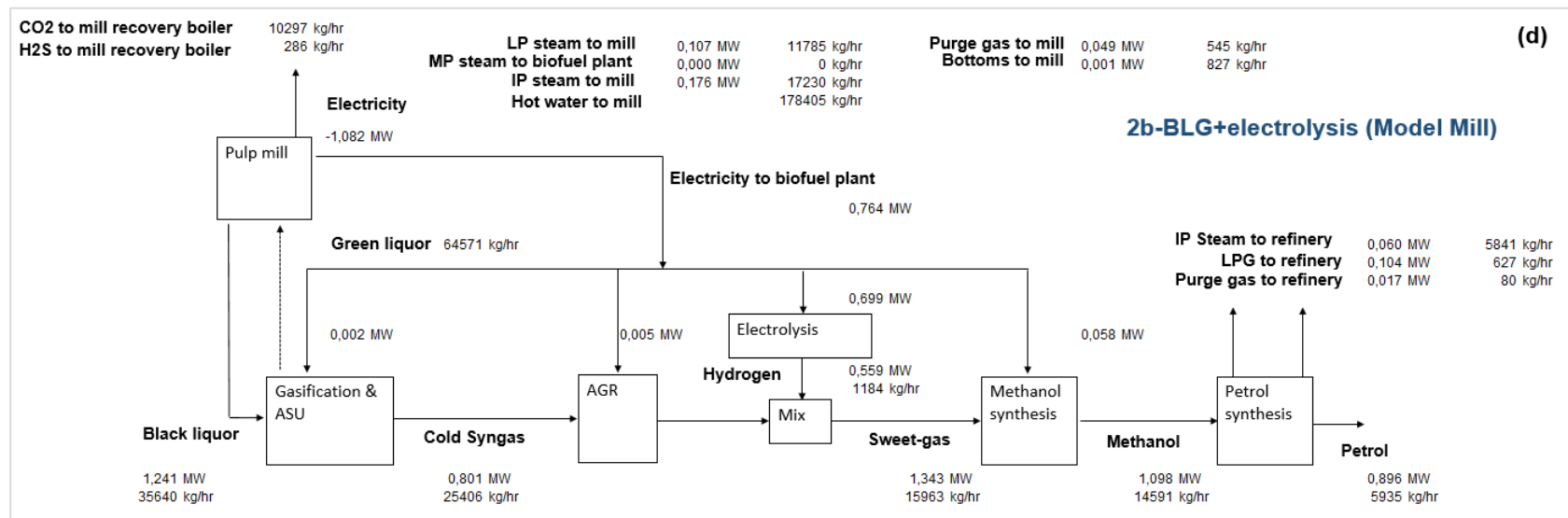
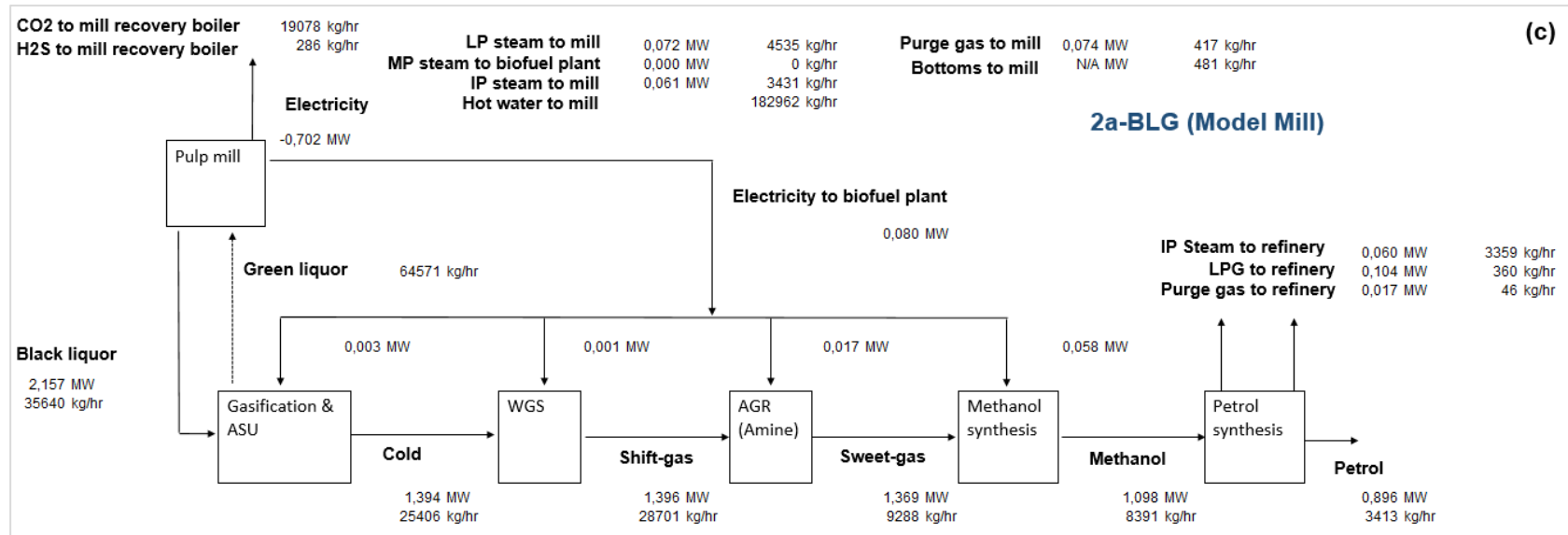
<sup>d</sup> Partly paper pulp, normally ~ 2/3 of the total, and partly textile pulp, normally ~ 1/3 of the total.

## APPENDIX C – DETAILED BALANCES FOR INTEGRATED BIOFUEL PRODUCTION

Figure C- 1, Figure C- 2 and Figure C- 3 show the detailed balances for Model Mill, Södra, and SKKP, respectively, for all five examined pathways. Note that figures break over pages.



DROP-IN FUELS FROM BLACK LIQUOR PART STREAMS –  
BRIDGING THE GAP BETWEEN SHORT- AND LONG-TERM TECHNOLOGY TRACKS





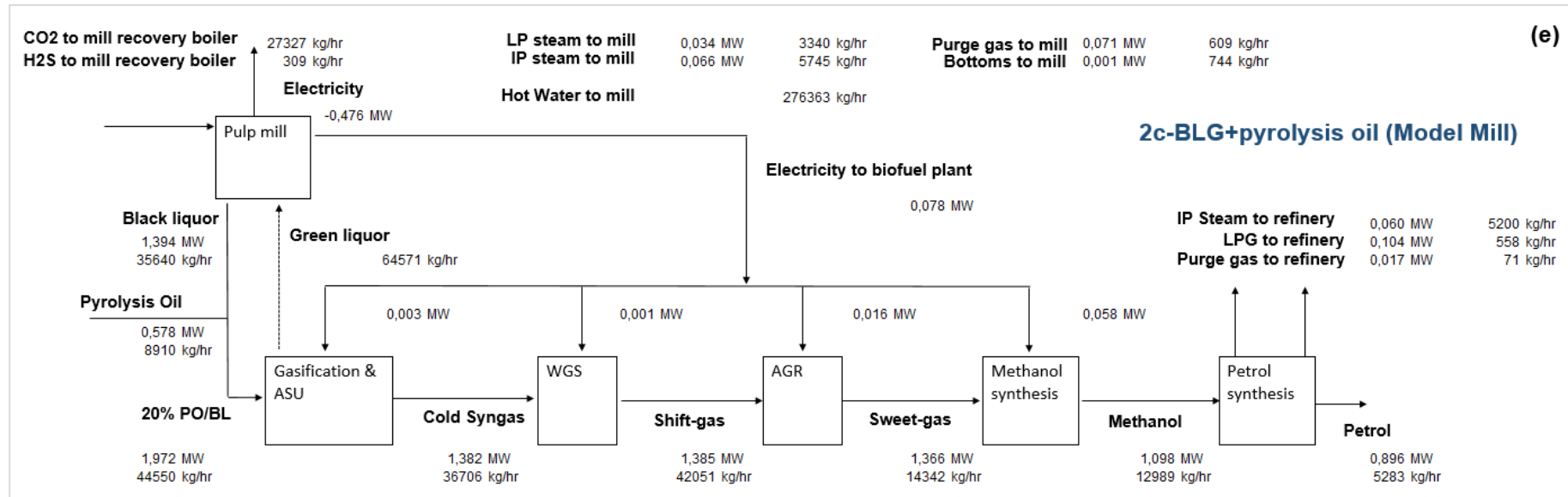
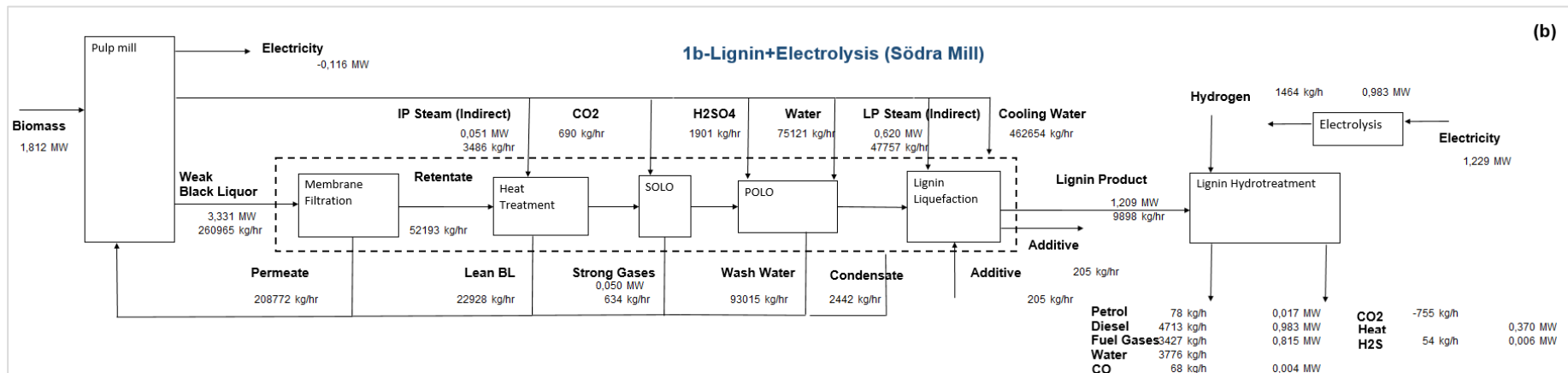
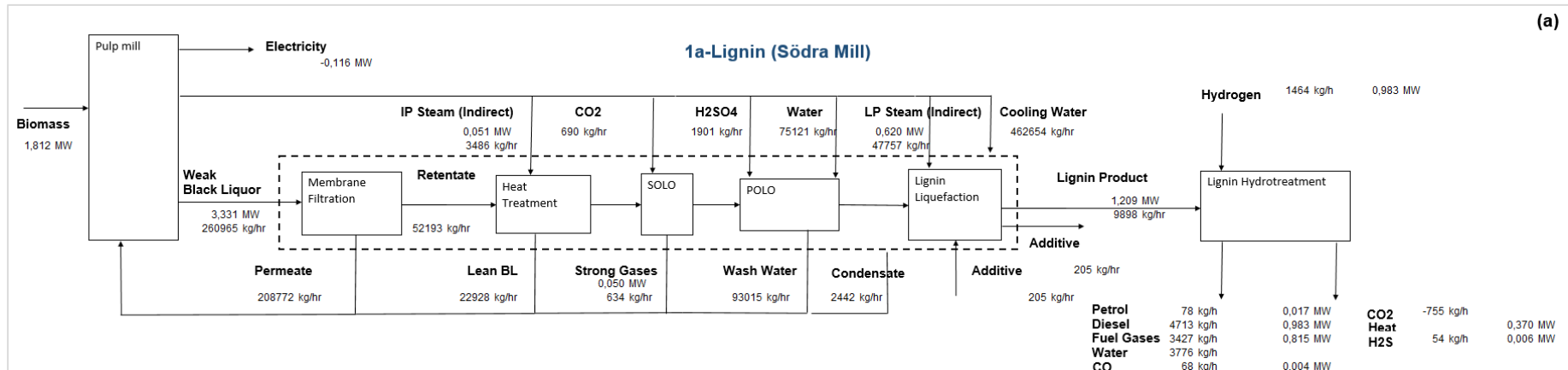
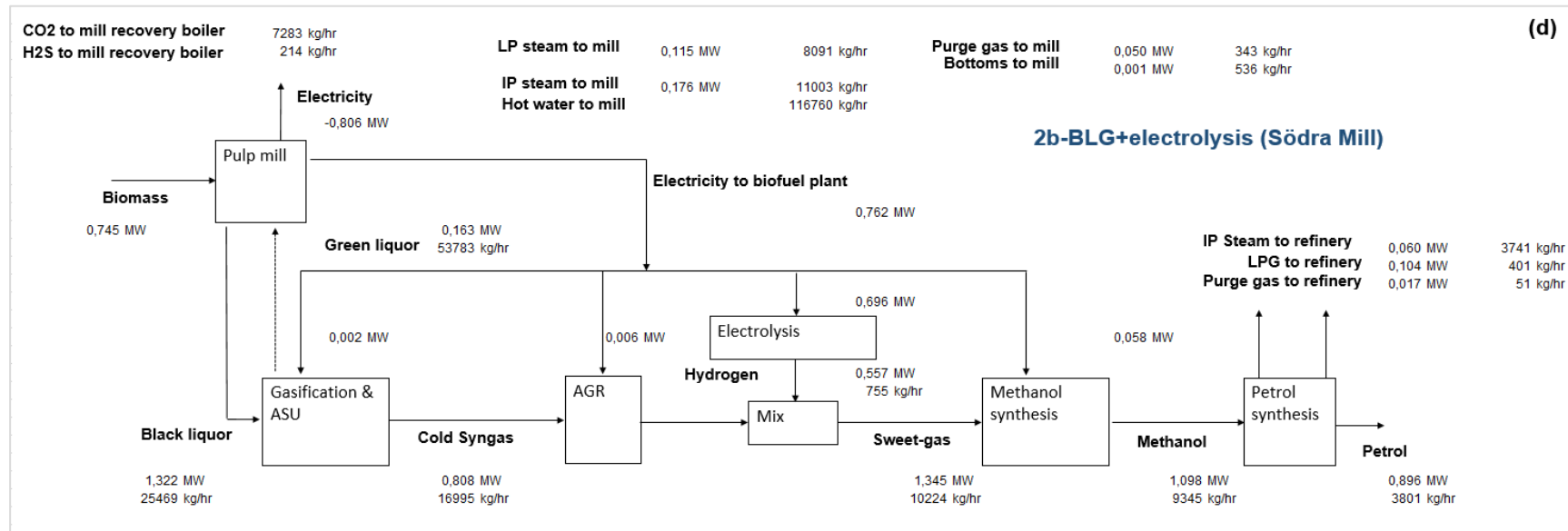
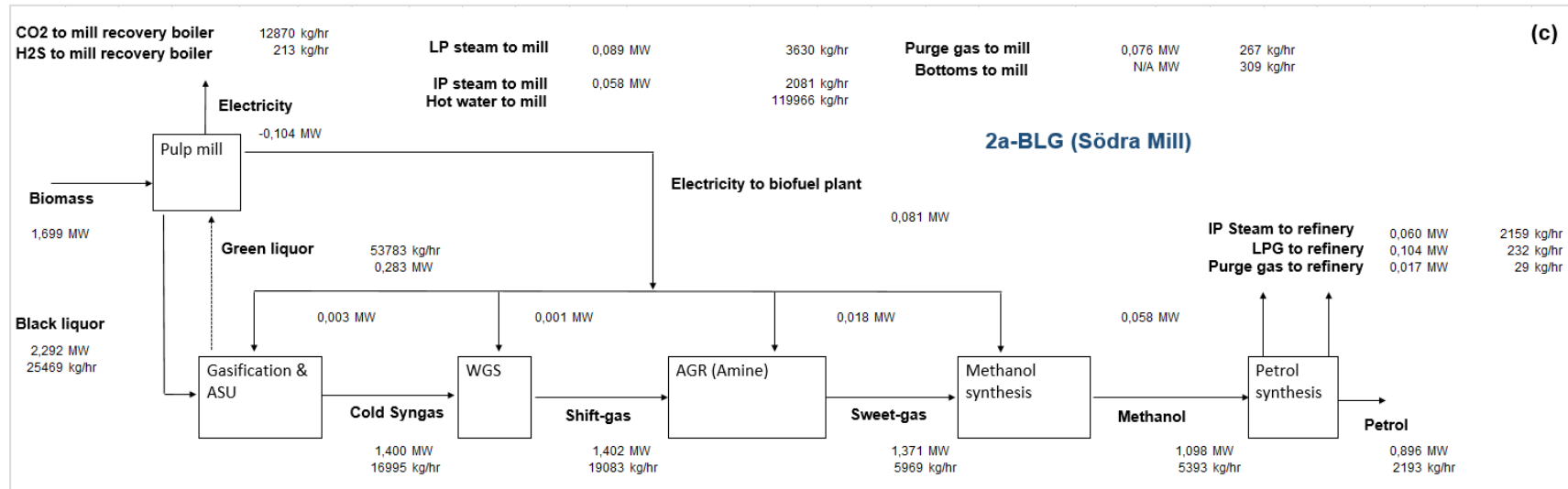


Figure C- 1. Energy balance for (a) 1a-Lignin, (b) 1b-Lignin+electrolysis, (c) 2a-BLG, (d) 2b-BLG+electrolysis and (e) BLG+pyrolysis oil in the Model Mill integration case normalized to 1 MW HHV of biofuel products. Selected mass flows are also shown for reference.

DROP-IN FUELS FROM BLACK LIQUOR PART STREAMS –  
BRIDGING THE GAP BETWEEN SHORT- AND LONG-TERM TECHNOLOGY TRACKS



DROP-IN FUELS FROM BLACK LIQUOR PART STREAMS –  
BRIDGING THE GAP BETWEEN SHORT- AND LONG-TERM TECHNOLOGY TRACKS



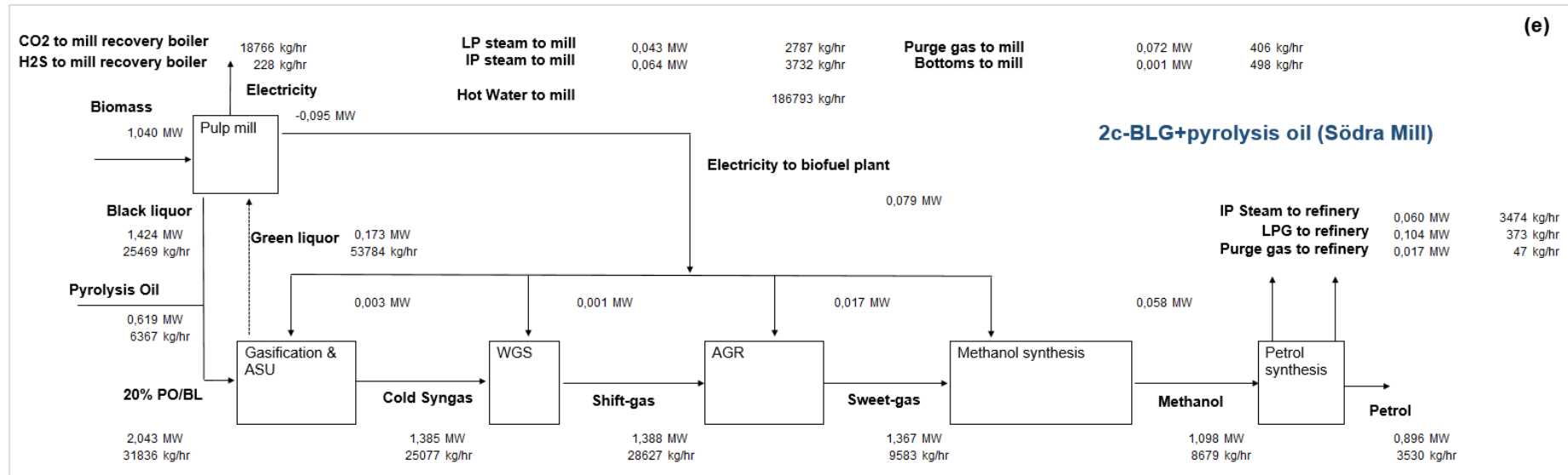
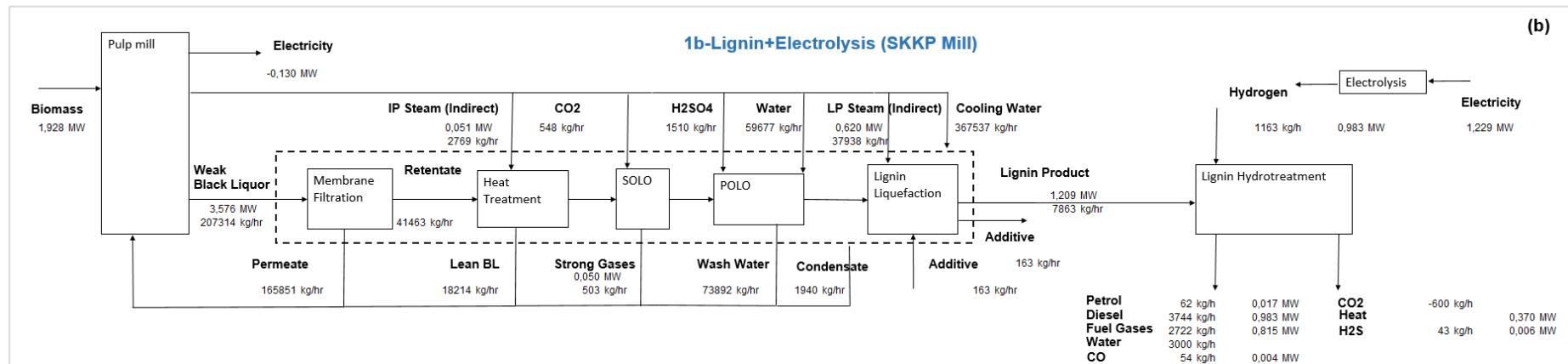
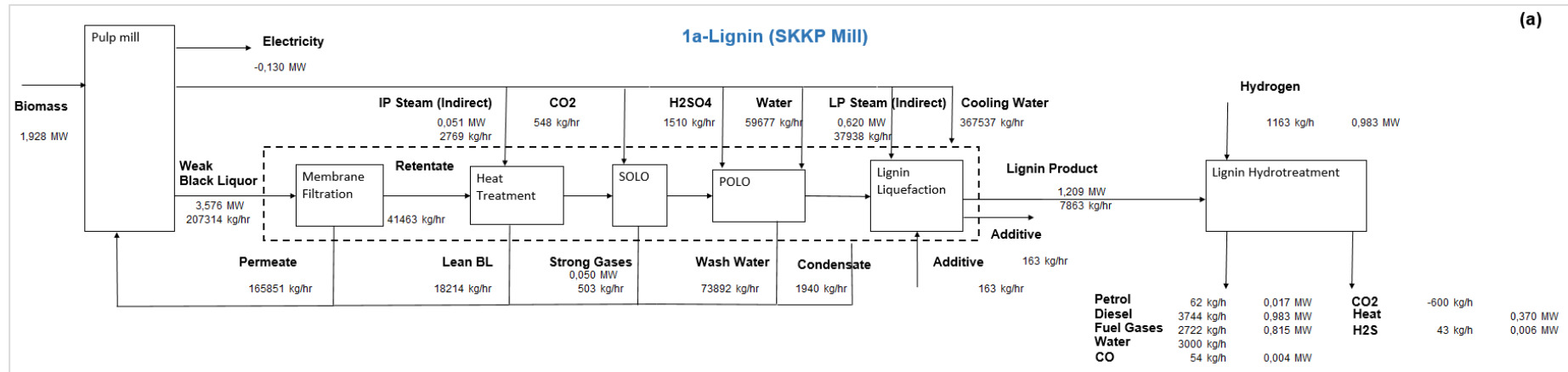
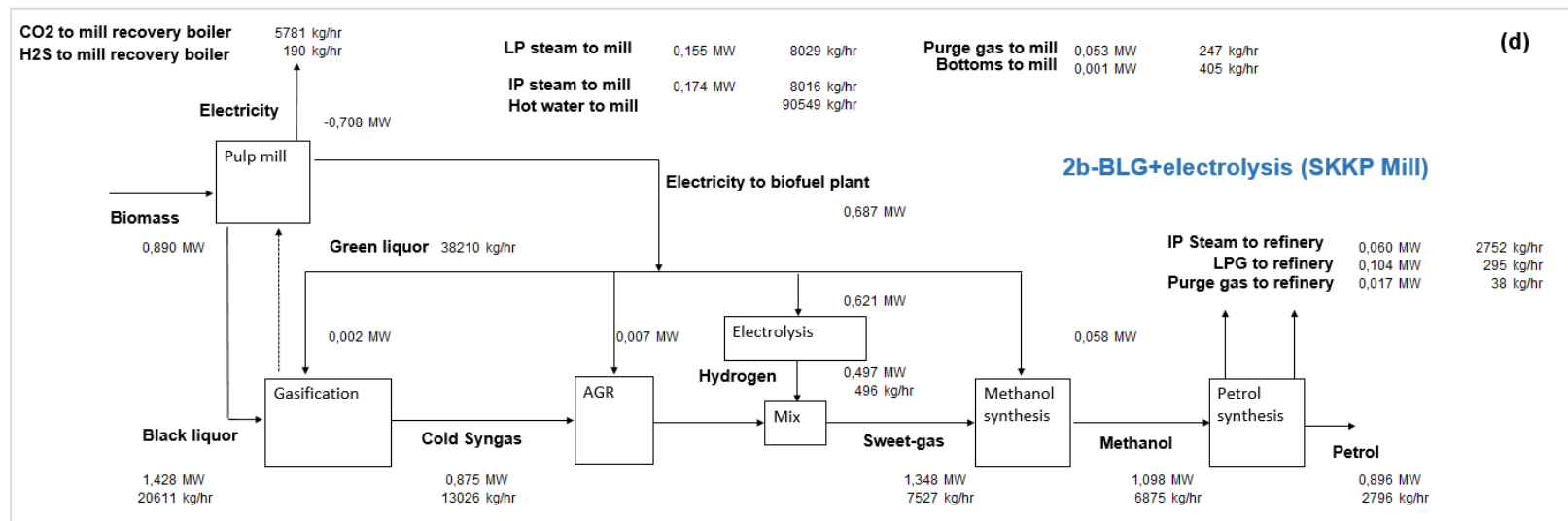
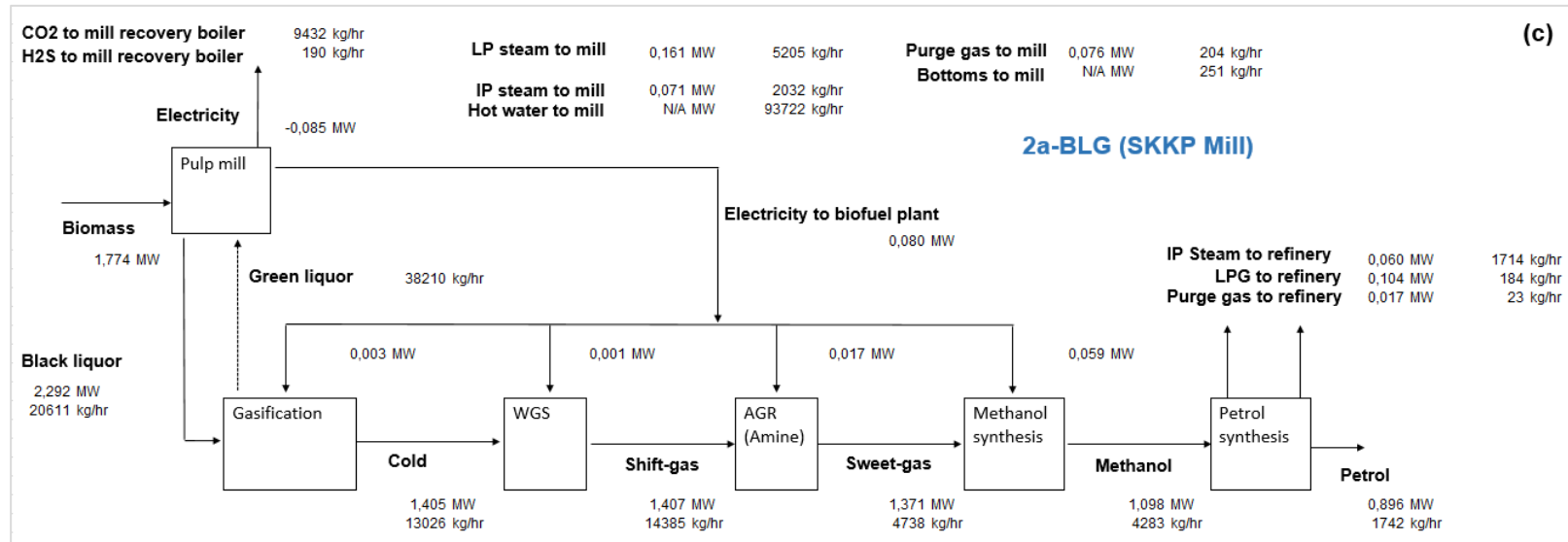


Figure C- 2. Energy balance for (a) 1a-Lignin, (b) 1b-Lignin+electrolysis, (c) 2a-BLG, (d) 2b-BLG+electrolysis and (e) BLG+pyrolysis oil in the Södra Mill integration case normalized to 1 MW HHV of biofuel products. Selected mass flows are also shown for reference.

DROP-IN FUELS FROM BLACK LIQUOR PART STREAMS –  
BRIDGING THE GAP BETWEEN SHORT- AND LONG-TERM TECHNOLOGY TRACKS



DROP-IN FUELS FROM BLACK LIQUOR PART STREAMS –  
BRIDGING THE GAP BETWEEN SHORT- AND LONG-TERM TECHNOLOGY TRACKS



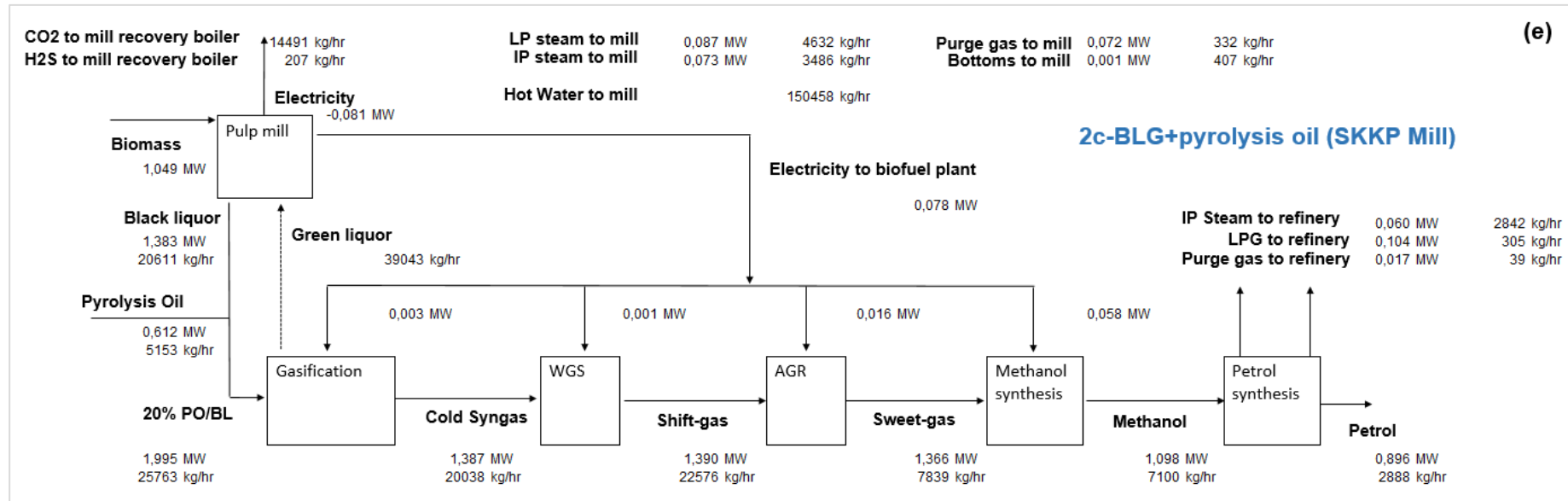


Figure C- 3. Energy balance for (a) 1a-Lignin, (b) 1b-Lignin+electrolysis, (c) 2a-BLG, (d) 2b-BLG+electrolysis and (e) BLG+pyrolysis oil in the SKKP Mill integration case normalized to 1 MW HHV of biofuel products. Selected mass flows are also shown for reference.

