**Final report** 

# ELECTROLYSIS ASSISTED BIOMASS GASIFICATION FOR BIOFUELS PRODUCTION

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## PREFACE

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## SUMMARY

### FLEXIBLE PRODUCTION OF DROP-IN BIOFUELS FROM FOREST INDUSTRY BYPRODUCTS AND RENEWABLE ELECTRICITY

The demand of drop-in biofuels compatible with the current transportation fleet in Sweden is expected to increase dramatically in the coming decade to meet the policy goals for 2030, 70% emissions reduction from domestic road transport, and for a fossil-free society by 2045. Though, the penetration of electric and hybrid vehicles in passenger fleet is increasing at promising rate, long distance trucks, aviation and marine sectors will depend on the renewable replacement for the foreseeable future.

The Swedish forest industry has annual capacity about18–19 million cubic meter of sawnwood and 9 million tons of pulp. In a typical Nordic sawmill about half of the incoming timber becomes byproduct in the form woodchips, sawdust and bark. Thus, utilization of these residual biomass for the production biofuels has large potential to contribute in the transition from fossil-based economy to a circular one.

Gasification is a promising pathway for converting forest residues to renewable transportation fuels and chemicals. The main challenge with gasification based pathways is that it requires multiple steps to clean and condition the produced synthesis gas prior to its final conversion into liquid fuels. Traditional technologies for cleaning and conditioning are capital intensive with high investment needs, which puts limitation on deplyoment of small to medium scale process. Integration of Molten Carbonate Electrolysis Cell (MCEC) improves the performance of gas cleaning and conditioning steps and brings the possibility of realizing economically feasibility even at small scale.

MCEC technology is a power to gas conversion process, which can enable long-term storage of intermittent renewable electricity in the form of liquid fuels. MCEC is flexible technique, for example when operated in a fuel cell mode it produces electricity.

#### OBJECTIVE

The project "Electrolysis assisted biomass gasification for biofuels production" evaluates the potential for converting sawmill byproducts to sustainable aviation fuel onsite with the help of gasification, gas conditioning and Fischer-Tropsch synthesis where the MCEC plays central role.

#### WHAT IS A MOLTEN CARBONATE ELECTROYSIS CELL (MCEC)?

MCEC is electrochemical process that builds on the concept of well-proven technology molten carbonate fuel cell (MCFC), currently available for electricity production at capacities as high as 3 MW. MCEC is essentially a reverse operation of MCFC. In this work, MCEC uses electricity and steam to increase hydrogen content of the syngas prior to its synthesis and upgrade to transport grade hydrocarbons in the final stage.

The chemical transformation in the MCEC can be described as a process in which electrons interact in the chemical reactions at the two electrodes, anode and cathode. Water  $(H_2O)$  and carbon dioxide  $(CO_2)$  are supplied to the cathode where the valuable syngas components hydrogen  $(H_2)$  and carbon

monoxide (CO) are formed. Carbonate ions are transported through an electrolyte consisting of molten carbonate salts from the cathode to anode and decompose into oxygen ( $O_2$ ) and  $CO_2$ . The oxygen can be separated and used as oxidant in the gasification or sold to generate income.



### EVALUATED PROCESS CONFIGURATIONS

The project evaluated different process configurations for integrated production of sustainable aviation fuel (SAF) at a sawmill facility using process simulation developed in MATLAB® Simulink and UniSim Design®. Production cost of total Fischer-Tropsch liquid (SAF, naphtha and diesel ranges) is evaluated as economic performance indicator of the cases analyzed. The size of the Nordic sawmill is chosen to meet feedstock demand of the gasification process (capacity 20 MW syngas LHV) and the heat demand of lumber drying kiln.



The SAF production facility includes mainly a gasification process, primary gas cleaning unit (particle filter), a steam reformer for methane (SMR – steam methane reformer), a MCEC unit, a final gas cleaning step to remove impurities and acid gas (among others  $H_2S$ ,  $CO_2$ ) and a Fischer-Tropsch synthesis unit (converts  $H_2$  and CO to hydrocarbons). The MCEC essentially replaces water gas shift (WGS) process, traditionally used in gasification-based biofuel production to adjust syngas composition to desired  $H_2/CO$  ratio before the final synthesis step.

The study evaluated eight cases based on three gasification technologies and two process configurations. The gasificaton technologies are:

- Dual Fluidized Bed (DFB) (indirectly heated)
- Bubbling Fluidized Bed (BFB) (directly heated)
- WoodRoll (WR) process

The process configurations differ by the heat source to the integrated reformer that converts methane and light hydrocarbons in syngas into CO and  $H_2$  components. The configurations are:

- a) Traditional steam reformer (SMR) part of the incoming gas and tar are combusted to generate heat to maintain reformer operating temperature
- b) Electric resistance heated steam reformer (eSMR) reactor surfaces heated with electric resistance

#### MAIN FINDINGS

The project have established operating range of the electrochemical unit for handling biomass derived syngas from three gasification technologies. The MCEC can not only replace the water-gas shift process of the conventional syngas conditioning step, but enables increased product throughput by as much as 15–31%. The MCEC also opens the option for linking the process to external carbon utilization in the context of power-to-X configurations.

The capacity of the plants considered in this study are for first-of-its-kind demonstration scale, about 30 MW LHV biomass feedstock, which resulted in about 5–7 kTPY (kilo-tons per year) production of biojet fuel and 4–5 kTPY of other hydrocarbon components, naphtha and diesel range. The economic assessment resulted in total FT liquids production cost range 1400–1600 SEK/MWh (SMR configurations) and 1300–1500 SEK/MWh (eSMR configurations). The processes turned out to be capital intensive with specific investment range 59–71 million SEK/MW FT liquid. About 55% of the production cost derives from the investment.



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## SAMMANFATTNING

### FLEXIBEL PRODUKTION AV BIODRIVMEDEL FRÅN RESTBIOMASSA OCH FÖRNYBAR ELKRAFT

Dagens växande elektrifiering av personbilar betraktas som en viktig del i övergången från fossilbaserade till förnybara transportbränslen, framför allt inom den landbaserade transportsektorn. En elektrifiering av luftfarten och marina transporter är inte lika självklar och här kommer förnybara biodrivmedel att spela en viktig roll för att möjliggöra förverkligandet av Sveriges nationella mål att minska utsläppen från inhemsk vägtransport med 70 % år 2030 och ett fossilfritt samhälle 2045.

Skogsindustrin i Sverige har idag en produktionskapacitet på 18-19 miljoner m<sup>3</sup> sågat trävirke och 9 miljoner ton pappersmassa. Mer än hälften av det timmer som levereras till ett typiskt nordiskt sågverk kommer att sluta som biprodukter i form av restbiomassa (flis, bark och sågspån). Nyttiggörandet av denna restbiomassa, som råvara för produktion av biobaserade drivmedel, har därför en stor potential att kunna stå för en betydande andel av framtida behov.

En strategiskt viktig teknik för omvandling av restbiomassa till bl.a. biodrivmedel är förgasning. Men konstruktionen av effektiva efterföljande processer för gasrening och -uppgradering innan biodrivmedel kan produceras är en utmaning. Traditionell teknik för processerna är generellt kapitalintensiv med hög initial investeringskostnad, vilket hindrar uppförande av mindre till medelstora processer. En integrering av den nya tekniken smältkarbonatelektrolyscell (MCEC) erbjuder en möjlighet att effektivisera gasrening och uppgradering och på så sätt möjliggöra processer som är kostnadseffektiva även i mindre skala.

MCEC-tekniken är en elkraft-till-gas-process, vilket möjliggör en större andel intermittent förnybar elkraft i energisystemet då den elkraften kan lagras i form av kemiska energibärare. Då MCEC-tekniken även kan köras i omvänt läge, som en bränslecell (MCFC), går det även att producera elektricitet beroende på behoven i samhället.

### SYFTE

Inom projektet utvärderades potentialen att omvandla restbiomassa från sågverk till biojetbränsle och andra kemiska energibärare med hjälp av förgasningsteknik och processer för gasrening och uppgradering där MCEC-tekniken integrerats. Information om driftsförhållanden och möjliga vägar till integration i t.ex. sågverk med tillgång på restbiomassa är viktigt för att stimulera introduktionen av processer för produktion av förnybart bränsle.

### VAD ÄR EN SMÄLTKARBONATELEKTROLYSCELL (MCEC)?

MCEC-tekniken bygger på den väl beprövade och redan kommersiellt introducerade smältkarbonatbränslecellen (MCFC) och säljs idag i enheter på 3 MW för installation i kraftverk för elproduktion.

Den elektrokemiska enheten MCEC förbrukar elektrisk energi och ånga för att öka vätgasinnehållet i den syntesgas som i ett senare steg används i det slutliga kemiska syntessteget för framställning av t.ex. biojetbränsle.

Den kemiska omvandlingen i en elektrokemisk smältkarbonatelektrolyscell (MCEC) kan förenklat beskrivas som process där elektroner deltar i kemiska reaktionerna vid två elektroder. Vatten (H<sub>2</sub>O) och koldioxid (CO<sub>2</sub>) matas till katoden där främst vätgas och koloxid bildas, värdefulla syntesgasmolekyler som kan vidareförädlas till olika kemiska energibärare och transportbränslen. Karbonatjoner transporteras i en elektrolyt som består av en saltkarbonatsmälta (600-800°C) från katoden till anoden och sönderdelas till syrgas (O<sub>2</sub>) och koldioxid (CO<sub>2</sub>). Syrgasen kan avskiljas och användas i den termiska förgasningen.



#### STUDERADE PROCESSALTERNATIV

Inom projektet utvärderades en rad olika processkonfigurationer för produktion av biojetbränsle genom processmodellering på olika nivåer och genom processintegrering i ett sågverk. Den totala produktionskostnaden för biojetbränsle och andra flytande Fischer Tropsch-produkter (FT) utvärderades för varje enskild processkonfiguration för att möjliggöra en jämförelse mellan de olika fallen.

I studien användes en för Norden typisk storlek på sågverk och utifrån storleken anpassades skalan för övriga processer i anläggningen för biojetbränsleproduktion i relation till sågverkets övriga energianvändning. Tekniskt innebär detta en dimensionering av förgasaren (20 MW syntesgasproduktion) för syntesgasproduktionen och en förbränningspanna som körs för att möta kraven på värme i det integrerade systemet och för att överhetta mättad ånga från anläggningen för biojetbränsleproduktion.



Anläggningen för biojetbränsleproduktion inkluderar huvudsakligen en förgasningsprocess, primär gasrengöring (partikelseparering), en ångreformer för metangas (SMR, steam methane reformer), en integrerad MCEC-enhet, ett steg för slutlig gasrening (bl.a.  $CO_2$  separering) och slutligen processen för FT-syntes (som använder vätgas, H<sub>2</sub>, och koloxid, CO, vid syntesen). Den integrerade MCEC-enheten ersätter en processenhet för vattengasskift (WGS), som traditionellt används för att skräddarsy sammansättningen av H<sub>2</sub> och CO i syntesgasen innan t.ex. FT-syntesen.

Studien innefattade åtta olika processkonfigurationer där tre olika förgasningsalternativ ingick:

- Tvåbäddsförgasare (DFB, Dual Fluidized Bed). Den mest använda tekniken för indirekt förgasning av biomassa, huvudsakligen uppbyggd av två fluidiserade bäddreaktorer (en för förgasning och en för förbränning) som sammankopplas genom bäddmaterialet (sand), som cirkulerar mellan de två reaktorerna. Bäddmaterialet värms upp i förbränningsreaktorn vilket möjliggör förgasning med enbart ånga i förgasningsreaktorn.
- Bubblande fluidiserad bädd (BFB, Bubbling Fluidized Bed). Teknik som består av endast en fluidiserad bädd där värmebehovet i reaktorn tillgodoses genom direkt förbränning av syrgas i bädden. Ånga används vid produktion av syntesgas för att öka mängden vätgas i syntesgasen.
- WoodRoll (WR): Tekniken innefattar flera olika steg där de viktigaste är torkning av råmaterial, pyrolys och förgasning i en suspensionsförgasare. Suspensionsförgasaren värms indirekt genom att gas från pyrolysen bränns. Biokol från pyrolysen förgasas med ånga i suspensionsförgasaren och omvandlar biomassa till en relativt ren förnybar gas med ett måttligt behov av gasrengöring, jämfört med t.ex. DFB och BFB.

Studien visade att en ångreformer för metangas som omvandlar metan och högre kolväten i produktgasen från förgasaren till CO och H<sub>2</sub> bör ingå i processen för att uppnå en kostnadseffektiv process. Därför inkluderades även två olika alternativa SMR-processer:

• En traditionell ångreformer för metangas (SMR): En del av gasen in till reformen förbränns för att generera värmen som krävs för att upprätthålla driftstemperaturen

• En elektriskt uppvärmd ångreformer för metangas (eSMR - Electric resistance heated Steam Methane Reformer): Reaktorns värmebehov tillgodoses genom elektrisk uppvärmning.



#### VAD HAR VI KOMMIT FRAM TILL?

Projektet har tagit fram information om driftintervall för den elektrokemiska enheten MCEC när den används i en process med en produktgas producerad med tre olika biomassaförgasningstekniker. Resultaten pekar på tydliga fördelar med MCEC-tekniken jämfört med den traditionella vattengasskiftsprocessen. MCEC-tekniken förbättrar utbytet genom en bättre kolomvandlingsgrad och öppnar samtidigt upp möjligheten för en direkt användning av extern koldioxid i en s.k. "power-to-X"-process. Sammantaget går det att dra följande slutsatser från

studierna av processerna inom projektet:

- För alla de utvärderade fallen fanns metan i den utgående gasen från katoden, som beroende på önskad slutprodukt antingen är attraktiv eller inte. Vid en syntesgasprocess, som t.ex. FT-syntes, är en ångmetanreformer (SMR) därför oundviklig för att uppnå ett acceptabelt utbyte.
- Graden av aktivitet hos MCEC-processteget varierar beroende på vilken produktgassammansättning gasen har. Fördelarna är mer uppenbara för förgasningsprocesser där produktgasens innehåll av vätgas är lägre, som för BFB-förgasare, då produktionen av vätgas i MCEC-processteget ger en högre aktivitet.
- MCEC-tekniken möjliggör en ökad produktgenomströmning med så mycket som 15–31 % beroende på processkonfiguration, jämfört med den traditionella vattengasskiftsprocessen.
- Elektrisk uppvärmning av ångmetanreformen (eSMR) förbättrar det totala utbytet flytande FTprodukter med 9–13 % jämfört med de traditionella SMR-fallet, och så mycket som 15–31 % jämfört med den traditionella vattengasskiftsprocessen.



- Storleken på de processkonfigurationer som utvärderades har en kapacitet på cirka 30 MW biomassa, vilket skulle resultera i en produktion av cirka 5–7 kton biojetbränsle per år, och 4–5 kton per år av andra kolväteföreningar såsom nafta och diesel.
- Den totala produktionskostnaden för den flytande fraktionen av FT-produkterna hamnar mellan 1400–1600 SEK/MWh för SMR-konfigurationen och 1300–1500 SEK/MWh för eSMR-konfigurationen.

Jämfört med de produktionskostnader som rapporterades i Biojetutredningen (Wetterstrand et al 2019), ligger resultaten i mitten av det intervall som utredningen utvärderade, dvs. 1850 SEK/MWh för 2021 och 1235 SEK/MWh<sup>1</sup>för 2030.

<sup>&</sup>lt;sup>1</sup> Räknade med jetbränsle värmevärde 35 MJ/liter

## ABBREVIATIONS

Abbreviation	Definition
AGR	Acid gas removal
BFB	Bubbling fluidized bed
BFBe	Bubbling fluidized bed best efficiency case
BFBy	Bubbling fluidized bed best yield case
CCS	Carbon capture and storage
CFB	Circulating fluidized bed
DFB	Dual fluidized bed
eSMR	Electric resistance heat steam methane reformer
FT	Fischer Tropsch
FTP/L	Fischer Tropsch products/liquid
FTR	Fischer Tropsch reactor
FTS	Fischer Tropsch synthesis
kTPY	kilo tons per year
MCEC	Molten carbonate electrolysis cell
SAF	Sustainable aviation fuel
SMR	Steam methane reformer
WR	WoodRoll
OHC	Other hydrocarbons
Ĥ	Enthalpy flow, kW
'n	Molar flow, kmol/s
F	Faraday constant, 95 485 A·s/mol
G	Gibbs free energy, kJ/kmol
p	Partial pressure, kPa
R	Ideal gas constant, 8.314 kJ/kmol K
Т	Temperature, K
TFCI	Total fixed capital investment
LPS	Low-pressure steam
LHV	Lower heating value
IPS	Intermediate pressure steam
MPS	Medium pressure steam
SMLPS	Sawmill low-pressure steam

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## 1 INTRODUCTION

Electrification of the transportation fleet is growing and considered as one of the best options to transition from fossil-based fuels, especially for passenger vehicles where its share has increased at a promising rate. Nonetheless, the role of biofuels cannot be overlooked given the varying nature of the transportation mode, such as heavy-duty trucks, aviation and marine where the need for carbon neutral substitution will remain for the foreseeable future. Thus, biofuels are expected to enable the realization of Sweden's national targets for fossil free vehicle fleet and carbon neutral society, for 2030 and 2045, respectively. Sweden is endowed with a vast forest coverage making it a major forest industry nation with production capacities of about 18.4 million cubic meter sawnwood (Eurostat, 2018b) and 8.6 million tonnes pulp (Eurostat, 2018a) in 2018. Evidently its forest industry can benefit from timely evaluation and knowledge of emerging technologies in the biofuel's arena. Early identification of resource effective, economically feasible and least carbon footprint value chains can facilitate deployment of commercial scale production of biofuels in the short term.

Biomass gasification is considered a strategic technology for converting residues from forest industry to transportation fuels, chemicals or electricity. A significant challenge in biomass gasification is the engineering of an efficient product gas cleaning and conditioning process before utilizing the produced synthesis gas in a syngas process. Consequently, it is essential to find ways to improve the process economics. In a previous study (Mesfun et al., 2019) evaluated the techno-economic potential of integrating molten carbonate electrolysis cell (MCEC) technology in a dual fluidized bed gasification system for bio-SNG production. The MCEC was integrated in the cleaning and conditioning process to reform hydrocarbons in the gas and to boost the production of a tailored syngas prior to bio-SNG production utilizing intermittent renewable power. The results from the study indicate that the production of SNG can be boosted by approximately 50-60% without the need of an additional carbon source. The study also demonstrated how MCEC could be utilized in a power-to-gas process, enabling a greater share of intermittent renewable power into the energy supply system.

This present work investigates the potential to valorize sawmill byproducts onsite using gasification technology enhanced by MCEC. The aim is to investigate the performance of MCEC technology to boost production of renewable transportation fuels via gasification of woody biomass and a Fischer Tropsch (FT) synthesis process. The results from the study may provide with information of importance to advance the engineering of gas-to-liquid transportation fuels utilizing renewable electricity.

## 2 PROCESS DESCRIPTION

Figure 1 illustrates a schematic of a biomass gasification process for production of transportation fuel, including primary gas cleaning, an integrated MCEC process and final trace gas cleaning. Starting from left, the process consist of a; 1) biomass gasifier; 2) a primary gas cleaning step, including removal of particle, tar and other impurities (H<sub>2</sub>S, etc) to concentration levels as specified by the MCEC; 3) the MCEC process, together with a  $CO_2/O_2$  separation unit; 4) a final gas cleaning step for removal of e.g. trace elements; 5) a chemical synthesis process.



(A) H<sub>2</sub>, CO, CO<sub>2</sub>, H<sub>2</sub>O. Hydrocarbons, tar, H<sub>2</sub>S, etc at concentration levels specified by MCEC
 (B) H<sub>2</sub>, CO, trace impurities

# Figure 1. A schematic of a biomass gasification process for production of transportation fuels, including a MCEC process step.

The focus of the project is the MCEC as the key technology introduced. Unlike other types of electrolysis technologies that require pure stream of water/steam (e.g. PEM, alkaline) or mixture of steam and CO<sub>2</sub> (e.g. solid oxide cells), the MCEC can utilize streams with wider variety of species such as a product gas from a gasification process. The MCEC process allows for a certain degree of hydrocarbon reforming, conditioning of the H<sub>2</sub>/CO ratio via WGS and carbon dioxide separation from the product gas in one process step. Another advantage is the production of oxygen that can be used in the gasification process, enabling the use of gasification technologies where air separation is needed for syngas production. Nevertheless, there are limitations for the concentration of tar and other impurities and, therefore, the primary gas cleaning process configuration, before the product gas entering the MCEC, could vary depending on the biomass feedstock and gasification technology selected. The raw product gas from a gasifier undergo several process steps of gas cleaning and upgrading to achieve tar and particulates-free syngas stream. The primary cleaning process is comprised of a hot gas cleaning step to primarily remove particles that follow the gas. Depending on the gasification technique, hydrocarbons, tar and impurities that can harm the electrochemical unit must be removed. This can be performed using e.g a RME scrubber or a catalytic tar reformer (Guan et al., 2016).

## 2.1 PROCESS CONFIGURATION EVALUATED

Figure 2 shows a simplified block diagram of the process configuration investigated in this study, a more detailed process flow diagram is presented in Appendix A. The main difference from Figure 1 is the introduction of a reformer before the MCEC, motivated by the following two reasons:

First, the syngas after hot gas cleaning may contain tar and hydrocarbons such as methane,  $C_2H_x$  (ethyne, ethylene and ethane) and benzene, which significantly reduce the overall carbon conversion from biomass to finished products when left unutilized. Initial evaluations were made to

investigate potential reforming activity in the MCEC by supplying a syngas containing methane 3– 8% vol. at temperatures 700–800°C. The equilibrium results indicated that methane reforming was not as expected and significant amount of methane was present at the fuel electrode.

Second, the Fischer Torpsch synthesis, the chemical synthesis process chosen, produces tail-gas rich in light-hydrocarbon components,  $C_1$ - $C_4$ . Therefore, it was considered important to reform these hydrocarbons to CO and H<sub>2</sub> components and subsequently tailor the syngas in the MCEC to achieve an optimal H<sub>2</sub> to CO ratio for upgrading.



(B) H<sub>2</sub>, CO, trace impurities

Figure 2 A. schematic of the process configuration evaluated in this work.

## 2.2 MOLTEN CARBONATE ELECTROLYSIS CELL (MCEC)

Experimental investigations have shown that high-temperature electrolysis of MCEC (Hu et al., 2016a, 2017) benefits from both thermodynamic and kinetic properties of the electrochemical process resulting in an improved overall conversion efficiency. High temperature also reduces the electricity required to drive the electrolysis process because part of the reaction energy is supplied in the form of heat. Besides, MCEC often use molten alkali metal carbonate salts as electrolyte, mandating operational temperatures in the range 600–800°C.

The working principle of MCEC is shown in Figure 3. In the electrochemical reactions that take place in the cell, electrical power is converted into energy-rich gas, or in case of the reverse operation as a molten carbonate fuel cell (MCFC), producing electrical power. Hydrogen is produced on the cathode side through the electrochemical reaction of water, carbon dioxide and 2 electrons forming hydrogen and a carbonate ion. The carbonate ions migrate in the electrolyte from one electrode to the other, where they are decomposed to oxygen and carbon dioxide, facilitating a separation of the carbon dioxide from the product gas. After separation the carbon dioxide is either recycled to the MCEC cathode or separated for sequestration. Oxygen can be utilized in the thermal gasification. In the output stream from the catode, the CO content can be controlled through the water-gas shift reaction and thus enabling a control of the H<sub>2</sub>/CO ratio.



#### Figure 3. MCEC operating principle (Hu, 2016).

The high operating temperature of the MCEC can potentially enable internal reforming of methane and other hydrocarbons, especially if Ni is used as electrode (cathode), which can also improve the overall efficiency of the system. Information about acceptable concentration levels of hydrocarbons is not available, but values for MCFC, as indicated in Table 1, are probably reasonable approximations. Values for selected impurities are also specified in Table 1.

Contaminants	Qualitative tolerance	Comment
Saturated hydrocarbons including methane (%-vol)	< 10	Decomposed (Dayton et al., 2001)
Aromatic and cyclic hydrocarbons (%-vol)	< 0.5	Decomposed (Dayton et al., 2001)
H <sub>2</sub> S (ppmv)	< 5	Recoverable (Dayton et al., 2001; Remick, 1986)
NH <sub>3</sub> (ppmv)	<10 000	No effect (Srinivasan, 2006)
HCl (ppmv)	< 0.1	Long term effects possible (Srinivasan, 2006)

Table 1. Acceptable concentration levels of hydrocarbons and other contaminents for MCFC

## 2.3 A MODEL OF THE MCEC

The behavior of the MCEC has been simulated with a numerical model developed in MATLAB/Simulink. The model of the MCEC stack used in the study is a zero-dimensional model in which molar and energy balances, chemical reactions (steam reforming and water gas shift) and electrochemical reactions (with the related aspects on current and voltage) are taken into account.

The MCEC stack model is part of a Simulink model developed for this project, Figure 4. The input to the fuel electrode consists of the mix among three streams: a syngas with given composition, a fixed amount of steam (usually given in proportion to the water vapor content of the syngas) and the recycled carbon dioxide from the oxygen electrode outlet. The input current to the MCEC stack is determined by a control system so that the proportion between the molar flows of hydrogen and carbon monoxide at fuel electrode outlet is equal to 2.



Figure 4. MCEC Simulink model.

The inputs to the MCEC stack model are:

- Molar flow rates of the components of the gas at the inlet of the fuel electrode (cathode) (the seven chemical species involved in the model are: CH<sub>4</sub>, H<sub>2</sub>, H<sub>2</sub>O, CO, CO<sub>2</sub>, N<sub>2</sub> and O<sub>2</sub>)
- Pressure and temperature of the gas at the inlet of the fuel electrode (the cell stack is assumed to operate at atmospheric pressure)
- Input electric current to the cell stack

Parameters of the stack are the number of cells and cell size, which are needed to calculate current density.

The outputs of the MCEC stack model are:

- Molar flow rates of the components of the gas at the outlet of the fuel electrode
- Molar flow rates of the components of the gas at the outlet of the oxygen electrode
- Cell stack temperature (which is also the temperature of the gases at the outlet of the electrodes)
- The electric power required by the cell stack

The calculation of the outputs requires the convergence of an iterative procedure, since all output quantities are heavily interdependent and cannot be determined separately from the knowledge of the inputs only. Details on the equations considered in this iterative procedure are given in the following subsections.

#### 2.3.1 Chemical reactions

Steam reforming and water gas shift are supposed to occur along the fuel electrode.

$CH_4 + H_2O \rightleftharpoons CO + 3H_2$	steam reforming (subscript R)		
$\rm CO + H_2O \rightleftharpoons \rm CO_2 + H_2$	water gas shift (subscript S)	(R2)	

Both reactions are considered to be in their chemical equilibrium at the outlet of the fuel electrode (cathode), i.e. at the composition of the gas at fuel electrode outlet and at cell stack temperature. For each reaction, R1 and R2, the equilibrium constant K is a function of the temperature (cell stack temperature  $T_{cs}$ ) and the variation of standard Gibbs free energy, and is also equal to the product of the partial pressures of the species,  $p_i$ , involved raised at the power of the respective stoichiometric coefficients in the reaction.

$$K_{R} = \frac{\Delta G_{R}^{0}}{RT_{cs}} = \left(\frac{p_{H_{2}}^{3} p_{CO}}{p_{CH_{4}} p_{H_{2}O}}\right)_{fe,out}$$
(1)

$$K_S = \frac{\Delta G_S^0}{RT_{cs}} = \left(\frac{p_{CO_2} p_{H_2}}{p_{CO} p_{H_2O}}\right)_{fe,out}$$
(2)

#### 2.3.2 Electrochemical reactions

The electrochemical reactions (E) occurring at the electrodes are:

$\mathrm{H_2O} + \mathrm{CO_2} + 2\mathrm{e}^{-} \rightarrow \mathrm{H_2} + \mathrm{CO_3}^{=}$	fuel electrode (cathode)	(R3)
$2\mathrm{CO}_2 + 2\mathrm{e}^{-} \rightarrow \mathrm{CO} + \mathrm{CO}_3^{-}$	fuel electrode (cathode)	(R4)
$\mathrm{CO}_3^{=} \rightarrow \mathrm{CO}_2 + 0.5\mathrm{O}_2 + 2\mathrm{e}^{-}$	oxygen electrode (anode)	(R5)

The electrochemical reaction (R4) producing carbon monoxide and carbonate ions from  $CO_2$  molecules at the fuel electrode is neglected because it is much slower than the reaction R3 (Pérez-Trujillo et al., 2020).

The molar flow rates involved in the electrochemical reactions depend on the input current I (A) to the cell stack through the Faraday equation, the molar flow rate of steam consumed at fuel electrode,  $\dot{n}_E$  (kmol/s), being:

$$\dot{n}_E = \frac{I}{2F} \tag{3}$$

#### 2.3.3 Cell stack voltage and power

Cell stack voltage depends on the operating conditions, i.e. gas compositions, temperature, pressure and current density. It is equal to Nernst voltage (the open circuit voltage, V) plus the overpotentials due to ohmic, activation (or polarization) and concentration losses.

$$V = V_{Nernst} + \Delta V_{ohm} + \Delta V_{act} + \Delta V_{conc}$$
(4)

Nernst voltage is calculated from the standard Gibbs free energy variation of the global electrochemical reaction at cell stack temperature and the partial pressures of the species involved

in the global electrochemical reaction raised at the power of the respective stoichiometric coefficients:

$$V_{Nernst} = \frac{\Delta G_{S}^{0}}{2F} + \frac{RT_{CS}}{2F} ln \left( \frac{p_{H_{2},fe} \, p_{CO_{2},oe} \, p_{O_{2},oe}^{0.5}}{p_{H_{2}O,fe} \, p_{CO_{2},fe}} \right)$$
(5)

The three overpotentials are calculated with empirical formulae from (Pérez-Trujillo et al., 2020). In particular, the concentration overpotential depends on how close current density is to the limiting current density that would make the concentration of one reactant fall to zero. This means that a limiting current density for each reactant at both electrodes has to be calculated using Fick's law (more details in (Pérez-Trujillo et al., 2020)).

Stack power can then be calculated as:

$$Power = V \cdot I \tag{6}$$

#### Molar balance

The following molar balances can be written at the two electrode outlets:

$$fuel electrode \begin{cases} \dot{n}_{CH_{4},out} = \dot{n}_{CH_{4},in} - \dot{n}_{R} \\ \dot{n}_{H_{2},out} = \dot{n}_{H_{2},in} + 3 \dot{n}_{R} + \dot{n}_{S} + \dot{n}_{E} \\ \dot{n}_{H_{2},out} = \dot{n}_{H_{2},in} - \dot{n}_{R} - \dot{n}_{S} - \dot{n}_{E} \\ \dot{n}_{CO,out} = \dot{n}_{CO,in} + \dot{n}_{R} - \dot{n}_{S} \\ \dot{n}_{CO_{2},out} = \dot{n}_{CO_{2},in} + \dot{n}_{S} - \dot{n}_{E} \\ \dot{n}_{N_{2},out} = \dot{n}_{N_{2},in} \end{cases}$$
(7)  
oxygen electrode 
$$\begin{cases} \dot{n}_{CO_{2},out} = \dot{n}_{E} \\ \dot{n}_{O_{2},out} = 0.5 \dot{n}_{E} \end{cases}$$
(8)

Following the equilibria equations 1 and 2, the molar flow rate of reformed methane is denoted with  $\dot{n}_R$  (kmol/s), and the molar flow rate of carbon monoxide consumed in the water gas shift reaction is denoted with  $\dot{n}_S$  (kmol/s).

#### Energy balance

The overall energy balance for the MCEC stack, which is the key to determine cell stack temperature can be expressed with the enthalpy flows associated with gas streams entering and exiting the stack:

$$Power + \dot{H}_{fe,in}(T_{in}) = \dot{H}_{fe,out}(T_{cs}) + \dot{H}_{oe,out}(T_{cs})$$
(9)

#### 2.4 GASIFICATION

There are several gasification technologies that can utilize sawmill byproducts. Given the nature of the feedstock, technology knowledge and specific site constraints, we opted to explore the following gasification configurations:

- The WoodRoll process
- Indirectly heated dual fluidized bed

- Directly heated fluidized bed

All the gasification processes are scaled to produce 20 MW of product gas, LHV basis. A brief description of these technologies is presented in the following text.

### 2.4.1 WoodRoll Process

The WoodRoll (WR) technology, developed by Cortus Energy AB, converts biomass to a clean renewable gas mainly composed of CO and H<sub>2</sub>. The WR process produces relatively high-quality syngas with moderate need for gas cleaning, compared to fluidized bed configurations, before its utilization to produce liquid fuels for transportation and industrial purposes. The WR process includes multiple processe stages, the major steps being feedstock drying, pyrolysis and gasification. The pyrolysis process generates char and gas. The char is gasified in an indirectly heated entrained flow gasifier, using steam in a subsequent step. The pyrolysis gas is combusted to sequentially supply the heat required in the gasification reactor (1100°C), the pyrolysis process (400°C) and the drier (100°C), as illustrated in Figure 5.



Figure 5. WoodRoll process. Source: Cortus Energy AB.

#### 2.4.2 Indirectly heated dual fluidized bed

Dual Fluidized Bed (DFB), the most widely used configuration for indirect gasification of biomass, essentially employs two fluidized bed reactors (one for gasification and one for combustion) linked by the bed material, circulating between them, Figure 6a. Different fluidization media can be used in the two reactors. A widely accepted configuration is to use steam and air in the gasification and the combustion reactor (Motta et al., 2018a), respectively. The gasification reactor is often a BFB and the combustor a CFB, as CFB boilers can utilize a wide range of feedstocks both in terms of moisture content and size. In a DFB configuration biomass is supplied to the gasification reactor and part of the produced char is combusted in the combustor to preheat the bed material prior to its re-entry to the gasifier. The capacity of the combustor can be scaled separately to boost steam generation by directly supplying additional biomass.

#### 2.4.3 Directly heated bubbling fluidized bed

An oxygen-blown fluidized bed gasification technology was considered as relevant given the fact that the MCEC produce an oxygen rich stream as byproduct, Figure 6b. To this end, the bubbling fluidized bed (BFB) configuration developed by Andritz Carbona is an interesting option to consider because it is a well-established technique with a commercial installation in Skive, Denmark (Motta et al., 2018b; Salo & Horvath, 2009) and a demonstration plant at GTI in Chicago, USA (Motta et al., 2018b).



(a) Dual fluidized bed (DFB) (b) Bubbling fluidized bed (BFB)



## 2.5 FISCHER TROPSCH (FT)

The FT process converts syngas to liquid fuels that can be readily upgraded to jet fuel. FT synthesis was established in early 20<sup>th</sup> century by Franz Fischer and Hans Tropsch for converting coal to liquid hydrocarbons. FT process is proven technology for converting synthetic gas to liquid crude and can help unlock the potential of forest feedstock to replace fossil use in the transport sector. Most published studies on production of FT liquid transportation fuels consider conventional integration of gasification and FT technologies (Ail & Dasappa, 2016; Baliban et al., 2013; Kim et al., 2013; Niziolek et al., 2014; Trippe et al., 2013). In addition, there are a couple of studies that consider integration between electrolysis and FT process (Chen et al., 2017; X. Li et al., 2016; Stempien et al., 2015). A handful of studies consider integration of either electrolysis or co-electrolysis with gasification and FT (Hillestad et al., 2018; M. Samavati et al., 2018; Mahrokh Samavati et al., 2017, 2018). Most of the work in this area (integration of high temperature co-electrolysis, gasification, and FT technology and refinery) has recently been performed at KTH (Mahrokh. Samavati, 2018).

The FTS is essentially an exothermic process operated at temperature in the range of  $200-350^{\circ}$ C and pressure in the range of 10-65 bar (Guettel et al., 2008). The major chemical reactions in the FTS are summarized in Table 2. The formation of the desired products, alkanes and alkenes, proceed according to the exothermic reactions (R6) and (R7) over metal catalysts. The water gas shift (WGS) (R8) also takes place over most metal catalysts balancing the CO and H<sub>2</sub> ratio. Side

reactions such as those producing alcohols (R9) and carbon deposits (R10) and (R12) may occur in the FTS reactor. Oxidation and/or reduction of the catalysts may also occur (R11). The conversion process is often catalyzed by metals such as cobalt, iron and ruthenium. In this project the FT process was operated at 240°C and 20 bar. The operational temperature is controlled with a continuous supply of cooling water.

Major re	eactions		
	Paraffins (alkane)	$(2n+1)H_2 + nCO \rightarrow C_nH_{2n+2} + nH_2O$	(R6)
	Olefins (alkene)	$2nH_2 + nCO \rightarrow C_nH_{2n} + nH_2O$	(R7)
	WGS reaction	$H_2O + CO \leftrightarrow CO_2 + H_2$	(R8)
Side reactions			
	Alcohols	$2nH_2 + nCO \rightarrow C_nH_{2n+2}O + (n-1)H_2O$	(R9)
	Boudouard reaction	$2CO \rightarrow C + CO_2$	(R10)
	Catalyst (Metals) oxidation/reduction	$M_xO_y + yH_2 \leftrightarrow yH_2O + xM$	(R11)
		$M_xO_y + yCO \leftrightarrow yCO_2 + xM$	
	Bulk carbide formation	$yC + xM \leftrightarrow M_xC_y$	(R12)

Table 2. Major overall reactions in the FTS reactor (Van Der Laan & Beenackers, 1999).

## 2.6 GAS CONDITIONING

For optimal conversion, the syngas composition prior to the FT reactor must satisfy the following criteria:

- $H_2$  to CO molar ratio 2
- inert components such as nitrogen and H<sub>2</sub>S should be below the tolerance limit specified by technology developers

As presented in Table 3, the syngas after primary cleaning contain  $H_2$  to CO ratios 2.04, 1.71 and 0.72 for the WR, BFB and DFB, respectively. At this stage, depending on the gasification technology, the syngas also contains tar, hydrocarbons such as methane,  $C_2H_x$  and benzene that reduce the overall carbon conversion from biomass to finished products when left unutilized. It would be wise to reform the methane and light hydrocarbons to CO and  $H_2$  components.

	WR	BFB	DFB
Reference	(Amovic et al., 2014)	(Worley & Yale, 2012)	(Alamia et al., 2017)
Feedstock	woodchips, sawdust, woodpellets	woodchips, woodpellets, forest residue, bark	woodchips, woodpellets, residue, bark
Pressure	atmospheric	atmospheric or pressurized	atmospheric
Temperature (°C)	1100	800–1000	800–900
Gasifying medium	steam	steam and oxygen	steam
Cold gas efficiency	75	65–70	70–75
Scalable	moderate	Yes	moderate
Feedstock flexible	Yes	Yes	Yes
Product gas composit	ion		
Gasification agent	steam	oxygen	steam
% vol. dry			
H <sub>2</sub>	57.0	23.9	42.1
со	28.0	33.1	24.6
CO <sub>2</sub>	12.0	26.8	18.3
CH <sub>4</sub>	3.0	9.2	6.8
$C_2H_4$	0.0	3.4	2.0
C <sub>2</sub> H <sub>6</sub>	0.0	0.0	0.2
N <sub>2</sub>	0.0	2.0	4.0
H <sub>2</sub> /CO	2.04	0.72	1.71
Tar (g/Nm³)	traces		20.5
BTX (g/Nm <sup>3</sup> )	0.7		7.8
Carbon conversion	very high	moderate	very high
Heat supply	Indirect	Direct	Indirect
ASU	No	Yes	No
Gas cleaning	low	moderate	high

#### Table 3. Gasification technologies.

There are several gas reforming technologies commercially available, such as steam reforming (SMR) and partial oxidation (POX). The SMR can be further classified depending on the source of the thermal energy required to drive the process, e.g. by partially combusting the incoming gas in an integrated furnace (Baltrusaitis & Luyben, 2015) or by using electrically (resistance) heated reactor surfaces (Wismann et al., 2019). Compared to the conventional side fired SMR, the resistance heated is expected to achieve remarkable reduction in reformer volume as the furnace and its accessories become unnecessary (Wismann et al., 2019). The resistance heated reformer is yet to be proven at commercial scale.

It was not straight forward which technology and process configuration would lead to the best yield and economic performance. A total of six cases were evaluated, two process configurations for each gasification technology. The configurations differ mainly on the downstream handling of light hydrocarbons ( $C_1$ – $C_6$ ) and tar in the raw gas. In summary, the following configurations were investigated:

- side-fired steam reformer (SMR)
- electric heated steam reformer (eSMR)

#### 2.6.1 Side-fired steam reformer (SMR)

In the SMR configuration, light hydrocarbons ( $C_1$ – $C_6$ ) in the raw gas, FT tail gas and off-gases from upgrading were reformed to CO and H<sub>2</sub> using side-fired SMR unit. In this configuration part of the FT, tail gas must be combusted to supply the heat required to drive the SMR and the remainder of the FT tail gas was mixed with the fresh raw gas entering the SMR.

Distinctive features are:

- Cooling of cathode stream generate saturated steam at 35 bar
- Cooling of anode stream produce saturated steam at 5 bar for internal use in the MCEC and the steam reformer
- SMR combustor exhaust gases sent to the integrated CHP system to supplement production of high-pressure steam
- Process steam deficit is assumed to be supplied from the CHP system

#### 2.6.2 Electric heated steam reformer (eSMR)

In the eSMR configuration, light hydrocarbons in the raw gas, FT tail gas and off gases from upgrading were reformed to CO and  $H_2$  using electric resistance heated reactor surfaces. A small fraction of the FT tail gas must be purged to limit accumulation of inert compounds in the FT reactor feed.

Distinctive features are:

- Cooling of cathode stream generate saturated steam at 35 bar
- Cooling of anode stream produce saturated steam at 5 bar for internal use in the MCEC and the steam reformer
- Purge adjusted to limit accumulation of inert components in the stream entering the FT reactor
- Purge stream used as fuel in the CHP system
- Process steam deficit is assumed to be supplied from the CHP system

## 2.7 IMPROVED CRYOGENIC SEPARATION PROCESS

The molar composition of the stream exiting the anode side of the MCEC is two-third  $CO_2$  and one-third  $O_2$ . With such high concentration of  $CO_2$ , the anodic stream is suitable for linking to carbon capture techniques. The benefit is twofold; to reduce the carbon footprint of the overall process and to derive income from the separated pure  $O_2$  stream. An improved cryogenic separation process (Xu et al., 2014) with multiple refrigeration and condensation steps was developed and applied to the anodic stream achieving molar purity of 99.9%  $CO_2$  in a final distillation step. Depending on the process configuration and gasification process, 20–50% of the separated  $CO_2$  is recycled back to the MCEC feed and the rest is assumed to be sequestered.

### 2.8 COMPRESSOR

The gasification technologies considered, the associated primary gas cleaning units and the MCEC are operated at atmospheric pressure, whereas the FT reactor benefit from high operational pressure both in terms of favourable reaction kinetics and reduced equipment size. Thus, the syngas after the MCEC is compressed to 22 bar using two stage compressor with an inter- and after-cooler.

### 2.9 ACID GAS REMOVAL (AGR)

Depending on the gasification technology the syngas initially contains 12-27% vol.  $CO_2$ . Though the MCEC utilizes  $CO_2$  to form the carbonate ion conductor through the electrolyte, part of the incoming  $CO_2$  inevitably follows the cathodic stream that proceed to the FT island.  $CO_2$  is not needed in the FT synthesis and must be removed to minimize reactor size and thereby cost. Besides, poisonous gases that are harmful to the FT catalysts such as H<sub>2</sub>S and ammonia may also be present in the syngas. Therefore,  $CO_2$  and other impurities must be kept below limits specified by FT technology developers. Amine-wash technology was implemented to ensure these impurities remain below the specs. Zinc guard bed ensures sulfur content is limited to a few ppm H<sub>2</sub>S.

## 3 METHODOLOGY

## 3.1 PROCESS MODELLING

Literature data was used for the gasification technologies evaluated. The conversion of synthesis gas to FT products was carried out based on the model developed in this work. The MCEC model was built in MATLAB Simulink environment based on experimental polarization data (Hu et al., 2014, 2016b). The MCEC model was soft linked to the rest of the process steps which were developed in UniSim Design. Yield and energy performance of the FT process was derived from published experimental data (Hanaoka et al., 2015; Snehesh et al., 2017) and implemented in UniSim as a yield reactor.

## 3.2 PROCESS INTEGRATION

The MCEC unit was integrated at a sawmill site. At a typical Nordic sawmill, significant part of the incoming timber ends up in byproduct category in the form of bark, woodchips and sawdust. Under business as usual operation, about 10.3% wt. dry basis of the byproducts is combusted onsite to preheat drying air that circulates through a drying kiln where the fresh sawn wood (lumber) is stored (Anderson & Toffolo, 2013). The rest of the byproducts are exported to other process industries such as pulp mills and wood pellet plants.

In the current framework, the byproducts are utilized to produce SAF where the heat demand of the drying kiln is satisfied using heat derived from the SAF plant. In case the SAF plant does not produce enough heat to satisfy the drying kiln, part of the byproducts will be combusted in the integrated boiler of the steam system. The gasification technologies considered in this study were scaled to produce 20 MW syngas (LHV basis). The feedstock required to achieve the target syngas energy was used to indicate sawmill size and derive the corresponding drying kiln thermal energy load. The sawmill was sized to match feedstock requirements of the gasification unit and the biomass boiler. The boiler was run to satisfy any heat deficit in the integrated system and to superheat saturated steam derived from the SAF facility at medium and intermediate pressure levels.

There are high temperature processes in the SAF island, e.g. SMR exhaust, conditioned syngas cooling, and low temperature heat demand in the sawmill, e.g. preheating of drying air, local room heating. To exploit the thermal energy of high temperature process streams and minimize exergy destruction, a common steam system can be suitably designed taking advantage of the temperature difference between the heat sources and sinks. Besides, steam is required in the SAF facility to drive the SMR and MCEC. Integrated design approach was implemented as illustrated in Figure 7. Pinch Analysis (Kemp, 2007) was applied to ensure heat transfer feasibility among the hot and cold process thermal streams thereby minimizing external utility requirements.



Figure 7. Integrated SAF production at sawmill facility – system boundary.

## 3.3 ECONOMIC ASSUMPTIONS

Production cost of total FT liquid and aviation fuel were evaluated as an economic indicator of the cases evaluated.

The total fixed capital investment (TFCI) of the process configurations were evaluated by accounting the equipment cost of major process units and applying factors to account for direct cost (equipment erection, piping, instrumentation and control, electricals, utilities, off-site, buildings) and indirect cost (design, engineering and construction, contingency) (Smith, 2005). Investment cost of most of the major process units are based on published data and the sources are outlined in Appendix B. All investment costs are scaled to fit equipment sizes required in this work and were adjusted for inflation to the reference year 2017. Monetary value conversion factors 0.1 and 0.85 are used for SEK to Euro and USD to Euro, respectively. Annuity method is applied to annualize the TFCI assuming 8% interest rate and 20 years economic lifetime, resulting in a capital recovery factor of 0.1. Annual O&M was fixed at 3% of TFCI (includes costs for operating supplies, planned and unplanned maintenance and repair, spare parts and payroll). The prices of energy carriers (feedstock, electricity, and co-products) and other materials (oxygen, scrubbing media) that constitute to the variable operating cost are presented in Table 4. A plant availability of 7884 hours (corresponding to 90% availability) was assumed.

	Unit	Value	Remark			
Feedstock*						
Forest residue	SEK/MWh	186				
Bark	SEK/MWh	159				
Sawdust	SEK/MWh	186				
Utility						
Electricity	SEK/MWh	400				
Oxygen	SEK/Mt	600				
Biodiesel	SEK/MWh	1200				
Scrubber oil	SEK/MWh	1060	DFB configuration			
*Source: Swedish Energy Agency; Statistics database: Trädbränsle och torvpriser (wood fuels and						

#### Table 4. Economic parameters.

peat prices); annual prices; Matrix: EN0307\_1; 2018.

#### 3.4 GHG PERFORMANCE EVALUATION

GHG performance of the cases were evaluted using a simplified approach based on the revised Renewable Energy Directives (RED II) guidelines. Accordinlyy, the allocations are made based on energy of products which stops where the product streams are distributed. In this study, the main product is FTL which fractionates into SAF and other hydrocarbons (diesel and naphtha ranges) components in the final stage. Emissions deriving from procurement of timber, processing of timber to lumber (electricity use) and export of lumber are not impacted by what the mill owners decide to do with their byproducts, hence are excluded from the GHG performance evaluation. The integrated processes produce two CO<sub>2</sub> concentrated streams, MCEC anode and AGR, which are suitable for carbon capture and storage (CCS). Thus, two GHG performance indicators are evaluated, with and without a CCS option. The following emission factors are assumed: Swedish electricity mix 13.1 gCO<sub>2</sub>eq/MJ (Drivmedel 2019- Redovisning Av Rapporterade Uppgifter Enligt Drivmedelslagen, Hållbarhetslagen Och Reduktionsplikten, 2020), transport of FTL 1.55 kgCO<sub>2</sub>eq/MWh, and transport of CO<sub>2</sub> by truck (0.108 kgCO<sub>2</sub>eq/ton-km) and ship (0.03 kgCO<sub>2</sub>eq/ton-km) (Innovation Fund (InnovFund) Methodology for GHG Emission Avoidance Calculation, 2021) for CCS cases.

## 4 RESULTS

### 4.1 MCEC PERFORMANCE

The performance of the MCEC is found to be highly linked to the incoming syngas composition and the downstream upgrading technique. The electrochemical unit consumes electrical energy and steam to boost the H<sub>2</sub> content of the incoming syngas to meet a target H<sub>2</sub> to CO ratio that is optimal for the downstream upgrading technique. In general, the MCEC's contribution is magnified when supplied with particle and tar free product gas lean in hydrogen content. In molten carbonate units  $CO_2$  must be present in the cell to transfer  $O_2$  in the form of  $CO_3^{2-}$  ion through the electrolyte.

The cathode exit of the MCEC is a syngas stream primarily composed of  $H_2$  and CO readily upgradable to liquid hydrocarbons in an FT unit, combined represent more than 88% vol. dry basis with  $H_2$  to CO molar ratio 2:1. Depending on the composition of the syngas feed to the MCEC, methane,  $CO_2$  and  $N_2$  may also be present in the cathode exit stream. The anode exit is a mixture of  $CO_2$  and  $O_2$  in the ratio 2:1, respectively. This stream undergoes a cryogenic separation process and part of the separated  $CO_2$  is recycled back to the feed of the MCEC. The amount of  $CO_2$  that can be recycled is directly proportional to the MCEC activity.

The molar ratio of  $H_2$  to CO in the syngas, derived from the WR and DFB gasification processes, are not far from 2, which is the optimal ratio for FT upgrading technique, hence the MCEC exhibited limited activity for these cases. The syngas from the BFB has however a very lean hydrogen content, which required increased activity of the MCEC to meet optimal syngas quality for the FT process. Through the application of the MCEC, the energy content of syngas magnified with factors 1.26, 1.30 and 1.4 for the WR, DFB and BFB cases, repectively. It should be noted that these figures correspond to the operating parameters of the MCEC, amount of steam feed and amount of CO<sub>2</sub> recycle chosen for each configuration. The methane content in the conditioned syngas marginally contributes to its total energy content, about 6 % on LHV basis.

To explore the most influential parameters of the MCEC and their pros and cons to its performance for syngas conditioning, the developed model was run under different operating conditions, such as current density, steam feed and  $CO_2$  recycle. The results presented in Figure 8 through Figure 12 refer to the BFB scenario as the case where the MCEC exhibited highest degree of activity. The WR and DFB scenarios also showed the same trend but to a lesser magnitude of activity due to their near optimal syngas composition for FT upgrading. Figure 8 presents polarization curve of the MCEC under a range of current densities and  $CO_2$  recycle for the BFB scenario. The MCEC unit was operated under atmospheric pressure and the corresponding temperature was estimated in the range 710–760°C, Figure 9.



#### Figure 8. MCEC polarization curve.

It is evident from equation 6 that the power required to drive the MCEC increases with current density, which in turn raises the operational temperature of the stack, Figure 9. Moreover, raising the  $CO_2$  composition of the feed stream magnify the power consumption whereas the temperature range fairly remains the same.



#### Figure 9. MCEC operational temperature.

Another aspect investigated was the capability of the MCEC to reform light hydrocarbons, such as methane, ethane and ethylene, and the results are presented in Figure 10Figure 10. The results turned out unfavorable, the amount of methane in the cathode exit stream magnifies with increasing CO<sub>2</sub> in feed. It seems equilibrium favors formation of methane at high CO<sub>2</sub> concentration. Presumably formation of CO proceeds via water gas shift followed by methane formation through reverse steam-methane reforming (Perez-Trujillo et al., 2018). Looking at Figure 10, it becomes clear that feeding the MCEC syngas that contains light hydrocarbons is counterproductive. It would



be beneficial to reform these species into  $H_2$  and CO components prior to MCEC and operate the MCEC at higher temperature to boost the yield conditioned syngas for FT upgrading.

Figure 10. MCEC cathode exit methane content.





It is not straight forward which operating conditions are optimal for the MCEC. High current density lead to high stack temperature (Figure 9), less methane in cathode exit (Figure 10 and Figure 11) and high power consumption (Figure 12). Energy indicator was introduced to measure performance of the MCEC in boosting the syngas energy, Figure 12. The yield of conditioned syngas magnifies with increasing recycled  $CO_2$  at the expense of increased power consumption under all the current densities evaluated.



#### Figure 12. Energy performance of the MCEC.

When production of biofuels is the priority, the MCEC should be operated at high current density under high  $CO_2$  recycling to maximize the yield of conditioned syngas and disfavor the formation of methane as a result of the associated high stack temperature. However, this configuration does not necessarily lead to the best energy efficiency, especially when the electricity that drive the MCEC is included in the equation. Both aspects are evaluated, one that maximize the yield of conditioned syngas and another that lead to optimal energy efficiency.

### 4.2 OVERALL PROCESS PERFORMANCE

#### 4.2.1 Mass and energy balance

Table 5 presents a summary of mass and energy flows for total FT liquids and SAF production of relevance for the techno-economic and greenhouse gas emission assessment. The investigated cases resulted in annual total hydrocarbon production of 9–12 kT, depending on the process configuration and gasification technique. Mass and energy balances of major streams can be found in Appendix C. The conversion efficiency (LHV basis) from biomass to renewable hydrocarbon components was estimated to 49–58 % and 57-61 % for SMR and eSMR configurations, respectively. It should be noted that the conversion efficiencies refer to the additional biomass, i.e. the difference between the internal consumption for a standalone operation of the sawmill and the integrated cases. On energy basis, the eSMR configurations improved the yield by about 9–13 % compared to their SMR counterparts.

Product selectivity, which leads to the yield of SAF, is strongly dependent on the FTS process configuration and catalyst. Upgrading of FT crude yields different fractions of hydrocarbon products, naphtha-, aviation-, and diesel-ranges. The results presented in Table 5 assume a 60 % wt. conversion FTP to SAF and the rest are grouped as other hydrocarbons (OHC).

		W	/R	D	FB			BFB		
						best yield (BFBy)		best efficier	best efficiency (BFBe)	
		SMR	eSMR	SMR	eSMR	SMR	eSMR	SMR	eSMR	
Input										
Additional biomass	GWh/y	211.14	206.45	224.23	210.83	223.21	235.83	234.92	237.10	
Electricity net	GWh/y	7.48	29.15	25.26	51.34	73.68	70.59	41.76	60.09	
Scrubber oil	GWh/y			7.07	7.07					
Side-products										
Oxygen	kTPY	2.54	2.55	5.96	5.74	17.68	8.18	6.04	6.04	
Tar oil	GWh/y			15.91	15.91					
Main products										
Total FTP	kTPY	8.81	9.91	8.85	9.67	10.44	11.49	9.82	11.10	
	GWh/y	110.08	123.88	110.57	120.92	130.48	143.59	122.79	138.76	
SAF	kTPY	5.28	5.95	5.31	5.80	6.26	6.89	5.89	6.66	
	GWh/y	66.05	74.33	66.34	72.55	78.29	86.15	73.68	83.26	
ОНС	kTPY	3.52	3.96	3.54	3.87	4.18	4.59	3.93	4.44	
	GWh/y	44.03	49.55	44.23	48.37	52.19	57.43	49.12	55.50	
Conversion efficiency	% (LHV)	52.14	60.00	49.31	57.35	58.46	60.89	52.27	58.52	

#### Table 5 Summary of inputs and biofuel production

The integrated process design approach showed that a significant part of the lumber drying process can be covered using steam generated from the high temperature process streams of the SAF plant. This is evident from Figure 13 (generation) where the production of high-pressure steam (HPS) from the biomass boiler is minor, about 1–25% of the total steam generation. Saturated steam at 35 bar (IPS) and 12 bar (MPS) are derived from cooling of conditioned syngas exiting the MCEC fuel electrode (cathode) and FT reactor, respectively. Both streams are superheated in the biomass boiler to allow mixing with expanding HPS at their respective pressure levels, thereby maximizing electricity production of the integrated process. The SAF facility both generates and consumes saturated steam at 5 bar (LPS). LPS is produced from cooling of the stream exiting the MCEC anode and consumed by the SMR and MCEC. The SAF facility has net deficit in LPS. Therefore, the LPS exiting the back-pressure turbine of the steam system is split between SAF plant and the sawmill drying process, Figure 13 consumption. The sawmill consumes LP steam (SMLPS) to preheat the drying air entering the drying kiln.



Figure 13. Integrated configurations steam balance.

### 4.3 ECONOMIC PERFORMANCE

Estimated specific capital investment per unit MW of FTP, and categorized by process section, are shown in Figure 14. For the WR cases, the gasification train (which includes a dryer and a pyrolysis reactor) is the largest contributor, about 30%, followed by the gas conditioning category (which includes the MCEC unit), about 25%. The order shifts for the DFB and BFB configurations in which the gas conditioning category becomes the largest contributor, 26% DFB and 30-35% BFB. For the BFB cases, this shift derives from the MCEC unit which resulted in capital expenditure about 1.5 and 2.5 times larger than that of the WR and DFB cases for BFBe and BFBy configurations, respectively. In general, the processes evaluated are predictably capital intensive due to the relatively small production scales, the technological maturity of the MCEC and limited integrability with the host facility.



#### Figure 14. Specific capital investment per MW FTP.

FTL production cost breakdowns for the configurations evaluated are shown in Figure 15. Accordingly, all cases resulted in production cost range 1 400–1 550 SEK/MWh-FTP. The eSMR

cases correspond to the lower end of the range, 1 397–1 460 SEK/MWh-FTP. Clearly, using electric resistance heated reformer has improved the yield which in turn reduced the production cost. On average, about 55 % of the production cost derives from the total fixed capital investment (TFCI), about 20 % feedstock procurement, and about 16% operation and maintenance.



#### Figure 15. FTL production cost breakdown.

To shed light on the SAF production potential, the FTL production costs were compared to the biojet fuel prices reported in the inquiry for Swedish biojet fuel blending mandate (Wetterstrand et al., 2019). The inquiry investigated SAF blending obligation in the Swedish aviation industry up to 2030 in which the contribution of biojet fuel was projected to increase from about 1 % vol. in 2021 to 30 % vol. in 2030. Biojet fuel prices 1 850 and 1 235 SEK/MWh<sup>2</sup> were estimated for 2021 and 2030, respectively. It should be noted that this comparison is only indicative since the assumptions behind the evaluations are not necessarily the same.

### 4.4 GHG PERFORMANCE

The GHG performance of the cases were evaluated with and without a CCS option and the results are shown in Figure 16. The SAF and hydrocarbons (diesel and naphtha range) produced will be used to replace fossil counter parts. Thus, the GHG performance of the cases were compared to a GHG performance of a fossil reference system evaluated by allocating emissions according to the energy share of SAF, petrol and diesel in the product using fossil emission factors 87 g CO<sub>2</sub>/MJ<sub>jetfuel</sub> (Tzanetis et al., 2017), 93 g CO<sub>2</sub>/MJ<sub>petrol</sub> and 95 g CO<sub>2</sub>/MJ<sub>diesel</sub> (Jafri et al., 2020). The CCS options consider 85 % efficiency on the CO<sub>2</sub> capture technology and the captured CO<sub>2</sub> is transported to long-term storage sites by truck and ship assuming 200 km and 1 200 km of land and maritime distances, respectively. GHG emissions saving potential of 87–97 % for the cases without CCS option and 120–150 % for those with CCS option can be realized. The carbon footprint of product distribution and CO<sub>2</sub> transport to the net GHG emissions are significantly smaller than that of

<sup>&</sup>lt;sup>2</sup> Recalculated assuming jet fuel calorific value of 35 MJ/liter

electricity. The cases were able to satisfy the 65 % GHG reduction for new plants set by the REDII, thanks to the low carbon footprint of the expanded system feedstocks, biomass and electricity.



Figure 16. GHG performance of the evaluated cases.

## 5 DISCUSSIONS

Given the fact that methane was present at the fuel electrode (cathode) under all scenarios, though at varying degree, a reformer is inevitable when the target product is other than methane. In this regard, the MCEC effectively replaces the water gas shift (WGS) process of the conventional gasification-based biofuels value chains evaluated. To indicate on the benefits of the MCEC, reference configurations were evaluated based on process flow diagrams that included a WGS and a SMR. These reference cases resulted in 7.86, 8.39 and 8.77 kTPY of FTP production for the WR, DFB and BFB cases, respectively. Compared to the MCEC results, Table 5, the increment in FTL productivity, depending on the syngas quality (gasification technology) and reformer configuration, were 5-19 % (SMR) and 15-31 % (eSMR). Thus, the benefit of the MCEC over WGS is twofold; the MCEC improves the yield by improving the carbon conversion efficiency, and creates a link for the direct utilization of external CO<sub>2</sub> in the context of power-to-X. It is noteworthy that the MCEC is the attractive option even without considering the later benefit.

It is evident that electrification of the SMR showed significant impact on the yield. This outcome sheds light on the importance of carbon tracking in the conversion processes, see Figure 17 (carbon balance of the BFB cases). The carbon balance of the DFB and WR cases are presented in the Appendix D. This study was limited to analyze the MCEC considering only the carbon content of the syngas, in the form of  $C_xH_y$ , CO and CO<sub>2</sub>. Consequently, the performance of the MCEC was constrained to meet the cathode exit syngas quality that was set as a target. Certain amount of the CO<sub>2</sub> exiting the oxygen electrode (anodic side) was recycled to the feed of the MCEC to increase the overall carbon efficiency. Under such configuration, the maximum attainable carbon conversion efficiency is set by the carbon content of the incoming syngas.

Depending on the gasification technology, about 63 % wt. WR, 68 % wt. DFB and 88 % wt. BFB of the carbon in biomass ends up in the syngas. Thus, to enhance the performance of the MCEC to increase carbon conversion, as much as possible of the carbon in biomass must be kept in the syngas. In this regard, finding alternative ways to supply the thermal heat required to drive the gasification process can bring the carbon conversion to syngas close to 100 %. These aspect of the MCEC performance are best clarified by the BFB cases, where the highest activity of the MCEC was observed, Figure 17. The MCEC adds flexibility to the process, for example, by adjusting the operating condition of the MCEC. The amount of carbon at the oxygen electrode can be controlled to reduce the load on the AGR.

It should be noted that the contribution of electricity to biofuels in the context of the configurations evaluated in this study was not limited only to the MCEC, but also to the reformer (eSMR scenarios) and the cryogenic separator that purifies  $CO_2$ . The oxygen electrode exhaust is rich in  $CO_2$  concentration (67 % vol.), the remainder being oxygen. This stream undergoes cryogenic separation to produce 99.9 % vol.  $CO_2$  and oxygen-rich fractions. A part of the  $CO_2$  separated (20-50 %) was recycled to the feed stream of the MCEC to facilitate the operation of the MCEC primarily serving as carbonate ion source, and part of it is captured.

![](_page_41_Figure_1.jpeg)

#### Figure 17. Carbon balance, normalized to carbon in syngas-BFB cases.

Electrochemical reduction of  $CO_2$  to CO and C in MCEC has been studied (Kaplan et al., 2010; Küngas, 2020; Ren et al., 2015). A recent experimental study of MCEC (Meskine et al., 2020) has confirmed that electrochemical reduction of  $CO_2$  to CO was observed at the fuel electrode in the presence of steam. Considering such reports, co-electrolysis (electrolysis of steam and  $CO_2$ ) operation mode of the MCEC is something that needs close investigation. This is particularly relevant due to its potential interplay with methane formation mechanism, assuming the most relevant gaseous products of  $CO_2$  electrolysis are CO and  $CH_4$  (Meskine et al., 2020).

One of the main drawbacks of carbonate cells is of course their low carbon conversion efficiency due to the continuous formation of  $CO_3^=$  at the cathode which gets protonated at the anode to release  $CO_2$ . For every reduction of  $H_2O$  to  $H_2$ , one  $CO_2$  is released at the anode. MCEC are often operated under excess  $CO_2$  supply to support high current densities required to maintain a steady stack temperature.

## 6 CONCLUSIONS

This study has established operating conditions of MCEC for syngas conditioning applications. Under low current density mode, the MCEC favors the formation of methane. Under high current density mode, the MCEC disfavors formation of methane but the power consumption is high. For all the cases evaluated, methane was present at the cathodic exit which may or may not be attractive depending on the final product. For FT applications, a methane reformer is inevitable to achieve acceptable yields. The activity of the MCEC varied when supplied with the different synthesis gases derived from the WR, DFB and BFB gasification techniques. The BFB technology generates syngas with the least hydrogen content and promoted highest activity in the MCEC. Both the DFB and WR technologies generate syngas quality that are not far from the optimal for FTS and, hence, the MCEC activity for these cases was low.

Electrification of the steam reforming (eSMR) improved the overall yield of FTP by 9–13 % compared to the SMR cases and by 15–31 % compared to the conventional configurations with WGS. This indicates that there is room for fine tuning gasification-based processes to increase carbon conversion efficiency.

Under the assumed economic conditions, biofuels can be produced at a cost of 1 400–1 550 SEK/MWh-FTP, the lower end of the range corresponding for eSMR configurations. Clearly, using electric resistance heated reformer has improved the yield by as much as 13 %, which in turn reduced the production cost.

## 7 FUTURE WORK

The capacity of the processes evaluated was limited to the syngas amount produced in the gasification step, which puts a limitation on the activity of the electrochemical unit. In this sense, scaling-up opportunity was not fully exploited. Future work should look into alternative biogenic  $CO_2$  sources that can be directly inserted into the MCEC, thereby increasing the product yield without additional biomass input.

Electrification of the steam methane reformer improved the overall carbon conversion of the cases evaluated. Future work should look into the possibility to use electricity to heat-up other process steps where carbon losses has been identified, such as gasification reactors of the WR and DFB cases.

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## APPENDIX A – PROCESS FLOW DIAGRAM

![](_page_49_Figure_2.jpeg)

Figure A-1. Process flow diagram – forest biomass to jet fuel.

## APPENDIX B – CAPITAL COST REFERENCE DATA

Process Unit	Scaling	Scaling parameter	TFCI Cost	Source
	Exponent		components	
Pretreatment				
Feedstock handling	0.77	t/h, wet biomass	a+d+e+f+g	(Thunman et al., 2019)
Belt dryer	0.65	MW <sub>th</sub> , LHV biomass	a+d+e+f+g	(H. Li et al., 2012)
Gasification island				
WR	0.65	MW <sub>th</sub> , LHV biomass	a+d+e+f+g	(Baldesca & Lasheras, 2012)
DFB	0.65	MW <sub>th</sub> , LHV biomass	a+d+e+f+g	(Thunman et al., 2019)
BFB	0.65	MW <sub>th</sub> , LHV biomass	a+d+e+f+g	(Worley & Yale, 2012)
Gas conditioning				
SMR	0.6	kmol/h (feed)	a+d+e+f+g	(Baltrusaitis & Luyben, 2015)
eSMR	0.6	kmol/h (feed)	c+d+e+f+g	assumed
Compressor	0.7	kmol/h (feed)	i+d+e+f+g	(Smith, 2005)
H <sub>2</sub> S scrubber	0.7	MW <sub>th</sub> , LHV syngas	a+d+e+f+g	(Thunman et al., 2019)
MCEC	1	m <sup>2</sup> (cell area)	j+d+e+f+g	(Calise et al., 2006)
Cryogenic separator	0.65	kg/s, CO <sub>2</sub> separated	a+d+e+f+g	(Xu et al., 2014)
Amine Wash	0.65	Nm <sup>3</sup> /h, shifted syngas	b+d+e+f+g	Commercial
Zinc Bed	0.65	MW <sub>th</sub> , HHV biomass	c+d+e+f+g	(Arvidsson et al., 2014)
Synthesis & upgrading				
Fischer-Tropsch reactor	0.75	Nm <sup>3</sup> /h, syngas feed	h+d+e+f+g	(Liu et al., 2011)
Initial catalyst fill	1	MW <sub>th</sub> , HHV FTP	h+d+e+f+g	(Liu et al., 2011)
Steam system	0.65	MW <sub>th</sub>	c+d+e+f+g	(Thunman et al., 2019)
Balance of Plant	-	-	g	(Liu et al., 2011)

<sup>a</sup> Reference equipment cost.

<sup>b</sup> Engineering, procurement & construction (EPC) costs.

<sup>c</sup> Assumed 50% of SMR cost. This is rather a conservative estimate considering substantial volume reduction obtained for eSMR reactor (Wismann et al., 2019).

<sup>d</sup> Other direct costs (6 % of total EPC or derived from commercial quotes for equipment evaluated based on commercial data).

<sup>e</sup> Indirect cost (3% of total direct cost or derived from commercial quotes for equipment evaluated based on commercial data).

<sup>f</sup>Contingency (10% of total direct cost).

<sup>g</sup> Balance of plant.

<sup>h</sup> FT equipment cost for a slurry type reactor with heat exchangers.

<sup>1</sup> Derived on a bottom-up capital cost estimation method outlined in literature (Smith, 2005), overall installation factor of 2.3 applied.

<sup>j</sup> Capital cost estimated using equations outlined in (Calise et al., 2006) for SOFC.

## APPENDIX C – MAJOR PROCESS STREAMS

![](_page_51_Figure_2.jpeg)

Figure C–1. Major streams WR cases.

![](_page_52_Figure_1.jpeg)

Figure C-2. Major streams DFB cases.

![](_page_53_Figure_1.jpeg)

Figure C–3. Major streams BFBy cases (best yield scenario).

![](_page_54_Figure_1.jpeg)

Figure C-4. Major streams for BFBe cases (best efficiency scenario).

## APPENDIX D – CARBON BALANCE

![](_page_55_Figure_2.jpeg)

WR\_SMR [C-balance]

WR\_eSMR [C-balance]

![](_page_55_Figure_5.jpeg)

![](_page_55_Figure_6.jpeg)

![](_page_56_Figure_1.jpeg)

DFB\_SMR [C-balance]

Figure D-2 Carbon balance DFB cases.

![](_page_57_Figure_1.jpeg)

Figure D-3. Carbon balance BFB cases (best yield scenario).

![](_page_58_Figure_1.jpeg)

BFBe\_SMR [C-balance]

Figure D-4. Carbon balance BFB cases (best efficiency scenario).

![](_page_59_Picture_1.jpeg)

![](_page_59_Picture_2.jpeg)

![](_page_59_Picture_3.jpeg)

![](_page_59_Picture_4.jpeg)

![](_page_59_Picture_5.jpeg)

![](_page_59_Picture_6.jpeg)

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