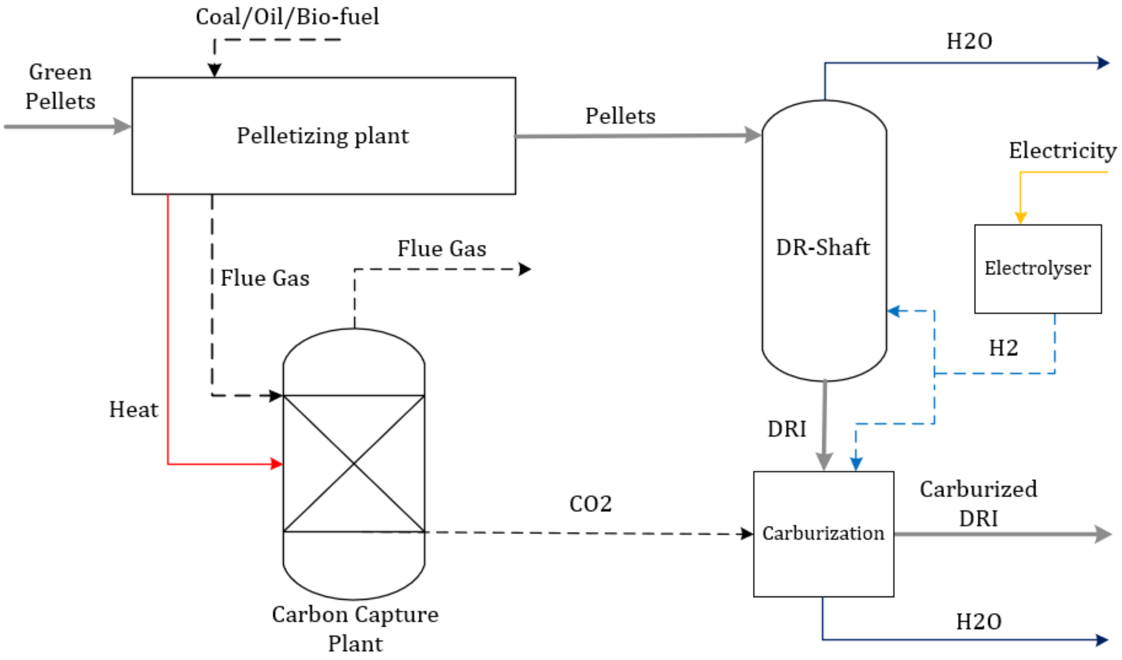

Carbon Capture and Utilization for Carburization of Hydrogen DRI

A techno-economical system analysis of possible synergies for future mine-to-iron plants at LKAB

Master's thesis in Sustainable Energy Systems

JAKOB ANDREASSON
CARL EMME

DEPARTMENT OF SPACE, EARTH and ENVIRONMENT
CHALMERS UNIVERSITY OF TECHNOLOGY
Gothenburg, Sweden 2021
www.chalmers.se

Master's thesis 2021

Carbon Capture and Utilization for Carburization of Hydrogen DRI

A techno-economical system analysis of possible synergies for future mine-to-iron plants at LKAB

JAKOB ANDREASSON
CARL EMME

Department of Space, Earth and Environment
Division of Energy Technology
Chalmers University of Technology
Gothenburg, Sweden 2021

Carbon Capture and Utilization for Carburization of Hydrogen DRI
A techno-economical system analysis of possible synergies for future mine-to-iron plants at LKAB
JAKOB ANDREASSON
CARL EMME

Supervisor: Jörgen Rosenqvist, LKAB
Examiner: Fredrik Normann, Division of Energy Technology

Master's Thesis 2021
Department of Space, Earth and Environment
Division of Energy Technology
Chalmers University of Technology
SE-412 96 Gothenburg
Telephone +46 31 772 1000

Cover: Visualization of a carbon capture and utilization system integrated with LKAB's future operation.

Typeset in LaTeX
Printed by Chalmers Reproservice
Gothenburg, Sweden 2021

Carbon Capture and Utilization for Carburization of Hydrogen DRI
A techno-economical system analysis of possible synergies for future mine-to-iron plants at LKAB
Master's thesis in Sustainable Energy Systems
JAKOB ANDREASSON & CARL EMME
Department of Space, Earth and Environment
Chalmers University of Technology

Abstract

This thesis investigates the technical and economic viability of capturing CO_2 from LKAB's pelletizing plants and utilizing the CO_2 as a carbon source for carburization of hydrogen reduced direct reduced iron (DRI). The CO_2 is captured through a post combustion carbon capture process utilizing an amine solvent. The thesis suggests two routes for CO_2 utilization where CO_2 is converted to CO or methane, which can carburize DRI, and additionally a CO_2 permanent storage route where DRI is instead carburized with bio-methane.

The work includes: mass and energy balances of each pelletizing plant to determine magnitudes of carbon and produced DRI, modelling of the carbon capture plant to validate technical viability of capturing from the low CO_2 concentrations found in LKAB's flue gases, and an economic assessment to compare the different routes.

The mass and energy balance shows that the CO_2 in the flue gases is sufficient to be of interest for the DRI carburization. For stand-alone application to LKAB's plants, i.e no integration between plants, the level of carburization possible using only CO_2 from pelletization are between 0.45-1.2wt% carbon content of the DRI. The CO_2 modelling concluded that it's feasible to capture CO_2 from LKAB's flue gases data at similar costs as for other industrial sites, due to the presence of available excess heat. The cost assessment showed that the CO-route has a similar cost to a reference case without CO_2 capture and where carburization occurs with biomethane. The methane and storage route were more expensive. A sensitivity analysis revealed that the cost results are highly sensitive to the price of electricity and solid biomass.

This thesis has shown technical and economical feasibility for LKAB to implement CO2 capture and/or utilization at scale together with the manufacturing of hydrogen DRI. Carburization of DRI with CO2 may be competitive to carburization with biomethane depending on the development of the energy system. If emissions from the pelletizing plant are reduced with other solutions, CO2 can be captured from other industries in the geographical proximity to LKAB's operations. The carburization of DRI has the potential to be a carbon sink if the CO2 comes from biomass combustion.

Keywords: Carbon capture, CCU, CCUS, DRI, Carburization, Pelletizing

Acknowledgements

It has been an exciting journey to both explore and develop fossil free ironmaking in this thesis, and being able to work on a wide scale from the small complex carburization mechanisms to the large systems studies. But this thesis would not have been possible without the support and guidance we received from several people. We are grateful for the insights and support we received from Jörgen Rosenqvist and Dr. T. K. Sandeep Kumar at LKAB who helped us put the work into context with the development occurring at LKAB with hydrogen reduction. We thank Elin Fahrman for the valuable support with the carbon capture modelling. And of course our examiner Fredrik Normann who provided us such an interesting project and supervising us. Fredrik, who works half time at LKAB, has been the perfect bridge between the Chalmers and LKAB.

Finally we would like to thank the staff at the Division for Energy Technology for inspiring conversations challenging our thoughts and giving us new ideas.

Gothenburg, June 2021
Jakob Andreasson
Carl Emme

Contents

List of Figures

List of Tables

List of Abbreviations

AEL	alkaline water electrolyser
CAPEX	capital expenses
CCS	carbon capture and storage
DCC	direct contact cooler
DRI	direct reduced iron
DR-shaft	direct reduction shaft
EAF	electric arc furnace
EIC	equipment installed costs
GHG	green house gas
HBI	hot briquetted iron
KPIs	key performance indicators
LKAB	Luossavaara-Kiirunavaara Aktiebolag
MEA	Monoethanolamine
OPEX	operational expenses
PH	preheating
RWGS	reverse water-gas shift
TIC	total installed cost

1

Introduction

Sweden has, in an international effort to limit global warming below 2°C, adopted a climate policy framework with a national goal of net zero carbon emissions by 2045 [1]. This requires carbon neutral transitions all throughout Swedish society and industrial sector. Luossavaara-Kiirunavaara Aktiebolag (LKAB) is a major actor in the iron mining and refining sector whose iron mines in northern Sweden represent 80% of the iron ore mined in Europe [2]. The global iron mining, iron making and steel value chain contributes about 7% of the world's total CO_2 emissions and LKAB's production of iron pellets accounts for 4% of Sweden's industrial emissions[3].

Steelmaking has since the 18th century relied on the coke-fired blast furnace process to that enabled the mass manufacture of steel but with the consequence of high green house gas (GHG) emissions. In 2016 the steel producer SSAB, the energy utility company Vattenfall and LKAB created the joint venture Hybrit Development which intends to produce emissions free steel by replacing coke coal with hydrogen and do the reduction in a direct reduction shaft (DR-shaft). In 2020 LKAB committed to exchanging its product portfolio from iron pellets to hydrogen direct reduced iron (DRI). The transition to DRI production makes the traditional blast furnace process obsolete for LKAB's customers. The transition is estimated to result in annual emissions reductions for LKAB's customers of 35 million tonnes CO_2, corresponding to 66% of Sweden's yearly CO_2 emissions. Since the hydrogen will be produced through electrolysis large amounts of electricity will be required, equivalent to one third of today's total Swedish production when fully developed. [3]

While LKAB has committed to ambitious goals there are still challenges and questions to be solved ahead. LKAB has internal process emissions mostly stemming from the pelletizing plants. Many pathways to reduce these emissions exists, such as electrification, biofuels or a carbon capture solution. At the same time carbon content in DRI is important for steel production and due to the new hydrogen reduction process the carbon content is missing. This opens up an opportunity for a solution that combines carbon capture and utilization of CO_2 to both reduce LKAB's emissions and provide a source of carbon for DRI carburization. LKAB stand before multiple process choices and since mining is a capital intensive industry with long lead times and investment horizons, process decisions made in the coming years will have effect and exist beyond 2050. Therefore all paths and process alternatives have to be evaluated.

1.1 Aim

The aim of this thesis is to determine the potential of post combustion carbon capture with Monoethanolamine (MEA) as a solvent at LKAB's pelletizing plants from a technical and economic perspective. Furthermore, the work evaluates CO_2 utilization for DRI carburization and compares it with a geological storage solution. Focus is on process design with moderate considerations to DRI metallurgy and chemistry. The thesis is put into context as if being included in LKAB's transition to produce hydrogen DRI, with the requirement of a carbon content in the DRI.

2

Theory

2.1 Pellets Production at LKAB

Iron mining and processing is a large and complex industry where products require several process steps before they can be transported to customers. LKAB currently produce and sell iron ore pellets, and will in the coming years expand the processing to instead produce DRI. In Figure 2.1 a simplified scheme of LKAB's existing and future operations are shown.

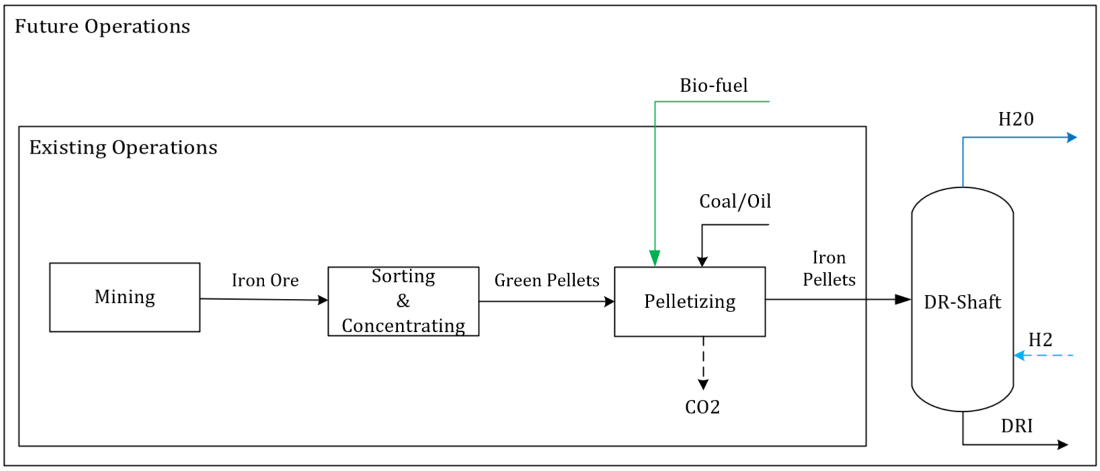

Figure 2.1: Schematic of LKAB's existing operations from mining to iron pellets, and future operations with the inclusion of a DR-shaft to instead produce DRI as a new product. Future plans also include switching to bio-fuels for the pelletizing plant.

LKAB's operations begin with ore being mined at approximately 1km depth by either drilling or blasting. The ore is then crushed before being transported to the sorting plant where the iron is further crushed and magnetically separated through several steps. As the iron feed still contains considerable amounts of impurities it goes to the concentration plant where grinding, further sorting and flotation is performed to separate impurities from the resulting slurry. Then so called green pellets are formed by rolling in large drums. The green pellets primarily consists of magnetite, with approximately 10% of the content being moisture and additives that have been added to give the pellets their desired properties. In the pelletizing plant the green pellets are thermally treated to improve strength and ability to withstand mechanical stresses, which is done by sintering the pellets at approximately 1200°C. As the pellets are dried and heated, the magnetite oxidizes to hematite in an exothermic reaction that supplies the main heat of the process according to Equation 2.1.[4]

$$4\,Fe_3O_4 + O_2 \rightarrow 6\,F_2O_3 \qquad (\Delta H_r = -122\ \text{kJ/mol}) \qquad (2.1)$$

Two types of pelletizing plant exist the grate-kiln and the straight-grate but their purpose is the same, to oxidise and strengthen the pellets. In theory the heat generated from oxidation is enough to fuel the process, but in reality a heat demand exists that has to be supplied at a temperature above 1200°C. To supply the heat the grate-kiln type use pulverized coal burner while the straight-grate type use oil burners resulting in CO_2 formation. The combustion occur at high air-fuel ratios as the main purpose of the pelletizing plant is to oxidise the pellets. Another source of carbon dioxide is the calcination of limestone additives (Eq. 2.2). The CO_2 from calcination is distributed on four stacks, per pelletizing plant, while the CO_2 from combustion is mainly localised to the preheating (PH) stack. The PH stack has a low but relevant CO_2 concentration and high flow rate at a high temperature. The other stacks have very low CO_2 concentration and also lower flow rates.

$$CaCO_3 \rightarrow CaO + CO_2 \qquad (\Delta H_r = 301\ \text{kJ/mol}) \qquad (2.2)$$

To achieve fossil free steelmaking LKAB are extending their processing to include a DR-shaft for hydrogen reduction of DRI. The direct reduction process is described in Chapter 2.2 and direct reduction with hydrogen in Chapter 2.2.2.

2.2 Direct Reduction of Iron Pellets

Direct reduction of iron pellets is a common steel production route in countries with low natural gas prices. The dominating process type of direct reduction use a counterflowing shaft where iron pellets are fed from the top and meet an upwards flowing reduction gas, a so called DR-shaft. Commonly the reduction gas is a syngas of primarily hydrogen (H_2) and carbon monoxide (CO) made from natural gas reforming [5] [6]. The reduction occurs by the following two reactions:

$$Fe_2O_3 + 3\,H_2 \rightarrow 2\,Fe + 3\,H_2O \qquad (\Delta H_r = 99.5\,\text{kJ/mol}) \qquad (2.3)$$

$$Fe_2O_3 + 3\,CO \rightarrow 2\,Fe + 3\,CO_2 \qquad (\Delta H_r = -24.8\,\text{kJ/mol}) \qquad (2.4)$$

The resulting DRI does not consist of pure iron, some iron oxide in the form of wüstite (FeO) remains, formed via the Reaction 2.5 and 2.6. The grade of metallisation (i.e. share of incoming hematite that is fully reduced) of DRI in DR-shaft process with high H_2 share is approximately 94-96% by mole [7].

$$Fe_2O_3 + H_2 \rightarrow 2\,FeO + H_2O \qquad (\Delta H_r = 18.6\ \text{kJ/mol}) \qquad (2.5)$$

$$Fe_2O_3 + CO \rightarrow 2\,FeO + CO_2 \qquad (\Delta H_r = -25.7\ \text{kJ/mol}) \qquad (2.6)$$

2.2.1 Carburization of DRI

In steelmaking it is important that the iron contains a certain carbon content, both for lowering melting temperature and giving the final steel product its desired properties. In industrial iron reduction processes, some carburization occurs simultaneously to the reduction. In the traditional blast furnace process iron is carburized with CO generated from coke through the Boudouard reaction (Eq. 2.7), creating a product with a high carbon content of 4-5 wt%. In the direct reduction processes, carburization stem from the Boudouard reaction but also from the hydrogen present (Eq. 2.8). Additionally, the carbon content can be boosted by injecting methane (Eq. 2.9) at the lower end at the shaft. DRI commonly has a carbon content ranging between 1-4.5wt%. [5][8]

$$3\,\mathrm{Fe} + 2\,\mathrm{CO} \rightarrow \mathrm{Fe_3C} + \mathrm{CO_2} \qquad (\Delta \mathrm{H}_r = -148.7\,\mathrm{kJ/mol}) \tag{2.7}$$
$$3\,\mathrm{Fe} + \mathrm{CO} + \mathrm{H_2} \rightarrow \mathrm{Fe_3C} + \mathrm{H_2O} \qquad (\Delta \mathrm{H}_r = -131.0\,\mathrm{kJ/mol}) \tag{2.8}$$
$$3\,\mathrm{Fe} + \mathrm{CH_4} \rightarrow \mathrm{Fe_3C} + 2\,\mathrm{H_2} \qquad (\Delta \mathrm{H}_r = 98.3\,\mathrm{kJ/mol}) \tag{2.9}$$

As the carburization with methane (Eq. 2.9) is endothermic the DRI is cooled. The cooling can be unwanted if the DR-shaft is integrated with an electric arc furnace (EAF) or a briquetting process to produce hot briquetted iron (HBI). If so, it is possible to inject a methane/CO mixture to increase the carbon content while keeping temperature stable which is a process concept presented in [5]. Briquetting DRI to HBI is typically done above 700°C with a carbon content in the DRI ranging between 0.5-1.6wt% [9] [10].

The carburization mechanisms behind 2.7 and 2.8 are further described in [8], where an experiment on carburization of iron foils at 920°C using different CO-H_2 mixtures was referenced. It concluded that CO carburization is always initiated with the decomposition of CO to C and adsorbed oxygen on the iron surface. The adsorbed oxygen then reacts with H_2 or CO. The reaction of adsorbed oxygen with hydrogen is considered very fast compared to removal by CO, which means that the presence of hydrogen can accelerate carburization. Additionally the study [11] investigated the carburization activity of CO, CO+H_2 and CH_4 on DRI at different temperatures. Main results from the study is that between 400°C and 900°C both CO and CH_4 are active as carburizing agents, however at higher temperatures CO becomes more stable and less active.

2.2.2 Hydrogen Direct Reduction

Using hydrogen as the sole reduction medium in the DR-shaft allows production of DRI without any GHG emissions, but comes with a number of challenges. As the reduction reaction (Eq. 2.3) is endothermic, heating is required in the process. This can be achieved by preheating pellets and the hydrogen gas with electricity [7]. The DR-shaft is operated at 800°C and this thesis assumes that the DRI leaves the shaft at this temperature [7]. Another challenge with hydrogen direct reduction is the lack of carbon content in the DRI. A carbon content in the DRI is important in steelmaking as during melting in the EAF the carbon content influences DRI melting temperature and impacts slag foaming which impacts energy consumption. Additionally DRI with no carbon content is very reactive and risks reoxidation during storage and transportation, however it can be pacified through briquetting where HBI is produced [9].

2.3 Carbon Capture

Carbon capture technologies allow for the generation of CO_2 from a industrial process that might have unavoidable emissions or emissions that are hard to reduce. For existing industrial processes post combustion techniques where the CO_2 is separated from the flue gas is a good alternative. In this thesis, post-combustion chemical absorption with MEA as solvent is the only technology considered, as this is a mature technology, commercially available and already implemented at industrial scale. In the study by Johnsson et al [12], carbon capture using MEA was investigated for industrial sites in Sweden. The study points out four factors for industrial sites to be good candidates for carbon capture plant integration; high volumes of CO_2, high concentrations of CO_2 in the flue gas, access to low cost excess heat and ocean access for lower transport costs. The study looks into 28 industrial processes together emitting more than 50% of Swedish total CO_2 emissions, each site produce at least 500ktCO_2/year and have flue gas concentrations above 5%vol. The study found that a range between 40 - 110 €/t CO_2 depending on emission source is expected.

2.3.1 Absorption based Carbon Capture with MEA

The chemical absorption process is based on the concept of temperature-swing absorption, schematically shown in Figure 2.2. Firstly the flue gas passes a direct contact cooler (DCC) to lower the temperature, partially remove water and reduce particulates in the flue gas. A liquid solvent (MEA) reacts with the CO_2 in the flue gas in a column, the absorber, at a temperature of $40 - 60°C$. The solvent absorbs the CO_2 and reaches a saturation rate, denoted rich loading. The CO_2 rich solvent is then passed to a separate column, the desorber, where the solvent is heated to $110 - 120°C$. The heat energy is supplied through steam. The high temperature releases the CO_2 from the solvent to the gas phase. The pure CO_2 gas can then be processed for storage or usage. After passing through desorber the solvent has a low concentration of CO_2, denoted lean loading, and is recirculated to the absorber. Capture rate is the amount of CO_2 removed from the flue gas, a normal target is 90% as higher capture rates in general requires significantly more heat energy in the reboiler of the desorber.[13]

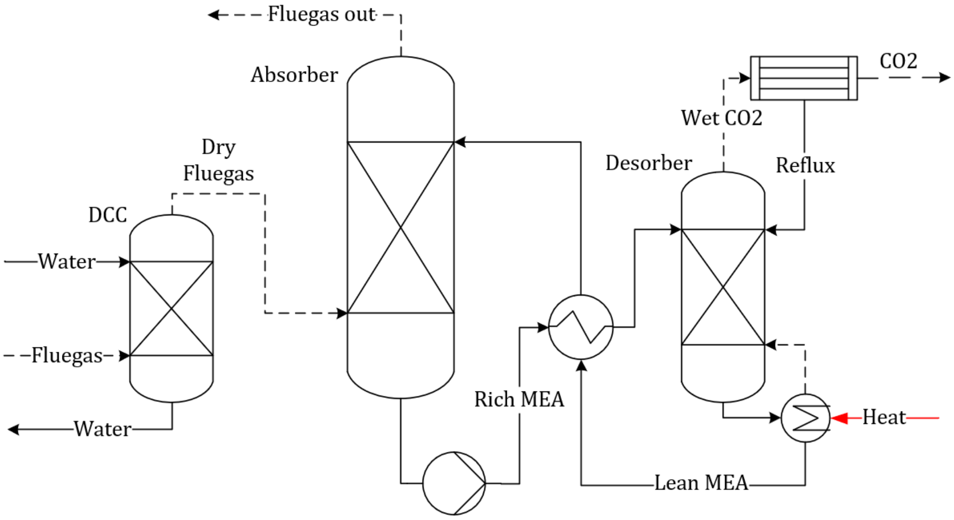

Figure 2.2: Schematic of a temperature swing based carbon capture plant using MEA as solvent.

MEA is a very efficient solvent and highly reactive with CO_2, resulting in high separation rates. A challenge for carbon capture with MEA is the high absorption enthalpy which causes the regeneration of CO_2 to be energy intensive. The energy consumed by the regeneration can therefore be a large contributor to the operational cost of the capture process. MEA has some constricting properties the first being that MEA is corrosive and may damage equipment, hence to reduce corrosiveness the solvent is diluted with water to a degree of 30 wt% MEA. The second property is that MEA is slowly degraded if oxygen is present above $100°C$ and quickly degraded by the presence of SO_x, often demanding flue-gas desulfurization. The degradation requires adding new MEA to the lean flow, the replacement increase OPEX of a plant. MEA that exits the absorber via the flue gas can form carcinogenic compounds in nature. To limit release of MEA to nature washing the flue gas with water is recommended. Degradation of the MEA and corrosive effects will not be handled in this thesis but a washer is included in capture plant to limit release of MEA in the flue gases.[13]

2.4 Carbon Utilization

Carbon utilization is a general term for when captured CO_2 is used in a product or process. This chapter will present two carbon utilization pathways that are relevant for this thesis.

2.4.1 CO Conversion

CO_2 can be converted to carbon monoxide in several ways, such as hot processing with coke through Boudouard reaction, or co-electrolysis in a solid oxide electrolysis. An additional way is through the reverse water-gas shift (RWGS) reaction (Eq. 2.10), which is an endothermic equilibrium reaction that favours CO formation as temperature rises. RWGS has an equilibrium of approximately 72% CO_2 conversion at 750°C and 1:1 H_2/CO_2 molar ratio [14].

$$\mathrm{CO_2 + H_2 \rightarrow CO + H_2O} \qquad (\Delta H_r = 41KJ/mol) \tag{2.10}$$

RWGS can be performed in the presence of a catalyst which improves activity of CO formation. Suitable catalytic materials are Pt, Pd, Cu, Ni and Fe. Constructing a RWGS reactor for CO production is a possible direction, however the high temperature required for good RWGS activity (>500°C) makes the alternative unattractive [15] [16]. Another interesting possibility is to use the hot DRI feed (800°C) exiting the DR-shaft as the heat supply. By injecting CO_2 and H_2 directly on the DRI, the gas temperature is raised and induces CO formation through RWGS. While studies on iron-oxide catalysts in CO production have been made [15] [16], only one study investigating the catalytic capabilities of DRI was found. The study which investigated DRI as a catalyst for methane reforming found that DRI can function as a catalyst for the investigated application, however with different gas compositions intense carburization could instead occur [17].

2.4.2 Methane Synthesis

Synthesising methane from CO_2 is a common process in industry where it is primarily used in purification of hydrogen in ammonia production . While many types of methanation reactors exists, the most established technology is fixed bed reactor with intercooling over a nickel catalyst. Methane is synthesised with the Sabatier reaction (Eq. 2.11), which has a two-step mechanism which combines RWGS and CO hydrogenation. The exothermic process is typically operated in a range of 200-500°C at an elevated pressure and provides heat available for recovery. A CO_2 conversion of 98% can be reached during operation at 20 bar, resulting in a methane content above 90%. [18] [19]

$$\mathrm{CO_2 + 4\,H_2 \rightarrow CH_4 + 2\,H_2O} \qquad (\Delta H_r = -165KJ/mol) \tag{2.11}$$

2.5 Carbon Storage

Carbon storage into geological rock formations has been conducted by the Norwegian oil and gas industry in the North Sea since 1996. This has proved the technology feasible but up until recently no concrete plans for large scale storage from land based sources has been realised in Europe. The most prominent solution for long term carbon dioxide storage is to liquefy the CO_2 which makes it easier to transport. The transport can be by truck, rail or ship with the final step being injection into underground geological formations.

2.5.1 Compression & Liquefaction

A liquefaction plant is needed for transportation of large volumes of CO_2. In the liquefaction plant the CO_2 is first pressurised to raise liquefaction temperature, and is then cooled. In this thesis, a liquefaction pressure of 20 bar and transportation at 7 bar and -50 °C is assumed, based on a report from Deng et al [20]. The liquefaction plant consists of a compression train with intercoolers, an ammonia refrigeration cycle, knock-out drums and a circulation pump.

2.5.2 Transport & Intermediate Storage

When the CO_2 is in a liquid state it may be transported. The challenges with transporting the liquefied gases are the large volumes, logistics and cost evaluation. There are four possible alternatives for transportation: by truck, by rail, by ship or by pipeline. Ship transport is often the preferable option, but it is determined by volumes, distance and ocean access. Which alternative of transportation that should be used is often location specific. Models have been used on regional scale to optimise capture, transport, storage and usage since shared transport and usage can bring down costs [21][22]. If multiple modes of transport are to be used or if the transport is intermittent, intermediate storage is required. The cost of intermediate storage depends on the required maximal stored volume [21].

2.5.3 Geological Storage

For Northern Europe the North Sea seabed has been pointed out as a good site for geological storage of CO_2. The Norwegian government has supported a project called Longship in an effort to kick-start a carbon capture and storage (CCS) market [23]. Longship will together with local industry finance the geological storage project Northern Lights at Øygardens, just outside Bergen, Norway. Northern Lights aims to sell carbon storage as a service where by Northern Lights collect liquefied CO_2 by ship at ports around Europe. The ship then transport the CO_2 to Øygardens to be injected into a geological storage below the North Sea seabed. The project will have an initial capacity of $1.5MtCO_2/year$ with possible expansion up to $5MtCO_2/year$ [24]. The Northern Lights project has not announced any costs for the service. But According to Skagestad et al. [25] an injection cost of 7€/tonCO_2 is possible for a location near to the one used in the Northern Lights project.

3

Method

The study has been conducted through three separate analyses. First a mass and energy balance for all of LKAB's pelletizing plants that investigated flows of iron, carbon and heat. The second part was a computer modelling of the carbon capture plant to evaluate technical feasibility, sizing of equipment and heat demand. The computer modelling analysis was only done on the flue gases from the PH-stack on LKAB's largest pelletizing plant KK4. The third part was an economic assessment for two proposed CO_2 utilization routes, the CO route and methane route where CO_2 was converted to CO and methane respectively which then carburizes DRI. Additionally a CO_2 storage route was included where the CO_2 was transported and stored in a geological storage, for the storage route biomethane was used for carburization. The routes were compared in an economic assessment where the costs for the carbon capture plant and relevant process steps was investigated. In Figure 3.1 the structure of the thesis is visualised.

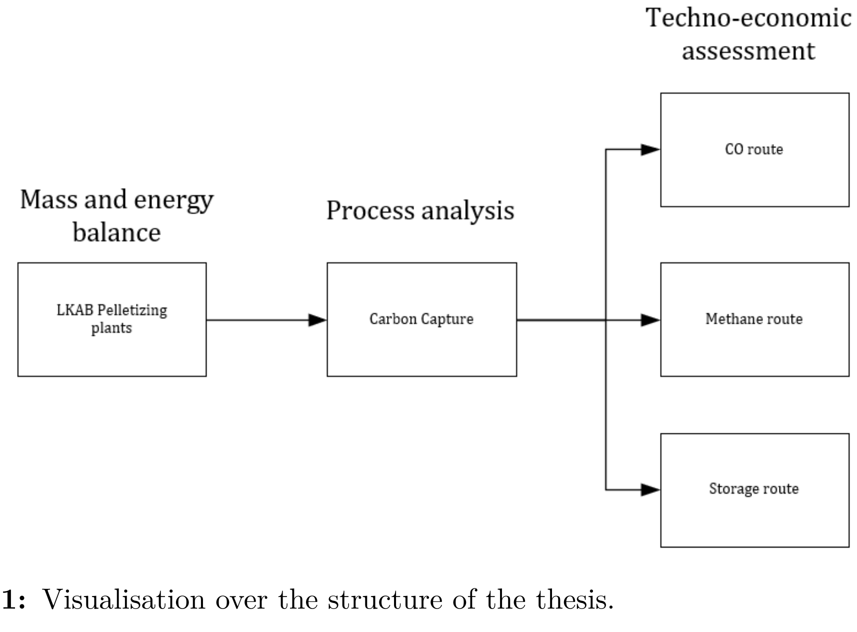

Figure 3.1: Visualisation over the structure of the thesis.

3.1 Mass and Energy analysis

A Mass and Energy analysis was performed for each plant to investigate several parameters of interest. In Table 3.1 the key performance indicators (KPIs) of the mass and energy analysis are displayed, as well as the input variables used in the analysis. The KPIs are either general for all route or route specific. General KPIs are the amount of carbon that can be captured and the heat demand of the carbon capture process, additionally the potential level of carburization of DRI for one hour of pellet production and carbon emissions is quantified as well. Route specific KPIs are further explained in the following chapters.

Table 3.1: Input and KPIs of mass analysis for each route.

Input		KPIs		
Pellet production	ton/h	Captured CO_2	ton/h	All routes
Flue gas volume	kNm3/h	CO2 Capture heat demand	MJ/h	All routes
Flue CO_2 conc.	CO_2vol%	Carburized DRI	ton/h	All routes
Flugas heat	MJ/h	Carburization degree	wt%	All routes
		Hydrogen for DRI	ton/h	All routes
		Hydrogen for Carbon carriers	ton/h	CO & CH4
		Methane production	ton/h	CH4
		Excess hydrogen	ton/h	CH4 & CCS
		Methanation excess heat	MJ/h	CH4 & CCS
		DRI heating/cooling	MJ/h	All routes

In Figure 3.2 the generic schematic of the CO_2 utilization system considered in this thesis is shown.

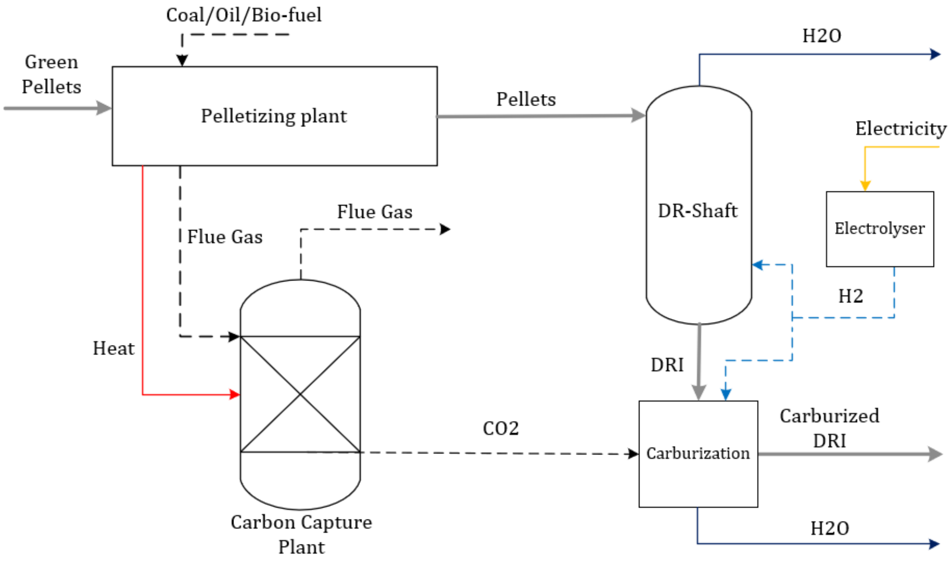

Figure 3.2: Generic process scheme of the CO_2 capture and utilization system.

The system consists of a pelletizing plant, carbon capture plant, DR-shaft and a carburization chamber in direct conjunction to the DR-shaft. The different routes and additional process steps are explained further in chapters below. Pellets produced in the pelletizing plant are sent to the DR-shaft to be reduced with hydrogen to DRI. The flue gases from the PH-stack are captured in a carbon capture plant with a capture rate of 90%. The energy demand in the carbon capture process is supplied from heat above 120°C in the flue gases, the amount of heat is further discussed in Chapter 4.2. The captured CO_2 is, in the CO-route, processed together with H_2 to carburize DRI. Assumptions for the pelletizing and CO_2 capture plant are shown in Table 3.2.

Table 3.2: Generic assumptions for all routes

Pellet production and CO2 capture assumptions	
Pellet hematite content	95%
Pellet inert substances content	5%
Flue gas available heat	>120°C
CO_2 capture rate	90%
Carbon Capture reboiler duty	3.44 MJ/kg CO_2

3.1.1 CO route

The CO route involves converting captured CO_2 to CO which is then used for carburization in direct conjunction to the hot DRI exiting the DR-shaft. For the CO-route the generic system displayed in Figure 3.2 is used with the carburization occurring as shown in Figure 3.3.

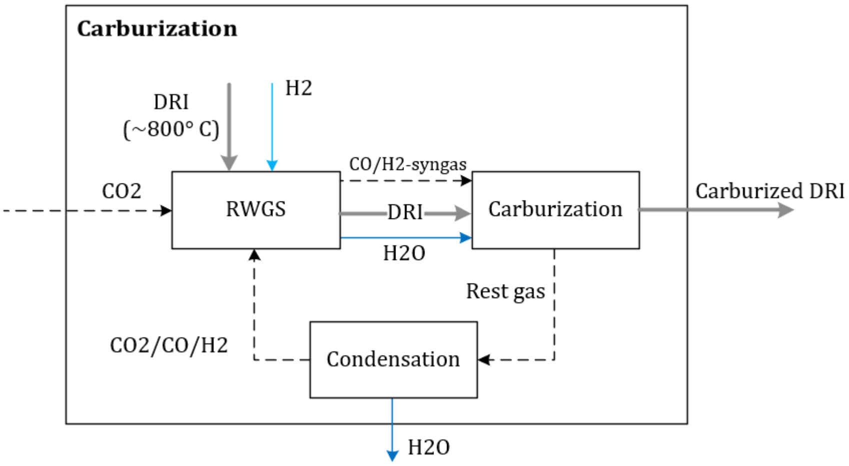

Figure 3.3: Process for joint CO conversion and carburization of DRI.

H_2 and CO_2 at a 2:1 molar ratio is injected on the hot DRI, increasing the gas temperature to one favourable for CO-formation via RWGS (Chapter 2.4.1), creating a CO and H_2 syngas. The syngas can then carburize DRI (Chapter 2.2.1). While RWGS and CO-carburization are well known, a joint process has not been studied before. The carburized DRI leaves the chamber and the rest gas has its water content separated before recirculation.

Relevant KPIs in this route are the amount of H_2 needed to fully reduce CO_2, and net heating of DRI after the endothermic CO formation and exothermic carburization. In an equilibrium state at 750°C only 72% of CO_2 conversion is expected. In addition a portion of the carburization could occur with Reaction 2.7. This would mean that a significant amount of the input CO_2 could remain in the rest gases. This would create a need for recirculation with a condenser to remove formed water. However it is assumed that with recirculation eventually all injected CO_2 undergoes a carburizing reaction. The oxygen leaves the system as H_2O and carbon in the DRI.

3.1.2 Methane route

The methane route includes a compression train for the captured CO_2 and a methanation reactor where CO_2 is converted to methane which can be used to carburize DRI. As this thesis assumes that all captured carbon is used for carburization, 100% methanation efficiency is assumed. A scheme of the methane route system can be seen in Figure 3.4.

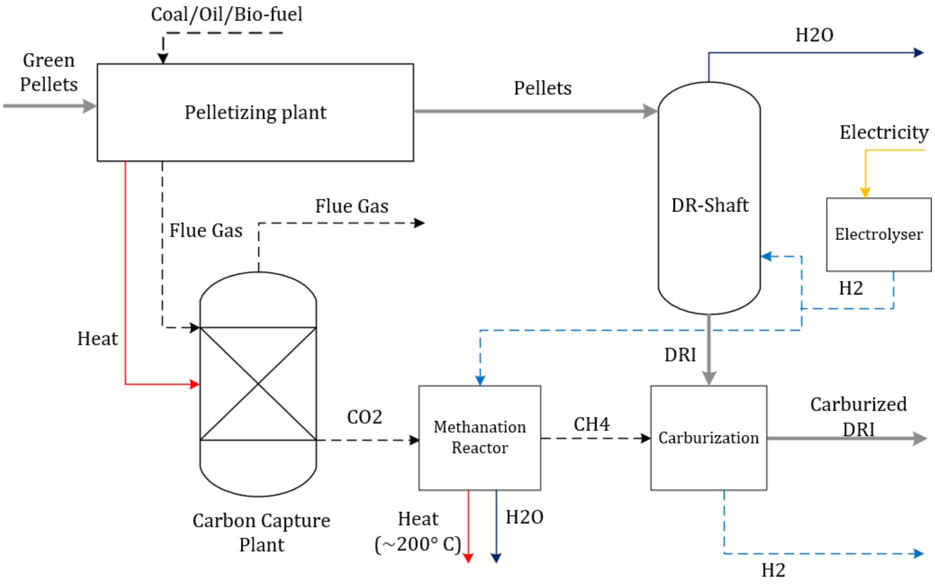

Figure 3.4: A schematic of the methane route used in the Mass & Energy balance.

The methane route differs from the CO route by introducing a methanation reactor to synthesise methane (see Chapter 2.4.2). The captured CO_2 is first pressurised to 20 bar before entering the exothermic methanation process together with hydrogen at a 4:1 H_2/CO_2 molar ratio. The excess heat from the methanation is assumed to sufficiently preheat the gases. The produced methane is injected to carburize the DRI (see Chapter 2.2.1), which also cools the DRI and releases hydrogen. It is assumed that the released hydrogen can be recovered and combusted to provide heat for the process and to reheat the DRI.

Relevant KPIs are methane production from the captured CO_2, H_2 demand and excess heat from the methanation reaction. In addition, the cooling of DRI from the endothermic carburization reaction is calculated. The excess hydrogen from the methane decomposition is quantified.

3.1.3 Storage route

The storage route sends captured CO_2 to long term storage while carburization occurs with biomethane. The amount of biomethane is equal to the amount of methane produced in the methane route for a fair economical comparison. A scheme of the storage route system can be seen in Figure 3.5.

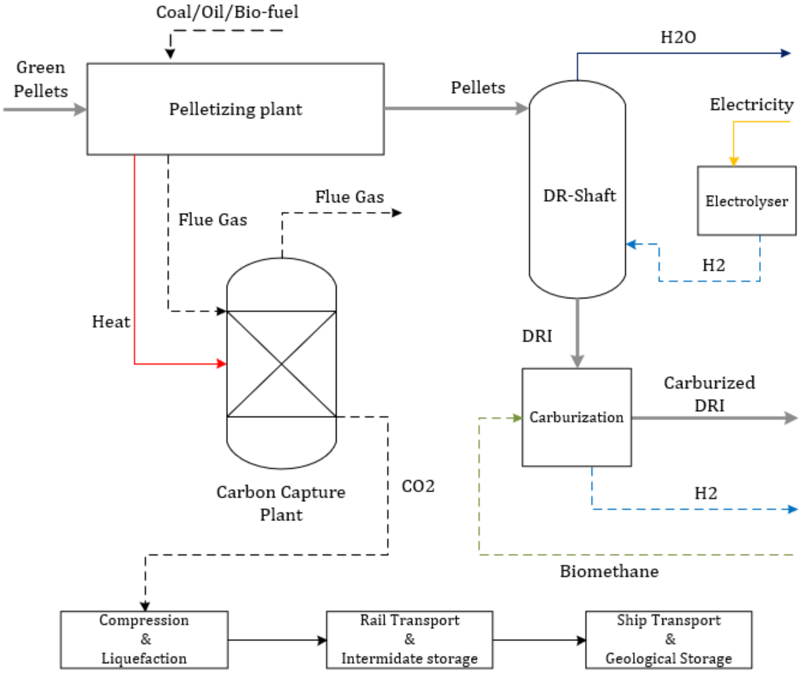

Figure 3.5: A schematic of the storage route used in the Mass & Energy balance.

In order to store the CO_2 long term it first has to be compressed and liquefied at the capture facility. The liquid CO_2 is then transported to a final storage location. In this thesis a fixed route originating from Kiruna is investigated, shown in Figure 3.6. The transport route is assumed to be a feasible alternative for LKAB's operations. Others routes may exists but are not explored in this thesis. The first transport step is rail transport by a lorry company along Malmbanan to Narvik. In Narvik intermediate storage is used for the CO_2 awaiting ship transport along the Norwegian cost down to Northern Lights geological storage at Øygardens. The carburization of DRI is achieved with biomethane, which similarly to the methane route cools DRI and releases hydrogen. For the storage route the most relevant KPIs are the hourly and yearly mass flows of captured emissions which scale the compression, liequefaction, rail transport and intermediary storage.

Figure 3.6: Map over the CO_2 transport route with key locations marked: (1) Kiruna, (2) Narvik , (3) Øygardens. The transport between (1)-(2) is by rail and the transport from (2)-(3) is by ship.

3.2 Carbon Capture Process Modelling

An ideal flue gas for carbon capture has a high volume of CO_2 at high concentrations. The flue gases from a single pelletizing plant have a volume of about 100-200ktonCO_2/year which is significant but with a CO_2 concentration below 3%vol it may not be economically viable to capture. Due to this, modelling and proper cost assessment was deemed necessary. The flue gases from the pelletizing plant are captured using a temperature-swing design with MEA as described in 2.3.1. The capture process was modelled in Aspen Plus V11 to determine two KPIs with specific heat required in the reboiler being the first. The second was the annual cost of the plant, described in 3.3.1. Due to the modelling of the Carbon Capture process being time consuming only the flue gases from the largest pelletizing plant KK4 was modelled, the other plants were then assumed to have a similar specific heat and annual costs. An analysis of CO_2 concentrations was made to assure that the design for KK4 flue gases representing the other plants. The model of the capture process was created by Garđarsdottir [26] and later extended and utilised for analysis of industrial processes in [27].

3.2.1 Simulation Setup

The input to the model was the flue gas specified by flow rate, temperature, pressure and volumetric composition, shown in Table 3.3. The flue gas specification will influence the dimensioning of components throughout the model.

Table 3.3: The properties of the flue gases for KK4.

Flue gas composition							
Flue gas	$\dot{V}[kNm^3/h]$	T [°C]	P [bar]	CO_2%	O_2%	N_2%	H_2O%
KK4	426.8	131	1	2.6	12.4	78	7

The first column the flue gas passes through is the DCC Track changes is off Everyone Track changes for everyone You Track changes for You carlem Track changes for carlem Guests Track changes for guests You: väldigt många varianter av "include" i det här stycket Jun 3, 2021 2:19 PM • Edit Hit Enter to reply Current file Overview 36 which removes water resulting in a flue gas saturated at 40°C. The cooled flue gas enter the absorber column at 1.1 bar where the lean solvent extracts the CO_2. The target of the model is to reach a capture rate of 90%. The solvent flow rate is adjusted until the target capture rate is reached. The CO_2 rich solvent from the absorber exchange heat with the lean stream and then enter the desorber. In the desorber the solvent is heated to 121°C to regenerate the CO_2, the heat demand is the first important KPIs for the capture plant. The released CO_2 gas is dewatered and a stream of pure CO_2 is achieved. In Table 3.4 the simulating setup specification is shown. The four columns are modelled with structured packing of the type Koch-Glitsch FLEXIPAC 2X. The separation in all columns is modelled through rate-based calculations, the height is fixed while the diameter is calculated so that flooding in the column does not exceed 80%. The washer diameter was set equal to the absorber, as it in reality constitutes the top part of the column. Therefore, instead the height was adjusted to avoid flooding.

The low concentrations in the flue gases from the pelletizing plant may lead to unnecessary reboiler heat. A higher absorber height can be tested to lower heat demand as investigated by [28, p.238]. In an effort to lower specific reboiler duty, changes to absorber height was made and are explained in Appendix A, with the result that a packing height of 27m is required to lower reboiler heat demand.

Table 3.4: ASPEN modelling settings

Aspen modelling setup	
Lean loading	23 %mol
Absorber packing height	27m
Stripper packing height	9m
DCC height packing height	7m
Cross heat exchanger ΔT	10°C
Lean solvent temperature into absorber	40°C
Lean solvent pressure into absorber	1.1 bar
Desorber reboiler temperature	121°C

3.3 Economic Assessment

An economic assessment was performed for each route with investment and operational costs for each process. For the CO route, the carbon capture process and hydrogen production cost was calculated. The methane route was similar but also included costs for a compression train and the methanation reactor. The storage route instead add costs for a CO_2 capture and liqufaction plant, CO_2 transportation and storage. Additionally, costs for producing biomethane were included. The method for calculating costs of each process are described further in this chapter.

3.3.1 Carbon Capture Plant

The economic evaluation of the carbon capture process was done according to the method proposed by Ali et al. [29] and used by Eliasson and Fahrman [27]. The method uses an estimation strategy where each piece of equipment is viewed as an independent plant resulting in a more accurate early cost estimation. The methodology provides an estimation of the equipment installed costs (EIC) for each piece of equipment and the total installed cost (TIC) for the carbon capture plant. TIC is used to estimate capital expenses (CAPEX) and combined with operational expenses (OPEX) an annualised cost can be found for the carbon capture plant. Results and stream data collected from the Aspen carbon capture model is shown in Table 3.5. Materials engineering cost data used was provided by Eliasson and Fahrman, described in [27, Appendix B]. Economic parameters of plant lifetime, interest rates and OPEX costs assumptions used are shown in Table 3.6.

Table 3.5: ASPEN data collection and Economic assumptions

Data collection	
Column and tank	Volume[m^3]
Heat exchanger	Area[m^2]
Pump and fan	Work[w]
Stream	flow rates [kg/h]

Table 3.6: Economic data for the carbon capture plant

Economic parameters	
Lifetime	23+2[y]
Interest rate	7.5 %
Full load hours	8200 [h/y]
Maintenance, insurance and labour	6 % of TIC
Electricity Price*	30 [€/MWh]
Steam price**	2 [€/MWh]
Cooling water price	[0.02€/m^3]
MEA price(including slugde disposal)	2000 [€/m^3]
NaOH price	270 [€/t]

* Assuming a low average electricity price in price zone SE1.
** Assuming all heat can be found on site.

3.3.2 Hydrogen Production

LKAB is investing in electrolysis and hydrogen storage capacity to supply the DR-shaft with hydrogen, but the cost of this is undisclosed. Costs for electrolysis investments was therefore adopted from [30], where the projected cost data for alkaline water electrolyser (AEL) in 2030 is used. Additionally, electricity cost for the hydrogen production was included. As the cost for hydrogen production is highly sensitive to the price of electricity, a sensitivity analysis of the electricity cost was done for a ±50% price change. The cost data for the hydrogen production is displayed in Table 3.7.

Table 3.7: Economic data for hydrogen production

Alkaline electrolyser cost data	
AEL efficiency	80%
AEL investment cost	580 €/kW
AEL lifetime	20 years
Full load hours	8200h
Stack replacement cost	305 €/kW
Stack lifetime	120000h

3.3.3 Methanation Reactor

The investment cost of a methanation reactor was estimated from the research of Götz et. al [19] which concludes that there is a strong uncertainty regarding methanation investment costs. However the costs calculated by Outotec GmbH was deemed realistic which are presented in Table 3.8. The cost data suggests lower investments cost with increased reactor capacity. In this thesis the 110 MW investment cost was used due to the required reactor sizes presented in Chapter 4.1.

Table 3.8: Economic data for methanation.

Methanation reactor cost data, 20 bar operating pressure	
Methanation 5 MW Investment cost	400 €/kW$_{CH4}$
Methanation 110 MW Investment cost	130 €/kW$_{CH4}$
Plant lifetime	25 years
Full load hours	8200h

3.3.4 Biomethane Production

Biomethane was assumed to be produced from the gasification of solid biomass. The method of calculating biomethane production costs was obtained from [31] where the gasifier equipment cost is added to the biomass cost at a conversion rate of 70%. The gasifier equipment cost was taken from [32]. The cost of solid biomass is sensitive, due to competition as more industry sectors choose to rely on biofuels for replacement of fossil fuels. A study on the access to biomass in more competitive environment has compared current biomass prices in Sweden with projected prices 2040, resulting in a cost range of 20-45 €/MWh for solid biomass [33]. For this thesis an average price for solid biomass of 30 €/MWh was assumed. The impact of biomass price uncertainty was analysed in a sensitivity analysis with cost changes of ±50%. The assumptions for the biomethane production are presented in Table 3.9.

Table 3.9: Economic data for biomethane production.

Biomethane gasification cost data	
Gasifaction equipment	20 €/MWh
Biomass cost	30 €/MWh
Biomass conversion efficiency	70 %
Plant lifetime	20 years
Full load hours	8000h

3.3.5 Compression and Liquefaction

The investment costs for the compression train and the liquefaction plant was evaluated as a single unit using cost data from literature and CO_2 flow as a sizing parameter [20] [29] [34]. The EIC was calculated by interpolating cost data for a plant operating at same transportation and delivery pressure acquired from the work of Deng et al [20]. An escalation factor of 0.6 was assumed and the entire power function can be seen in Equation 3.1. The TIC for the plant was calculated by multiplying the equipment cost with cost factors, which can be seen in Table 3.10. A 6% fixed OPEX was assumed. The electricity and cooling consumption of the compression train was determined through Aspen modelling. The economic parameters used for the evaluation was the same as for the carbon capture plant, seen in Table 3.6. The investment and operational costs for the compression train and liquefaction plant are presented separately due to the methane route requiring CO_2 compression without liquefaction.

$$C = C_0 \cdot (\frac{\dot{m}_{CO2}}{\dot{m}_{\mathrm{CO}_{20}}})^{0.6} \tag{3.1}$$

Table 3.10: Cost functions for the Liquifaction plant

Liquefaction plant cost functions	
Process contingency	15%
Owners cost	7%
Indirect costs	14%
Project contingency	20%

The compression train was modelled in ASPEN to find the power and cooling consumption, a process scheme can be seen in Figure 3.7. The model included a three-stage compression train with intercoolers and knock-out drums to separate remaining water. The compressors was modelled with an isentropic efficiency of 85% with the Peng-Robinson equation of state with Boston-Mathias extrapolation for the compression sequence [20].

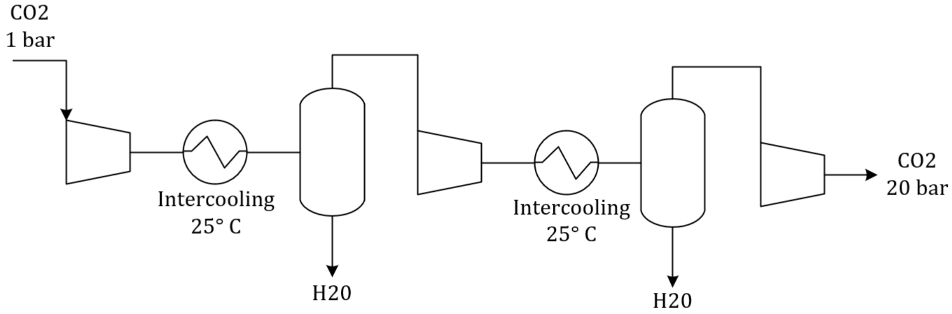

Figure 3.7: Process scheme of the compression train modelled in Aspen.

3.3.6 Rail Transportation and Intermediary Storage

In [21, Section 2.5] Karjunen presents a method for transport and intermediate storage, this method is used for optimising multiple nodes of CO_2 emitters , users and storage. This method was used but changed to apply to a single fixed route. For transport by train, Equation (3.2)-(3.5) were used to find the number of lorries, N, required and the final OPEX cost. The assumptions for the specific route Kiruna-Narvik used in the storage route are shown in Table 3.11.

$$L_{tot} = 2L \cdot \frac{Q_{tot}}{Q_{Load}} \tag{3.2}$$

$$t_{tot} = \frac{L_{tot}}{v} + \frac{Q_{tot}}{Q_{Load}} \cdot t_{load} \tag{3.3}$$

$$N = \frac{t_{tot}}{t_{ava}} \tag{3.4}$$

$$OPEX = L_{tot} \cdot (FP + M) + t_{tot} \cdot DW + N \cdot G \tag{3.5}$$

Table 3.11: Data used for liquid CO_2 transport from Kiruna to Narvik, partly based on [21, Table 5]

Rail transport assumptions	
Variable	**Value**
Distance, L	180 km
Annual emissions, Q_{tot}	140 ktonCO$_2$
Availability, t_{ava}	4380 h/y
Investment	487577 €/unit
Fuel cost, FP	1.34 €/km
General Expense, G	3175 €/y
Hourly costs, DW	27.55 €/h
Loading time, t_{load}	12 h/trip
Maintenance, M	0.0886 €/km
Capacity, Q_{load}	80 $tCO_2/trip$
Speed, v	45 km/h
Interest rate	10%
Lifetime	10 years

For intermediary storage in Narvik a storage facility of 33kton CO_2 was assumed, this would allow for storage of 80 days for the case of CO_2 captured at KK4. The cost data for intermediate storage was taken from [21] and scaled using Equation (3.1). The annual OPEX was assumed to be 1% of CAPEX.

3.3.7 Shipping and Geological Storage

Costs from the Northern Lights project are not yet known but general cost for shipping and geological storage exists. The shipping cost depends on a number of factors including the size of the ship used, how the ship is operated, fuel prices and if it is a joint venture with other companies looking to transport liquid CO_2. Kjärstad shows in [22] how different sizes of ships and transport distance affect the shipping cost. For this thesis a fixed cost of 25 €/tonCO$_2$ was assumed, equivalent to 0.5 Mton ship. For geological storage the cost was assumed to be 7 €/tonCO$_2$ as proposed by Skagestad et al, [25].

4

Results

In this chapter the results from the mass and energy balance, the carbon capture modelling and the economic assessment are presented. Individual mass and energy balances were calculated for each of LKAB's pelletizing plants, while the carbon capture evaluation and economic assessment was performed only for KK4.

4.1 Mass and Energy Balance

The mass and energy balance was done for each route and pelletizing plant. The analysis resulted in three significant results which are presented in this chapter. Firstly, the mass of elemental carbon in the captured CO_2 is enough to carburize the DRI to an interesting degree of carbon content, as highlighted in Figure 4.1. The results should be compared to carbon content in commercial DRI of 1-4.5%wt or HBI of 0.5-1.6%wt. Secondly, the available heat in the flue gases is enough to supply the carbon capture process. Thirdly, the CO_2 utilization routes uses significantly less H_2 then what the DR-shaft would consume, visualised in Figure 4.2. This means that the additional electrolysis and hydrogen storage capacity needed to realise the CO and methane route is relatively small when compared to LKAB's investments for the direct reduction process.

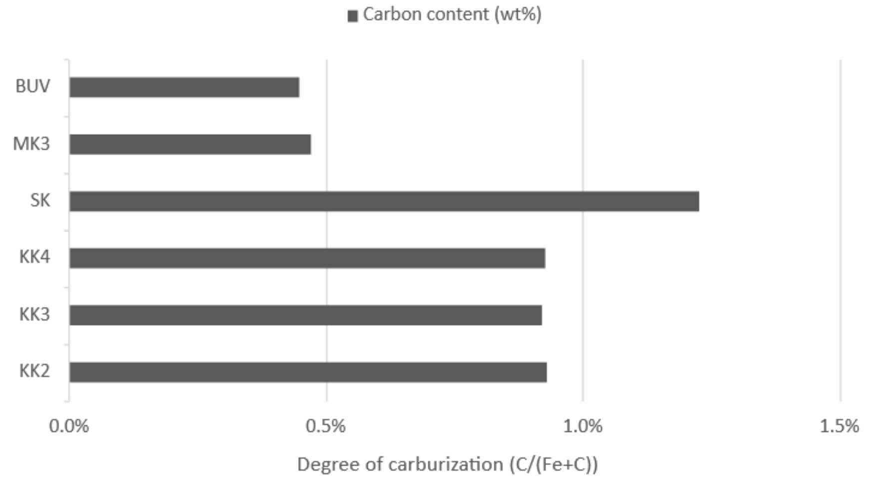

Figure 4.1: Potential degree of carburization of DRI with captured CO_2 emissions from the pelletizing plants. Assuming all captured CO_2 is used to carburize DRI.

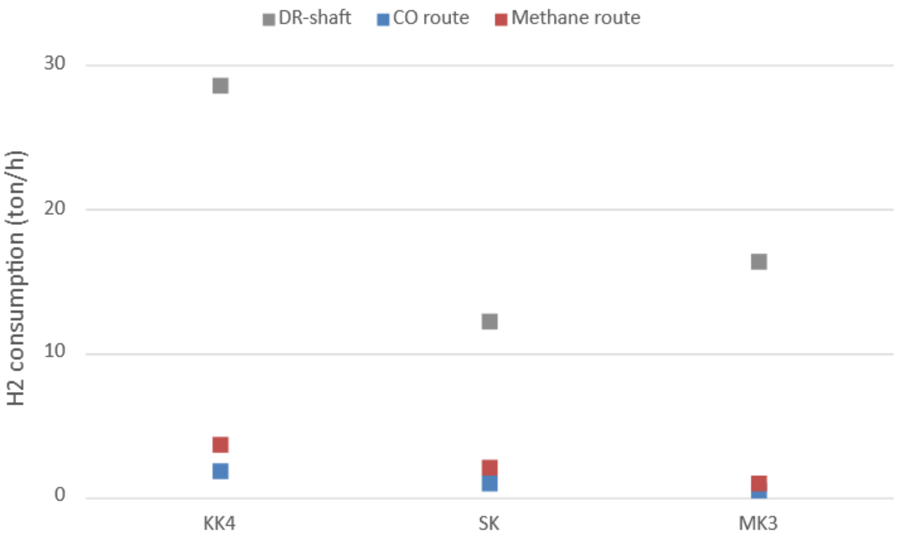

Figure 4.2: Comparison of H_2 consumption between the DR-shaft, CO route and methane route for three pelletizing plants.

The CO route is the most simple route as it has the fewest number of process steps. However while CO_2 to CO conversion through RWGS and CO carburization both are known mechanics, no studies for a joint process were found meaning that additional research is required to validate that both reactions can work together. The methane route has the benefit that it utilizes well known technologies: carbon capture, methanation reactor and carburization with methane. The storage route has the potential to be CO_2 negative if the emissions are biogenic, additionally the carburization degree is not set by CO_2 emissions captured as the other routes. Regarding transportation possibilities the CO route promotes CO_2 capture and carburization at the same location as efficient CO_2 transportation would require compression and liquefaction. The methane route has more options as the methane is produced at 20 bar. The results also showed a possible synergy between the excess heat from the methanation reactor and heat demand of the carbon capture process, as both are in the same magnitude and at the right temperature. The methanation reactor capacity required for the different pelletizing plants are in the range of 35 MW (MK3) to 110 MW (KK4).

The carburization reactions can impact the heat balance of the DR-shaft and possible HBI production. In the CO route the DRI would be heated by roughly 140°C which could be beneficial for HBI production. On the other hand carburization with methane and biomethane cools the DRI by roughly 170 °C. To compensate for the cooling of the DRI, it could be reheated by combusting approximately 20% of the excess hydrogen.

4.2 Carbon Capture Modelling

The Carbon capture modelling produced two important results. The carbon capture plant with MEA as solvent can be designed to capture CO_2 at low concentrations. The carbon capture modelling was only made on the flue gases from KK4 at 2.6%vol but results showed that the design for KK4 can function for a span of concentrations from 1.5% to 3.25%. The second result was that this can be done with an acceptable reboiler heat demand per kg CO_2 captured. The modelling in ASPEN showed that with the right setup a CO_2 concentration of 2.6%can be captured with reboiler heat duty of 3.44 MJ/kgCO_2 at a capture rate of 90%. The reboiler heat duty will vary with CO_2 concentration and capture rate, therefore an off-design analysis was made.

4.2.1 Sensitivity Analysis of CO_2 Concentration

To investigate how sensitive the plant design was to different CO_2 concentrations, an off-design analysis was made. When the concentration in the flue gas changed, the solvent flow rate was adjusted to keep the capture rate at 90%. In Figure 4.3 it is shown that concentrations from 1.5-3.25%vol are possible to capture with the modelled design with only a small increase in heat requirement for lower concentrations. For concentrations above 3.25%vol and the capture rate at 90% the absorber reaches 100% flooding. A larger design or lower capture rate would allow for higher concentrations of CO_2 without reaching the flooding limit. For concentrations bellow 1.5%vol the rich stream loading starts to decrease rapidly leading to a higher specific heat consumption. The only pelletizing plant that would require a significant redesign of the carbon capture plant is BUV which has a steady state operation flue gas flow far below CO_2 1.5%vol, this was not further investigated in this thesis. From this it was assumed that design for the carbon capture plant of KK4 was representative for all pelletizing plants except BUV.

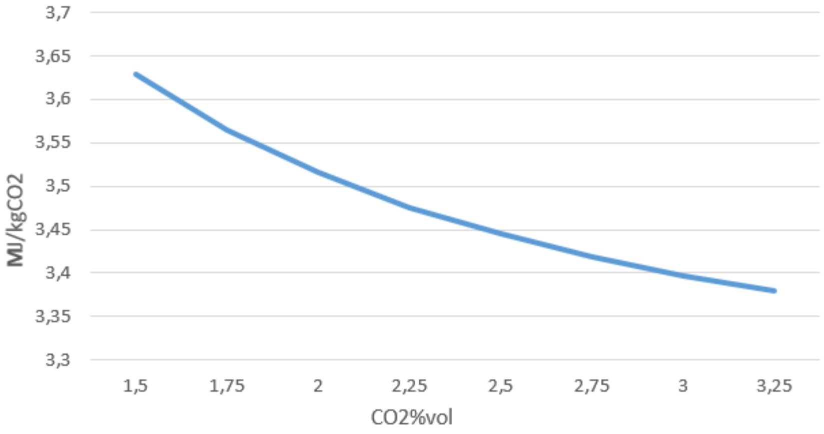

Figure 4.3: The specific energy demand for different CO_2 concentrations. For all points the design of the plant is the same and Capture rate is fixed at 90%

4.3 Economic Assessment

In this chapter the results from the economic assessment are presented. The total cost of each route are presented in the Figures 4.4-4.5 as a specific cost with a different denominator: per ton captured CO_2 and per ton DRI. In the mass and energy analysis it was determined that 20% of the excess H_2 released from methane carburization needs to be combusted to reheat DRI, which means that the remaining 80% is available for use and is presented as a negative cost. One additional scenario was added where no carbon capture, which is a reference case for if the carburization would occur with only biomethane and emissions from the pelletizing plants are eliminated through other measures, eg. electrification. Black bars are added in the figures which represents sensitivity of the results, further explained below.

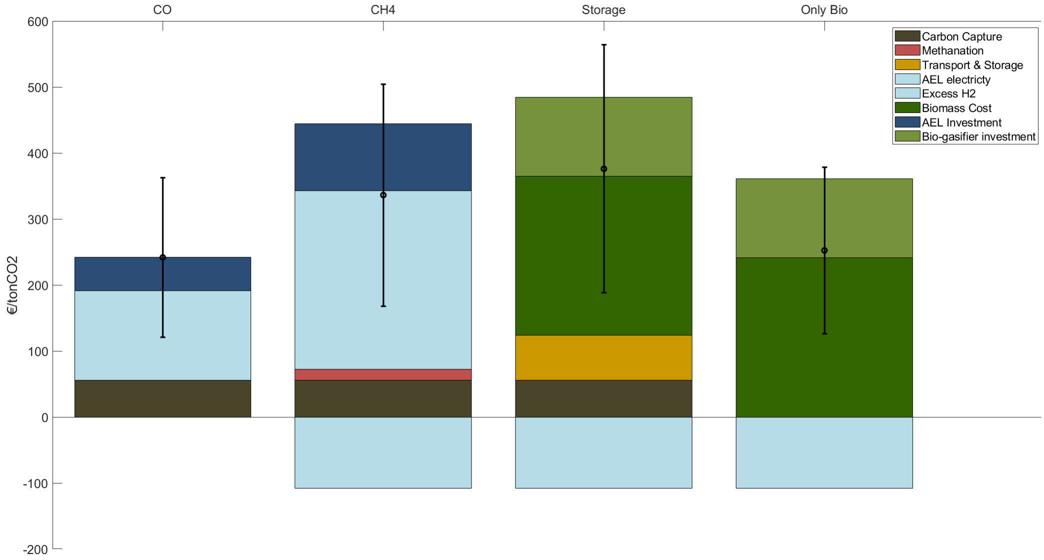

Figure 4.4: Specific cost per kg captured CO_2 for the CO, methane, storage and no carbon capture route. With combined results from the sensitivity as error bars.

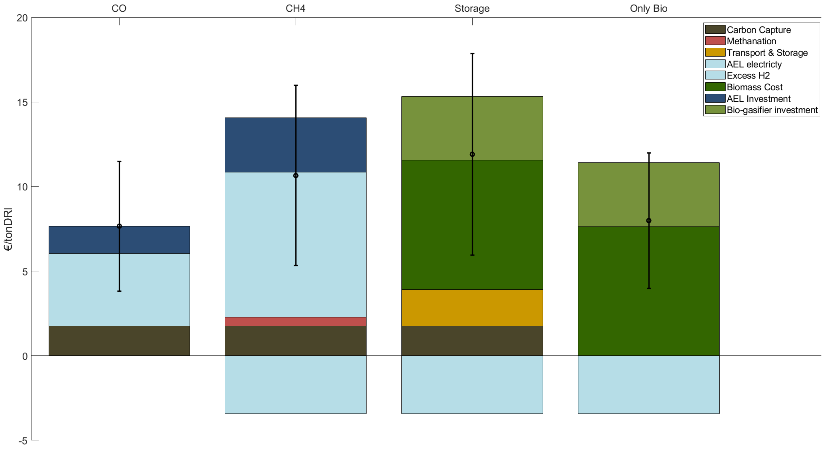

Figure 4.5: Specific cost per ton carburized DRI for the CO, methane, storage and no carbon capture route. With combined results from the sensitivity shown error bars.

The economic assessment showed that the CO route had the lowest cost closely followed by the no carbon capture case and with the methane route and storage route being more expensive. The costs for electricity consumption and biomass feedstock was dominating in all routes, indicating that the cost results are very sensitive to the cost of electricity and biomass. Additionally the sensitivity to CAPEX cost was of interest. To investigate this, a sensitivity analysis was made where the cost of CAPEX, electricity and solid biomass was all altered by ±50%. The results of sensitivity analysis can be seen in Figure 4.6.

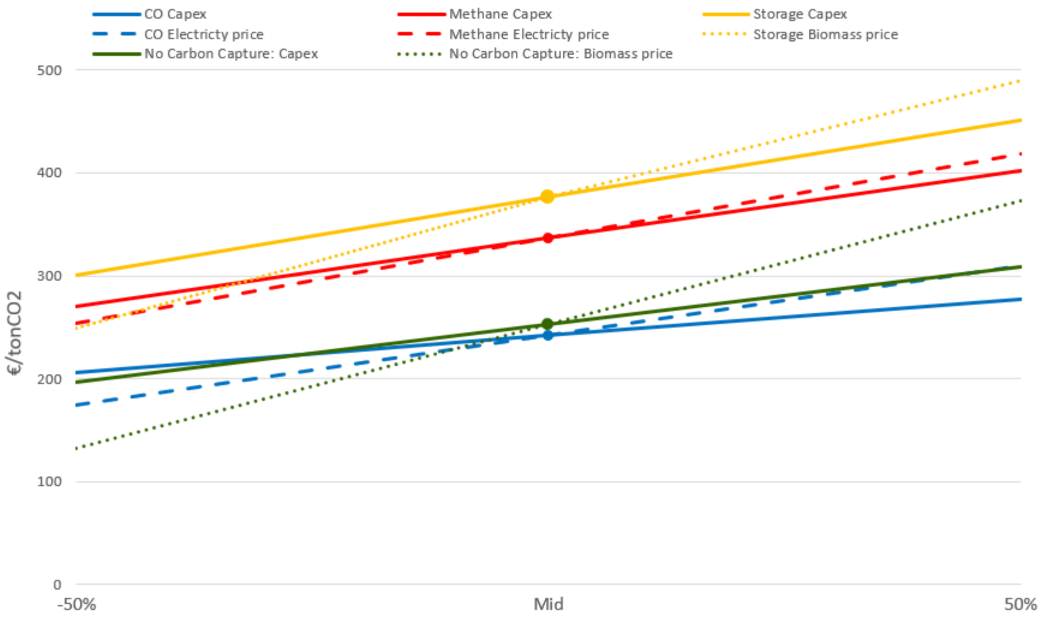

Figure 4.6: The figure show how sensitive each route is to CAPEX changes, electricity and biomass cost variety of ±50%.

The primary results from the sensitivity analysis was that the economic results are more sensitive to the price of electricity and biomass costs than CAPEX costs. This means that future price of electricity and solid biomass is important when comparing the different routes.

4.3.1 Two Cases for Higher Carburization Degree

As shown by the mass and energy analysis, each pelleting plant can only reach a carbon content of 0.5 - 1.2 wt%. But compared to commercial DRI with 1-4.5%wt a higher carbon content might be necessary. To facilitate this, it is possible to either inject additional biomethane, or to capture CO_2 from several plants to carburize the DRI production of one plant. For this analysis, a system was proposed where CO2 is captured from all three plants in Kiruna (KK2, KK3 and KK4) and used to carburize the DRI production of only KK4. This is relevant if KK2 and KK3 remains as only pellet production facilities. These two possibilities shown in Figure 4.7.

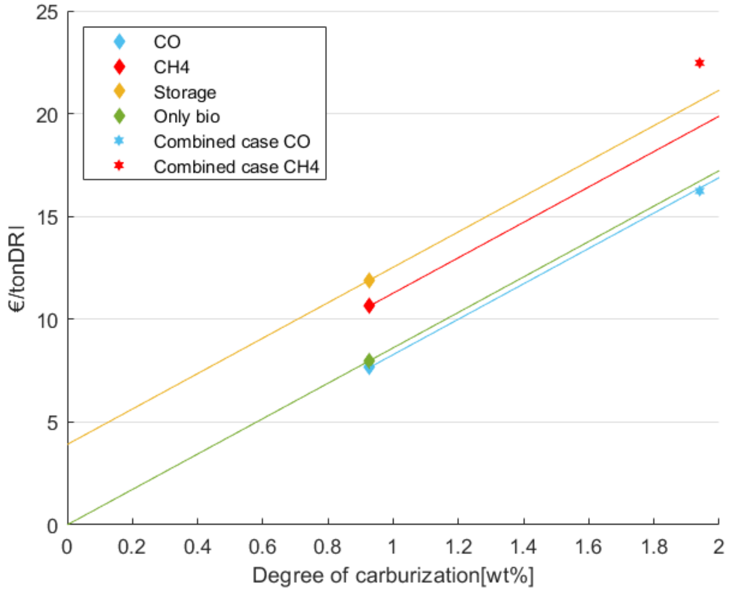

Figure 4.7: The figure shows how higher degree of carburization can be achieved either by biomethane(lines) or combined carbon capture of several plants(stars). The diamonds show each routs costs for KK4.

In Figure 4.7 results from the economic assessment of KK4 are shown as diamonds, at 0.92 wt%, with flexible biomethane carburization shown as lines. For Only bio or COtogether with biomethane a 2wt% can be achieved for about 17 €/tonDRI. In the combined cases (shown as stars in Figure 4.8), a carburization degree of 1.94% can be reached. For the CO route, the combined case results in a similar cost as COtogether with biomethane. While the methane route combined case is significantly higher than its counterpart.

4.3.2 Carbon Capture Plant

The economic evaluation for the carbon capture plant resulted in an investment cost of 40.6M€. Adding annualised CAPEX and OPEX resulted in a final cost of approximately 56€/tonCO_2. This cost is within range of what Johnsson et al. specifies expected costs for a carbon capture plant of 40-110€/tonCO_2 [12]. The low concentration requires some components, like the absorber, to be larger than usual, while some other components such as heat exchangers becomes smaller. Due to excess heat being available on site the steam price and in extension the OPEX becomes quite low. If heat had to be produced on site, for instance from natural gas combustion, the cost for carbon capture would be higher.

4.3.3 Transport and Storage Costs

The cost of CO_2 storage is divided in several sections, which are presented in Figure 4.8. Train transportation costs could be overestimated as the method assumes a lorry company while LKAB could have the infrastructure to do the transportation themselves. And the cost for the geological storage was assumed from literature and could be different when the Northern Lights project comes to fruition.

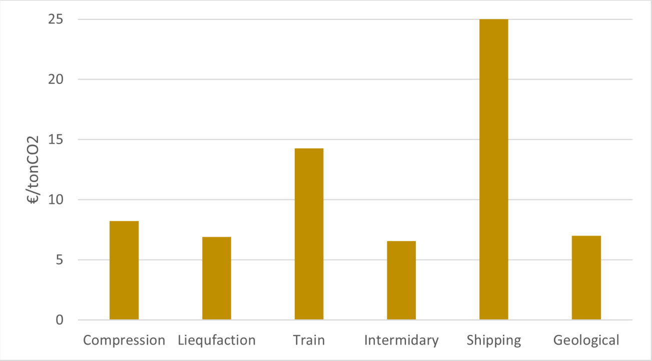

Figure 4.8: Costs for each step of the transportation and storage, showed in €/tonCO_2

5

Discussion

5.1 Methodology and Fundamental Assumptions

The thesis was made from a systems perspective with as low number of steps as possible for each route to be comprehensive. For most of the processes the mass and energy flows are simplified to input and output and costs are set for CAPEX and OPEX from literature. The most detailed process analysis was for the carbon capture plants, this was mainly due to the low CO_2 concentrations in the flue gases which brought uncertainty of technical and economical viability. Due to this the carbon capture modelling was made. This thesis is built as a continuation of LKAB's transition to fossil free DRI production, and thereby keeps some of the same central assumptions. These are the access to low cost electricity and lower investment costs for electrolysis capacity as production volumes ramps up. An additional central assumption for this thesis is that a carbon content in DRI is necessary in LKAB's products. In fact HBI briquetting and EAF melting with DRI with no carbon content is possible but has its own challenges.

5.2 Carbon Capture Plant

The carbon capture modelling on KK4 showed that it is possible for LKAB to implement a CO_2 capture process from both a technical and economical perspective. While some concerns regarding feasibility of capturing low CO_2 concentrations existed prior to the thesis, the modelling showed that with a sufficiently large absorber it was still possible to capture 90% without increasing the specific heat demand in the reboiler. LKAB has one advantage for CO_2 capture implementation: availability to excess heat on site from the flue gases. Without excess heat the heat demand would have to be satisfied by biomass or natural gas combustion, making the case for implementing carbon capture much weaker.

5.3 Emissions and Biomass

The Paris Agreement requires that European countries cut all GHG emissions by the year 2050. The carbon capture proposed in this thesis would allow LKAB to cut emissions in a relatively short time frame. The carbon capture technology proposed is mature and could be implemented without invasive changes to current and future operations. However it is not possible to reach 100% CO_2 capture rate today, higher capture rates than 90% are possible but often at higher costs. Additional measures would thereby be required to eliminate all fossil emissions. One way could be through the use of biofuels in the pelletizing plants. However reducing emissions by converting to biofuels does not necessarily mean that carbon capture is irrelevant. As this thesis demonstrated, it is technically and economically viable to capture CO_2 for DRI carburization compared to carburization using biomethane. By using biogenic emission sources it can even be argued that the LKAB's processes would function as a carbon sink. A portion of the carbon in DRI would be locked in, as steel retains its carbon content over its lifetime.

While this thesis is built on the assumption of continued emissions in LKAB's pelletizing factories from which CO_2 is captured, it is not necessary for the carburization part of the thesis. LKAB has the possibility to become a carbon sink of approximately 1.4Mton CO_2 annually, assuming Kiruna, Sappavaara and Gällivare current pellet productions instead produces DRI with 2 wt% carbon content. This is in the same order of magnitude as several high CO_2 emission sources in close geographical proximity to LKAB's operations, visualised in Figure 5.1. This means that if LKABs pelletizing factories have their emissions massively reduced, carburizing DRI with CO_2 is still possible and may be viable. Carbon capture at these sources and transportation costs have not been investigated in this thesis.

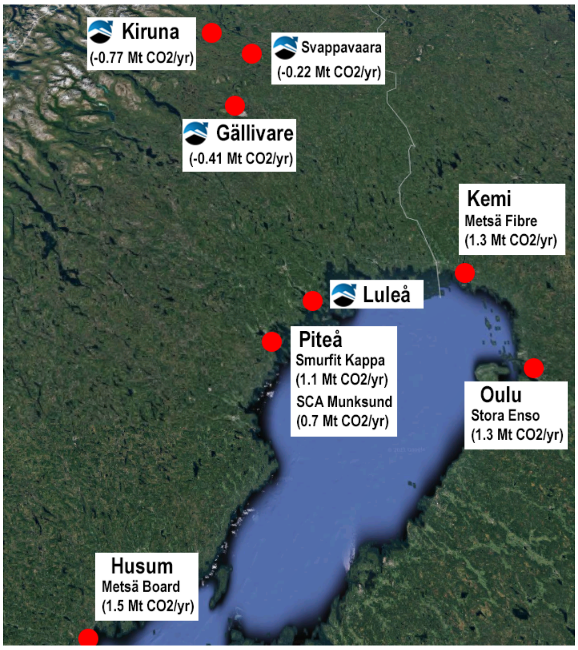

Figure 5.1: LKAB potential CO_2 consumption for 2 wt% carburization and external biogenic CO_2 emission sources in proximity of Norrbotten [12] [35].

6

Conclusion

This thesis investigates if carbon capture and utilization is a viable alternative for LKAB to reduce CO_2 emissions from pelletizing plants while simultaneously providing carbon to hydrogen DRI. The mass and energy balance show that the magnitudes of carbon in the captured CO_2 and the carbon needed in the produced DRI are compatible. The potential level of carburization for stand-alone application to LKAB's pelletizing plants are: Kiruna plants 0.92wt%, Malmberget 0.45 wt% and at Svappavaara 1.2wt%. This can be compared with the carbon content in commercial DRI (1-4.5wt%) and HBI (0.5-1.6wt%). The CO_2 capture plant modelling concluded that it is technically possible to capture the rather low CO_2 concentrations in the pelletizing plants flue gases. The cost analysis for the capture plant resulted in a specific capture cost at 56€/tonCO_2, which is equivalent to costs found in other studies of carbon capture at industrial sites. Carbon capture at LKAB is favourable in part due to that the excess heat in the pelletizing plants flue gases can supply the process.

This thesis has presented three routes for the captured CO_2. The CO route and methane route utilize CO_2 for DRI carburization. The storage route includes a long-term CO_2 storage solution, where DRI is instead carburized using biomethane. Additionally a reference case with no CO_2 capture and DRI carburization with biomethane is included. A cost assessment case for KK4 (0.92wt% carbon content in the DRI) resulted in final costs for the CO route 7.6€/tonDRI, methane route 10.6€/tonDRI, storage route 11.9€/tonDRI and biomethane reference case 8€/tonDRI. The sensitivity analysis showed that the results are highly sensitive to the price of electricity and solid biomass, and therefore are important for the economic viability of the routes. In the CO and methane route the investment in electrolysis capacity would in itself be a large investment for LKAB, if it was not for that LKAB are committed to massive investments for hydrogen reduction of pellets.

Other options to reduce pelletizing plants emissions includes electrification or fuel switch to biomass, the results shown here indicate that LKAB's pelletizing plant and future DRI production have an opportunity for carbon capture and utilization. Additionally, with several considerable biogenic CO_2 emission sources in the geographical proximity, CO_2 utilization can be a permanent solution for LKAB even if the pelletizing plants have their emissions eliminated.

6.1 Recommended Future Work

The work is a synthesis of several rapidly developing areas and the evaluation should be reassessed as the understanding of the different areas progresses. The methodology developed in this work should be a useful tool in these evaluations. The assumptions made in the necessity of carbon content in DRI, the mechanisms for DRI carburization, and projections of future electricity and biomass prices are important to the results of this work. Furthermore, the mapping and infrastructure required for utilization of external CO2 sources should be assessed in greater detail.

Bibliography

[1] Miljödepartementet, *Det klimatpolitiska ramverket*, 2017. [Online]. Available: `https://www.regeringen.se/artiklar/2017/06/det-klimatpolitiska-ramverket/`.

[2] LKAB, *LKAB in brief*, 2020. [Online]. Available: `https://www.lkab.com/en/about-lkab/lkab-in-brief/`.

[3] ——, *Climate transition will require a new fossil-free Swedish model*, 2020. [Online]. Available: `https://www.lkab.com/en/news-room/news/climate-transition-will-require-a-new-fossil-free-swedish-model/`.

[4] ——, *Processing.* [Online]. Available: `https://www.lkab.com/en/about-lkab/from-mine-to-port/processing/`.

[5] Midrex, *DIRECT FROM MIDREX 3RD QUARTER 2017*, 2017. [Online]. Available: `http://www.midrex.com/wp-content/uploads/Midrex_2017_DFM3QTR_FinalPrint.pdf`.

[6] R. Béchara, H. Hamadeh, O. Mirgaux, and F. Patisson, "Optimization of the iron ore direct reduction process through multiscale process modeling," *Materials*, vol. 11, no. 7, pp. 1–18, 2018, ISSN: 19961944. DOI: `10.3390/ma11071094`.

[7] A. Bhaskar, M. Assadi, and H. N. Somehsaraei, "Decarbonization of the iron and steel industry with direct reduction of iron ore with green hydrogen," *Energies*, vol. 13, no. 3, pp. 1–23, 2020, ISSN: 19961073. DOI: `10.3390/en13030758`.

[8] A. Jess and H. Depner, "Reduction and carburization of iron-ore in a fluidized bed by CO, H2 and CH4," vol. 69, no. 3, 1998.

[9] Hybrit development AB, "Slutrapport HYBRIT – Hydrogen Breakthrough Ironmaking Technology," 2018.

[10] International Iron Metallics Association Ltd., *Use of Hot Briquetted Iron (HBI) in the Electric Arc Furnace (EAF) for Steelmaking*, 2017.

[11] T. Ramos Ribeiro, J. Ferreira Neto, C. Takano, J. Poço, L. Kolbeinsen, and E. Ringdalen, "Ternary Diagram for Evaluation of Carbon Activity in CH4-containing gas mixtures," *Journal of Materials Research and Technology*, 2021. DOI: `https://doi.org/10.1016/j.jmrt.2021.05.033`.

[12] F. Johnsson, F. Normann, and E. Svensson, "Marginal Abatement Cost Curve of Industrial CO2 Capture and Storage – A Swedish Case Study," *Frontiers in Energy Research*, vol. 8, no. August, pp. 1–12, 2020, ISSN: 2296598X. DOI: `10.3389/fenrg.2020.00175`.

[13] Chalmers University, University of Oslo, Göteborgs University, and Tel-tek, "Carbon Capture and Storage in the Skagerrak/Kattegat region," no. Febru-

ary, 2012. [Online]. Available: http://interreg-oks.eu/webdav/files/gamla-projektbanken/se/Material/Files/Kattegat/Skagerrak/Dokumenter+projektbank/CCS%20final%20report.pdf.

[14] L. Pastor-Pérez, F. Baibars, E. Le Sache, H. Arellano-García, S. Gu, and T. R. Reina, "CO2 valorisation via Reverse Water-Gas Shift reaction using advanced Cs doped Fe-Cu/Al2O3 catalysts," *Journal of CO2 Utilization*, vol. 21, no. September, pp. 423–428, 2017, ISSN: 22129820. DOI: 10.1016/j.jcou.2017.08.009.

[15] C. Y. Chou, J. A. Loiland, and R. F. Lobo, "Reverse water-gas shift iron catalyst derived from magnetite," *Catalysts*, vol. 9, no. 9, 2019, ISSN: 20734344. DOI: 10.3390/catal9090773.

[16] M. Zhu and I. E. Wachs, "Iron-Based Catalysts for the High-Temperature Water-Gas Shift (HT-WGS) Reaction: A Review," *ACS Catalysis*, vol. 6, no. 2, pp. 722–732, 2016, ISSN: 21555435. DOI: 10.1021/acscatal.5b02594.

[17] T. Ramos Ribeiro, J. B. Ferreira Neto, J. G. Rocha Poc¨o, C. Takano, L. Kolbeinsen, and E. Ringdalen, "Steam reforming of methane on sponge iron: Influence of gas composition on reaction rate," *ISIJ International*, vol. 61, no. 1, pp. 182–189, 2021, ISSN: 09151559. DOI: 10.2355/isijinternational.ISIJINT-2019-477.

[18] P. E. Miranda, "Application of Hydrogen by Use of Chemical Reactions of Hydrogen and Carbon Dioxide," in *Science and Engineering of Hydrogen-Based Energy Technologies*, 2019, ch. 5.3.3, pp. 279–289. [Online]. Available: https://www.sciencedirect.com/science/article/pii/B9780128142516000137.

[19] M. Götz, J. Lefebvre, F. Mörs, A. McDaniel Koch, F. Graf, S. Bajohr, R. Reimert, and T. Kolb, "Renewable Power-to-Gas: A technological and economic review," *Renewable Energy*, vol. 85, pp. 1371–1390, 2016, ISSN: 18790682. DOI: 10.1016/j.renene.2015.07.066.

[20] H. Deng, S. Roussanaly, and G. Skaugen, "Techno-economic analyses of CO2 liquefaction: Impact of product pressure and impurities," *International Journal of Refrigeration*, vol. 103, pp. 301–315, 2019, ISSN: 01407007. DOI: 10.1016/j.ijrefrig.2019.04.011. [Online]. Available: https://doi.org/10.1016/j.ijrefrig.2019.04.011.

[21] H. Karjunen, T. Tynjälä, and T. Hyppänen, "A method for assessing infrastructure for CO2 utilization: A case study of Finland," *Applied Energy*, vol. 205, no. August, pp. 33–43, 2017, ISSN: 03062619. DOI: 10.1016/j.apenergy.2017.07.111. [Online]. Available: http://dx.doi.org/10.1016/j.apenergy.2017.07.111.

[22] J. Kjärstad, R. Skagestad, N. H. Eldrup, and F. Johnsson, "Ship transport—A low cost and low risk CO2 transport option in the Nordic countries," *International Journal of Greenhouse Gas Control*, vol. 54, pp. 168–184, 2016, ISSN: 17505836. DOI: 10.1016/j.ijggc.2016.08.024. [Online]. Available: http://dx.doi.org/10.1016/j.ijggc.2016.08.024.

[23] Norwegian Ministry of Petroleum and Energy, "Longship-Carbon capture and storage," vol. 33, 2020.

[24] *What we do - Northern Lights*. [Online]. Available: https://northernlightsccs.com/what-we-do/.

[25] R. Skagestad, H. A. Haugen, and A. Mathisen, "CCS case synthesis Final Report," no. October, 2015.

[26] S. Ó. d Signarsdóttir, F. Normann, K. Andersson, and F. Johnsson, "Postcombustion CO2 capture using monoethanolamine and ammonia solvents: The influence of CO2 concentration on technical performance," *Industrial and Engineering Chemistry Research*, vol. 54, no. 2, pp. 681–690, 2015, ISSN: 15205045. DOI: `10.1021/ie503852m`.

[27] Å. Eliasson and E. Fahrman, "Utilization of Industrial Excess Heat for CO 2 Capture Effects on Capture Process Design and District Heating Supply," 2020. [Online]. Available: `https://hdl.handle.net/20.500.12380/300819`.

[28] TCM, "A summary of published papers and reports on four MEA Campaigns," [Online]. Available: `www.tcmda.com%0Awww.catchingourfuture.com%20%7C`.

[29] H. Ali, N. H. Eldrup, F. Normann, R. Skagestad, and L. E. Øi, "Cost Estimation of CO2 Absorption Plants for CO2 Mitigation – Method and Assumptions," *International Journal of Greenhouse Gas Control*, vol. 88, no. December 2018, pp. 10–23, 2019, ISSN: 17505836. DOI: `10.1016/j.ijggc.2019.05.028`. [Online]. Available: `https://doi.org/10.1016/j.ijggc.2019.05.028`.

[30] A. Krüger, J. Andersson, S. Grönkvist, and A. Cornell, "Integration of water electrolysis for fossil-free steel production," *International Journal of Hydrogen Energy*, vol. 45, no. 55, pp. 29 966–29 977, 2020, ISSN: 03603199. DOI: `10.1016/j.ijhydene.2020.08.116`.

[31] L. Göransson, M. Lehtveer, E. Nyholm, M. Taljegard, and V. Walter, "The benefit of collaboration in the North European electricity system transition—System and sector perspectives," *Energies*, vol. 12, no. 24, 2019, ISSN: 19961073. DOI: `10.3390/en12244648`.

[32] H. Thunman, C. Gustavsson, A. Larsson, I. Gunnarsson, and F. Tengberg, "Economic assessment of advanced biofuel production via gasification using cost data from the GoBiGas plant," *Energy Science and Engineering*, vol. 7, no. 1, pp. 217–229, 2019, ISSN: 20500505. DOI: `10.1002/ese3.271`.

[33] G. Sandin, T. S. Zetterberg, and T. Rydberg, "Tillgång på skogsråvara – sammanfattning och scenarier," IVL Svenska Miljöinstitutet, Tech. Rep. C, 2019.

[34] M. van der Spek, S. Roussanaly, and E. S. Rubin, "Best practices and recent advances in CCS cost engineering and economic analysis," *International Journal of Greenhouse Gas Control*, vol. 83, no. February, pp. 91–104, 2019, ISSN: 17505836. DOI: `10.1016/j.ijggc.2019.02.006`. [Online]. Available: `https://doi.org/10.1016/j.ijggc.2019.02.006`.

[35] S. Teir, E. Tsupari, A. Arasto, T. Koljonen, J. Kärki, A. Lehtilä, L. Kujanpää, S. Aatos, and M. Nieminen, "Prospects for application of CCS in Finland," *Energy Procedia*, vol. 4, no. December 2011, pp. 6174–6181, 2011, ISSN: 18766102. DOI: `10.1016/j.egypro.2011.02.628`.

A

Absorber Dimensions

The modelling of the Carbon capture plant had previously been used to capture flue gases with significantly higher CO_2 concentrations about 7-20%vol. The modelling used stages as in trayed columns but in reality the carbon capture plant uses packed columns. The number of stages in the absorber in the initial model was 30 stages with a stage distance of 0.6m adding up to a total packing height of 18m, this is represented by the blue line in Figure A.1. The initial result with a optimum energy demand of $5.08 MJ/kgCO_2$ was deemed high and to decrease heat consumption the height of the absorber column was altered. The analysis resulted in a reduced energy demand and increased optimal lean loading with a taller absorber, visualised in Figure A.1. The best result was the 45 stages absorber equivalent of a stacked packing of 27m. A taller absorber gave diminishing returns in energy savings with linearly increased investment costs.

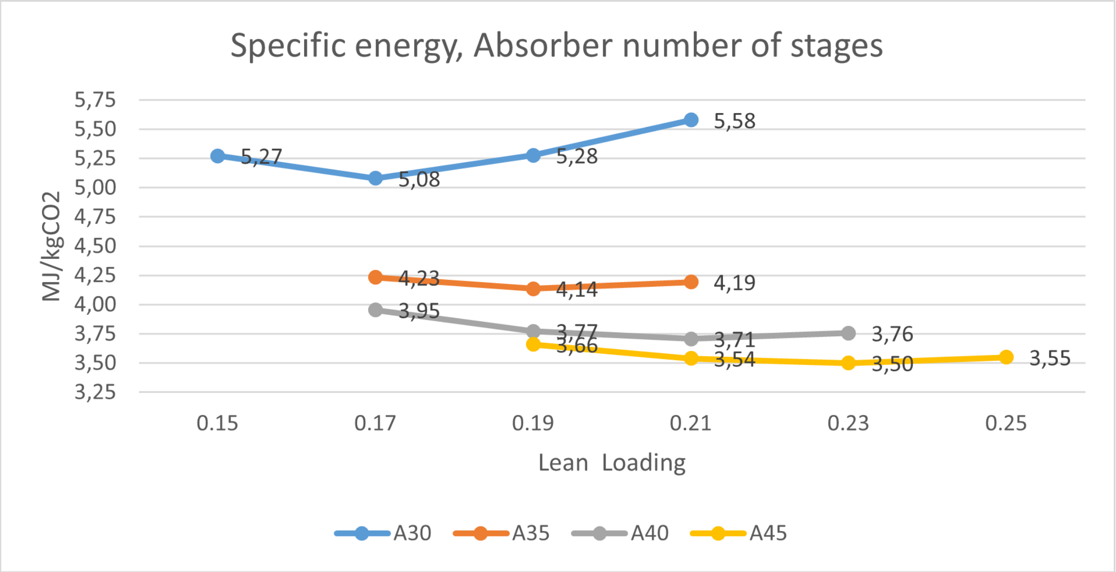

Figure A.1: Energy consumption of the desorber for different absorber heights and lean loading.

By increasing absorber height the MEA residence time increases, and therefore captures more CO_2 per unit MEA leading to increased rich loading. The increased rich loading leads to reduces energy regeneration and heat consumption in the desorber. A higher absorber comes with a higher investment cost but other components can be smaller, eg. the heat exchanger in the reboiler. The lower heat demand also means lower steam consumption which results in lower OPEX. The total cost of the system will therefore be similar with a higher absorber but with significantly lower heat demand. Due to this the absorber was set to 45 stages and 27m for the carbon capture modelling.

DEPARTMENT OF SOME SUBJECT OR TECHNOLOGY
CHALMERS UNIVERSITY OF TECHNOLOGY
Gothenburg, Sweden
www.chalmers.se