THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Partial carbon capture – an opportunity to decarbonize primary steelmaking

A techno-economic assessment of amine absorption of carbon dioxide at an integrated steel mill

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Abstract

Climate change requires that all energy-related sectors drastically reduce their greenhouse gas emissions (GHG). To have a high likelihood of limiting global warming to 1.5°C, large-scale mitigation of GHG has to start being implemented and cause emissions to fall well before Year 2030. The process industry, including the iron and steel industry, is inherently carbon-intensive and carbon capture and storage (CCS) is one of the few options available to achieve the required reductions in carbon dioxide (CO_2) emissions. Despite its high technological maturity, CCS is not being implemented at the expected rates due *inter alia* to the low value creation of CCS for process industries, which is often attributed to uncertainties related to carbon pricing and the considerable investments required in CO_2 capture.

This thesis deals with the concept of partial carbon capture, which is governed by market or site conditions and aims to capture a smaller fraction of the CO_2 emissions from an industrial site, thereby lowering the absolute and specific costs (€ per tonne CO_2) for CO_2 capture, as compared to a conventional full-capture process. Depending on the scale and market conditions these savings hold true especially for a process industry that has large gas flows with concentrations of $CO_2 \geq 20$ vol.% and access to low-value heat. Integrated steel mills typically fulfill these conditions.

The value of partial capture for the steel industry is assessed in a techno-economic study on the separation of CO_2 from the most carbon-intensive steel mill off-gases. The design for partial carbon capture using a 30 wt.% aqueous monoethanolamine (MEA) solvent is optimized for lower cost. Powering the capture process exclusively with excess heat entails a cost of 28–35 (±4) €/tonne CO_2-captured and a reduction in CO_2 emissions of 19%– 43% onsite, depending on design and CO_2 source. In contrast, full capture requires external energy to reduce the CO_2 site emissions by 76%, entailing costs in the range of 39–54 (±5) €/tonne CO_2-captured. Furthermore, the use of excess heat has impacts on the cost structure of partial carbon capture, i.e., increasing the ratio of capital expenditures to operational expenditures, as well as on the relationship between carbon and energy intensity for primary steel as an industrial product.

The present work concludes that near-term implementation of partial carbon capture in the 2020s will be economically sustainable if average carbon prices are in the range of 40–60 €/tonne CO_2 over the entire economic life-time of the partial capture unit (ca. 25 years). Once implemented, partial capture could evolve to full capture over time through either co-mitigation (e.g., with biomass utilization or electrification) or efficiency improvements. Alternatively, partial capture could act as a bridging-technology for new, carbon-free production. In summary, partial carbon capture is found to be readily available and potentially economically viable to initiate large-scale mitigation before Year 2030. Partial capture may represent a starting point for the transition to the carbon-constrained economies of the future in line with the 1.5°C target.

Keywords: Partial CO_2 capture, process industry, steel making, amine absorption, excess heat, CCS, cost estimation

List of publications

This thesis is based on the following papers, which are referred to in the thesis by their Roman numerals:

I. Biermann, M.; Normann, F.; Johnsson, F.; Skagestad, R. *Partial Carbon Capture by Absorption Cycle for Reduced Specific Capture Cost. Ind. Eng. Chem. Res.* **2018**

II. Sundqvist, M.; Biermann, M.; Normann, F.; Larsson, M.; Nilsson, L. *Evaluation of Low and High Level Integration Options for Carbon Capture at an Integrated Iron and Steel Mill. Int. J. Greenh. Gas Control* **2018**.

III. Biermann, M.; Ali, H.; Sundqvist, M.; Larsson, M.; Normann, F.; Johnsson, F. *Excess-Heat Driven Carbon Capture at an Integrated Steel Mill – Considerations for Capture Cost Optimization. Submitted for Publication.* **2019**.

In addition, **Paper A** is included in the Appendix:

A. Martinez Castilla, G.; Biermann, M.; Montañés, R. M.; Normann, F.; Johnsson, F. *Integrating Carbon Capture into an Industrial Combined-Heat-and-Power Plant: Performance with Hourly and Seasonal Load Changes. Int. J. Greenh. Gas Control* **2019**, *82*, 192–203.

Authors' contributions

Maximilian Biermann is the principal author of **Papers I** and **III**. As second author, he has contributed to Paper II with modeling, data processing, writing, discussions and editing and to **Paper A** with steady-state process modeling, data interpretation, discussions and editing. Associate Professor Fredrik Normann has contributed with discussions and editing to all four papers. Professor Filip Johnsson has contributed with discussions and editing to **Papers I**, **III** and **A**. Maria Sundqvist is the principal author of **Paper II** and has contributed to **Paper III** with modeling and discussions, mainly regarding the performance of the steel mill. Hassan Ali has contributed to **Paper III** with the techo-economic assessment and discussion of the steam generation cost. Ragnhild Skagestad has performed the underlying cost calculations for **Paper I** and **III**. Dr. Mikael Larsson has contributed with discussions and editing to **Paper III**. Leif Nilsson has contributed with discussions to **Paper II**. Guillermo Martinez Castilla is the principal author of **Paper A**. Dr. Rubén Mocholí Montañés has contributed to **Paper A** with model development, discussions and editing.

Additional work related to the topic has been carried out, resulting in the publications listed below. These have not been included in the thesis because they are either outside the scope of the thesis or overlap with the appended papers.

- Biermann, M.; Alamia, A.; Normann, F.; Johnsson, F. *Evaluation of Steel Mills as Carbon Sinks*. *International Conference on Negative Emissions*; Chalmers University of Technology: Gothenburg, **2018**
- Skagestad, R.; Sundqvist, M.; Biermann, M. Webinar: *Cutting Cost of CO2 Capture in Process Industry (CO2stCap) Project Overview & First Results for Partial CO2 Capture at Integrated Steelworks* GCCSI **2017.** http://www.decarboni.se/insights/webinar-cutting-cost-co2-capture-process-industry-co2stcap-project-overview-first-results-partial-co2-capture-integrated-steelworks

Acknowledgments

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List of abbreviations

BAT	Best available technology
BECCS	Bio-energy CCS
BF	Blast furnace
BFG	Blast furnace gas
BOF	Basic oxygen furnace
BOFG	Basic oxygen furnace gas
CAPEX	Capital expenditure
CCS	Carbon capture and storage
CDA	Carbon direct avoidance
CDQ	Coke dry quenching
CHP	Combined heat and power
COG	Coke oven gas
CS	Crude steel
DRI	Direct reduced iron
DSG	Dry slag granulation
EAF	Electric arc furnace
EU ETS	EU emissions trading system
EUA	European Union Allowance
EW	Electrowinning
FGHR	Flue gas heat recovery
GHG	Greenhouse gas emissions
H-DR	Hydrogen direct reduction
ICA	Intercooled absorber
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
MEA	Monoethanolamine
NG	Natural gas
OPEX	Operational expenditure
POX	Partial oxidation
PV	Photovoltaic
RSS	Rich solvent splitting
SEWGS	Sorption-enhanced water-gas shift
SR	Smelting reduction
SRP	Separation rate path
SSP	Split stream path
TGRBF	Top gas recycling blast furnace
TRL	Technology readiness level
WGS	Water-gas shift
VPSA	Vacuum pressure-swing adsorption

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1 Introduction

Climate change is one of the major global environmental challenges of the 21st Century. In the Paris Agreement the world's nations have agreed to limit the global temperature rise to well below 2°C above the pre-industrial level[1]. In their recent special report SR15, the Intergovernmental Panel on Climate Change (IPCC) motivates that humanity should further limit the global temperature rise to 1.5°C[2], so as to minimize the impacts of climate change, including the loss of human life. The remaining carbon budget for 1.5°C, estimated at 420–770 $GtCO_2$[2], is currently being depleted at a rate of 42±3 $GtCO_2$ per annum[2], with the depletion rate still rising[3,4]. Therefore, emissions have to fall significantly before Year 2030 to restrict global warming to 1.5°C[2], hence the urgency for large-scale mitigation. Fortunately, the electricity sector is showing a positive trend with increasing shares of renewable electricity generation, especially from solar photovoltaic (PV) but also from off-shore wind, at costs (especially for solar PV) that are much lower than previous expectations[5]. The industrial sector is, however, not 'on-track' and requires attention[5]. Together, the cement, petrochemical, pulp and paper, and iron and steel industries account for ca. 19%[6] of the global energy-related CO_2 emissions. These emissions are more difficult to mitigate as the use of carbon is inherent to the manufacturing process. Nevertheless, the process industries including the steel industry, should align to the reduction targets of greenhouse gas (GHG) emissions for industry, which in the EU are 34%–40% by Year 2030 and 83%–87% by Year 2050, as compared to Year 1990[7,8].

Carbon capture and storage (CCS) can play significant roles in decarbonizing industry and addressing the need for large-scale and timely mitigation. Carbon capture entails the separation of CO_2 from a CO_2-rich gas, which is compressed for transport by ship or pipeline to (preferably) off-shore geologic storage sites, such as saline aquifers or depleted oil fields. CCS is: 1) capable of reducing emissions at scale and is expected to mitigate a considerable share of the cumulative emissions[9]. It presents, therefore, a crucial technology in most emission pathways that are consistent with the 1.5°C[2] or 2°C[10] target; and 2) Concerning the above mentioned timeliness for climate mitigation, CCS is, readily available at commercial scale when using amine absorption processes for capture and storage in saline formations[11], and is, thus, implementable today.

However, cross-sector deployment of CCS is lagging[10]. This is due to a number of reasons[12], such as a lack of binding policies, legal issues related to cross-border CO_2 transport with storage intention (London Protocol), public acceptance, and, perhaps most importantly, the low value creation of CCS under present market conditions. As an example, the iron and steel industry has not applied CCS despite the fact that integrated steel mills fulfill the prerequisites for low cost for capture, such as large gas flows with high concentrations of CO_2. With estimated costs of 42–100 €$_{2015}$[13–20] per tonne CO_2-avoided for CO_2 capture from steel mill off-gases and considering that CO_2 emissions allowances in the EU (EUA) have been traded at around 10 € per tonne CO_2 for most of the time that the market has been in place[21], the value creation of CCS has been probably too low for steelmakers who face severe global competition, trade tariffs, low profit margins, and long investment cycles[22]. Mitigation options that match

investment cycles with the support of long-term policies, e.g., mechanisms that allow steelmakers to pass on costs for low-carbon technology to the end-consumer[23], may facilitate large-scale CO_2 mitigation in the steel industry in an economically sustainable manner.

In light of the urgency regarding a fast-shrinking carbon budget and the need for economic sustainability, the present thesis addresses the challenge of initiating large-scale, near-term mitigation in the process industry using the steel industry as an example. More specifically, the work evaluates partial carbon capture as a means to reduce the capture cost, and thereby lower the hurdles for CCS deployment. Partial capture is here defined as a CCS concept, in which only a fraction of the accessible CO_2 is separated from a CO_2-rich gas. The magnitude of this fraction is determined by economic factors, such as energy prices, and policy-driven requirements, such as the Emission Performance Standards. Partial capture comes with a reduced absolute energy penalty and reduced absolute capital expenditures, which reduce the investment risks[24,25], as compared to CCS with a so-called *full capture* approach. Full capture represents almost-maximized separation rates (e.g., 90%) of CO_2 from CO_2-rich gases, so as to minimize the specific capital expenditures per tonne CO_2-captured through economy of scale. To be clear, partial capture aims to reduce the total specific cost, i.e., capital and operating expenditures, as compared to full capture. Overall, partial capture is evaluated as a first step towards decarbonization of the process industry.

1.1 Aims and scope

The overarching aim of this thesis is to support a rapid and sustainable transition of the carbon-intensive industries to operation within a carbon-constrained society. The focus is on investigating the technical dependencies between the carbon-intensity and energy-intensity of the industrial product. More specifically, this thesis aims to:

i. Contribute to the cost-effective design of amine absorption cycles for partial capture of CO_2 from industrial processes that have large gas flows with high concentrations of CO_2;

ii. Evaluate the relationships between cost, energy consumption, and carbon capture rates of CCS in primary steelmaking that uses blast and basic oxygen furnaces,

iii. Assess the near-term implementation of partial capture in primary steel making; and

iv. Construct an overall perspective on partial capture in synergy with and in the transition to other mitigation options for the steel industry over time.

1.2 Outline of the thesis

This thesis comprises a summary essay and four appended papers. The seven chapters of the essay describe and contextualize the key findings of the papers. Chapter 2 gives the background to the work by reviewing mitigation options for primary steelmaking in terms of emissions intensity and technology readiness levels. Chapter 3 gives an overview of the applied methods. The outcomes of the work are presented in chapters 4 and 5. Chapter 4 describes the concept of partial capture and the design of amine absorption cycles for partial capture from process

industry. Chapter 5 summarizes the findings of the techno-economic assessment of partial capture applied to a reference integrated steel mill, and lists the implications of partial capture for the carbon- and energy-intensity of the produced steel. Chapter 6 discusses partial capture as a near-term mitigation option for the steel industry in terms of economic viability with respect to lifetime and possible synergies with other mitigation options. Chapter 7 concludes the essay and provides an outlook on future work in this area. The focuses of the appended papers are briefly described below. Their relationships are illustrated in Figure 1-1.

In **Paper I,** two design paths for partial capture are described, modeled, and evaluated based on their energy demand and capture cost in relation to the amount of CO_2 separated from a CO_2-rich gas, i.e., assuming high concentrations of CO_2 of around 20 vol.% , which is typical for process industries, such as pulp and paper, cement, petroleum refining, and iron and steel production. This paper focuses on the design of amine absorption cycles with regard to scale, CO_2 concentration, and CO_2 separation rate in the absorber.

Paper II assesses the amount of available excess heat as the yearly average in a reference integrated steel mill for the purpose of powering partial CO_2 capture from either a blast furnace gas or CHP plant flue gases. The paper emphasizes the different levels of integration of CCS within the steel mill and compares the levels of CO_2 capture that are achievable, using the designs from **Paper I,** from these two sources depending upon the amount of retrieved heat.

Paper III extends the technical assessment made in **Paper II** to include a third CO_2 source and to include the economic dimension as a criterion for performance. Full capture from all three CO_2 sources is compared to the best-performing partial capture scenarios. In addition, the paper incorporates the full-chain cost for partial capture, including transport and storage, into a relation with carbon price projections to assess the conditions for near-term implementation of partial capture in the steel industry.

Paper A investigates the assumption made for the yearly averaged excess heat in **Papers II** and **III** and illustrates the dynamic performances of partial capture with varying heat loads to the reboiler and feed gas flows to the absorber.
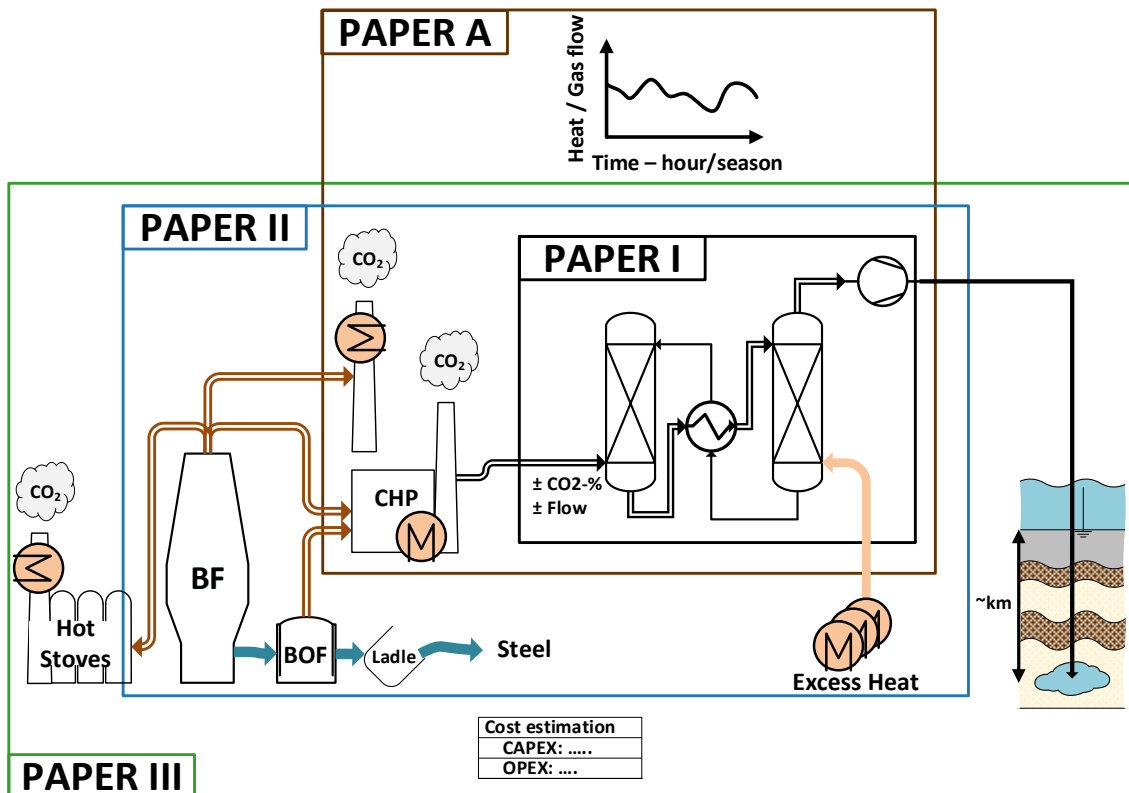
Figure 1-1: Overview of the topics covered and the linkages between the papers appended to this thesis.

2 Mitigation options for the steel industry

Numerous mitigation measures for the steel industry have been discussed in the literature. Here, an overview of the main low-carbon technologies for primary steel production is given with a focus on CO₂ intensity and technological readiness. Figure 2-1 illustrates the typical routes of steel production. In primary steelmaking, virgin iron ore is reduced with carbon to form metallic iron in blast furnaces. The hot metal is then refined to steel in a basic oxygen furnaces (BOF). In secondary steelmaking, predominantly recycled scrap steel is melted down and refined using electric power. Around 72% of the global crude steel (CS) is produced in the BOF, whereas 28% of the global crude steel is produced in electric furnaces[26]. Despite the increasing amounts of available scrap[27], substantial levels of primary steelmaking will be required throughout the 21st century on a global level, due to: 1) an expected increase in global steel demand as developing countries build up their steel stock[28,29]; 2) the longevity of blast furnaces, with lifetimes of 40–60 years[30]; and 3) the purity demands of high-quality steel[29,30]. Therefore, it is, not a viable option simply to replace all primary steelmaking with secondary steelmaking, but rather necessary to enable primary steelmaking to align with the required carbon intensity.

This overview is confined to primary steelmaking and the permanent storage of CO₂ in the case of carbon-capturing technologies, and therefore excludes carbon capture and usage. Estimates at technology readiness levels (TRLs) and CO₂ emission intensities for primary steelmaking are listed in Table 2-1. It should be emphasized that the outcomes of this thesis focus on amine absorption, which is at TRL9 and is, thus, the technology used for the partial capture approach, as described in chapter 4.

Figure 2-1: Categorization and overview of the typical routes of steelmaking highlighting their reliance on carbon as fuel and reducing agent.

Table 2-1: Mitigation options for production routes of primary steelmaking including CO_2 intensity and technology readiness levels (TRLs). See[11,31] for a description of the TRLs.

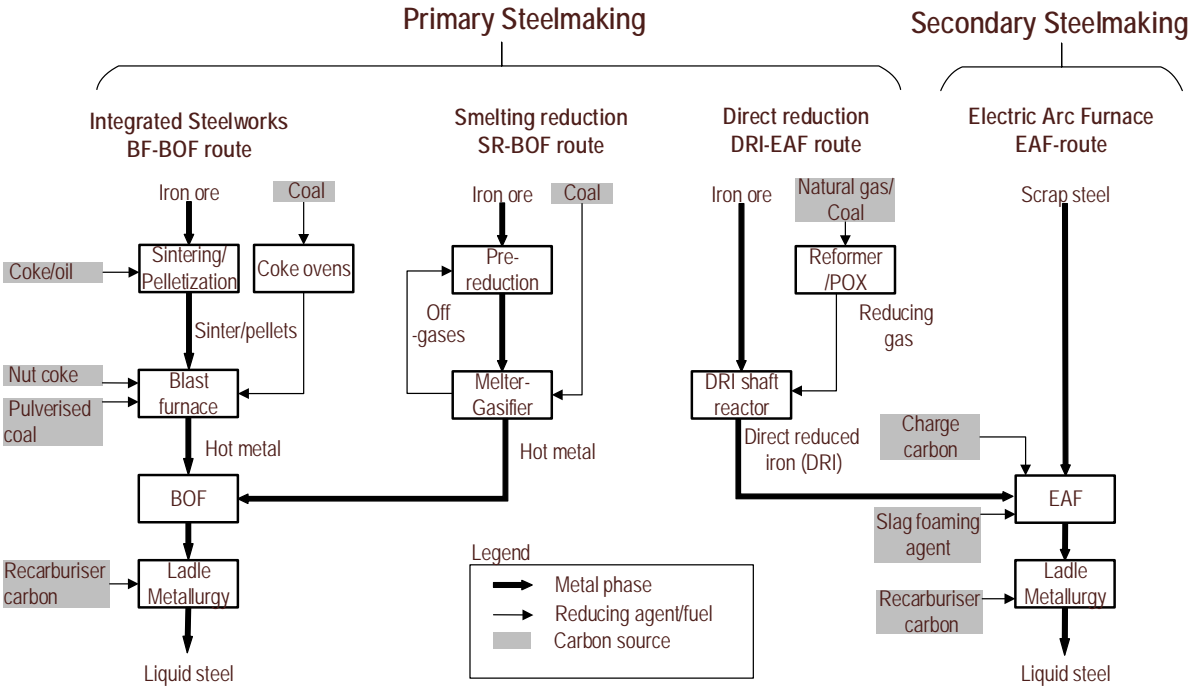| Technology	Status	Reducing agent/fuel	CO_2 intensity kg CO_2/ tonne CS [a]	Corresponding avoided emissions % [a]	Reference
BF-BOF route	Commercial	Coke/Coal	1600[b]–2200 EU: ~1880	-	32
TGRBF	TRL 7		1560–1670	20–25%	33
TGRBF+CCS	TRL 7		920–1360	45–60%	16,33
Amine Absorption	TRL 9[11]		300–1400	19–80 %	16,20,34
SEWGS	TRL 3–4[35], TRL 6[36,37]		500–1300	35–75%	36
SR-BOF route	Commercial	Coal	~2250		16
Amine absorption	TRL 9[11]		~1600	30%	16
HIsarna	TRL 7[35]		1200–1500	20–35%[c]	35,38
HIsarna+CCS			~400	80%[c]	35,38
DRI-EAF route	Commercial	Natural gas/	630–1500		33,39,40
with CCS	TRL 9	Coal/ Electricity	300–1200	25–50 %	33,41
CDA					
H-DR	TRL 1–4[35]	Hydrogen[d] /Electricity	~25	26–95%[c]	35,40
EW	TRL 4-5[35]	Electricity	~240 [e]	87%[c]	42

[a] Note that the baselines for the different references vary, so the comparability of references may, thus, not be provided.

[b] BAT.

[c] Compared to BF-BOF route.

[d] From electrolysis with 100 % renewables.

[e] Calculated: assuming 87% [42] reduction of 1880 kg/t CS [32].

Blast furnace route (BF-BOF). The majority of primary steel is produced in large-scale integrated steelworks, where coal and coke are used to reduce the iron ore and smelt the formed iron in the blast furnace (BF). The produced pig iron is converted to steel in oxygen-blown furnaces (BOF) to reduce the carbon content of the steel. The carbon that is used forms CO_2 and CO with oxygen originating mostly from the iron ore itself and ending up in the off-gases, i.e., blast furnace gas (BFG) and basic oxygen furnace gas (BOFG). Together with coke oven gas (COG), these gases are used as heating gases in, amongst others, the combined heat and power (CHP) boilers, and the hot stoves, which supply the blast furnace with hot air. It is important to note that the BFG alone contains around 70% of the carbon emitted from the site.

The CO_2 emission intensities of blast furnace processes that apply the BAT in Europe have reached levels close to those that are technically and theoretically possible[39]. Many studies have, therefore, evaluated CCS as an option for removal of CO_2 from the steel mill off-gases[13,15,17,34,43]. In summary, those studies have reported on 50%–80% CO_2 avoidance if the CO_2 is captured from the largest direct emission points onsite (stacks of CHPs, hot stoves, lime kilns, sinter plants, coke ovens), depending on the number of stacks included. Applying amine absorption to capture CO_2 from BFG alone could reduce emissions by 19%–30%[13,16]. The use

of different capture techniques, such as physical solvents (e.g., Selexol) or membranes could achieve similar emission reductions for CO_2 capture from BFG in common, air-blown BFs[16]. However, leading BFG into a water-gas shift (WGS) reactor, so as to shift the CO to CO_2 and H_2 in combination with CO_2 capture, achieves higher reductions in CO_2 emissions because the yield of CO_2 in the BFG is enhanced[16,37]. Similar emissions reductions can be achieved through the sorption-enhanced water-gas shift (SEWGS) technology developed by ECN[44], in which CO_2 is adsorbed simultaneously in a WGS reactor. SEWGS is currently being tested in the STEPWISE project. A prominent technology proposed by the ULCOS[45] consortium is called Top Gas Recycling Blast Furnace (TGRBF), involves a switch from air-blown to oxygen-blown BF and recirculation of the top gas into the BF as a reducing gas. TGRBF thereby decreases coal consumption and, consequently, the levels of CO_2 emissions, as compared to a common BF. The separation of CO_2 from the recycled top gas through amine absorption or vacuum-PSA (VPSA) could further boost CO_2 avoidance, as shown inTable 2-1.

Apart from CCS, the introduction of biomass as a source of biogenic carbon has been assessed and could theoretically deliver a 38%–55% reduction in emissions[46,47]. In addition to the practical limitations and the biomass supply, a major restriction is the mechanical strength of coke required to support the burden in large blast furnaces and to maintain gas permeability. The reader is referred to publications on the potential for bio-energy CCS (BECCS) in the steel industry[48,49]. Moreover, slag carbonation could be applied, although it achieves rather modest reductions in emissions of 8–20%[20].

Smelting reduction route (SR-BOF). In smelting reduction (SR), hot metal is produced in a similar way as in the blast furnace, i.e. a reduced, molten iron phase is produced and then refined to steel. However, SR does not require pre-treatment of the iron ore and onsite coke production. The iron production takes place in a two-stage process: first, the iron ore is pre-reduced in a shaft reactor using off-gases from the second stage, a smelter-gasifier, in which the final reduction and melting are achieved. The considerable amount of surplus off-gas is commonly used for heat and power production. Commercially available technologies at medium scale include COREX, using pellets or lump ore, and FINEX, using fine ore for pre-reduction in fluidized beds[33]. These are operated in South Africa, South Korea, China, and India – no SR plants currently exist in Europe. According to Eurofer[32], typical SR-BOF plants have a higher emission intensity than BF-BOF, depending on how the off-gases are used.

CCS for SR-BOF may reach similar CO_2 intensities per tonne of steel as does BF-BOF when using amine absorption or Selexol[16] (*cf.* Table 2-1). However, the application of WGS combined with CO_2 capture could reduce substantially the emissions from COREX compared to not applying any capture[16]. The ULCOS consortium has developed the HIsarna[50] process, in which a smelt cyclone for pre-reduction is placed on top of a coal-fed smelter within a single unit. It operates with pure oxygen, making CCS comparatively easy to integrate. Compared to the BF-BOF route, HIsarna is expected to have a lower carbon intensity and could reduce the most of the emissions when combined with CCS (*cf.* Table 2-1). HIsarna has been tested in pilot scale with a demonstration expected in The Netherlands in the period 2020–2025[35], and it is expected to be commercially available in 2030–2035[30].

Direct reduction route using electric arc furnaces (DRI-EAF). Direct reduced iron (DRI) or sponge iron is produced in a reducing gas atmosphere, commonly in the form of reformed natural gas, at a temperature below the melting point of iron. The solid, porous product has a high degree of metallization (similar to pig iron), yet it still contains gangue, which has to be removed in a subsequent melting process, usually in electric arc furnaces (EAF). If it is not immediately processed, DRI is prone to re-oxidation (pyrophoricity) due to its high specific surface area, and it has to be passivated for transport and storage. The share of DRI in global steel production is about 4%–6%[26,33,51], with most plants being located in regions with access to natural gas and low prices for electricity, e.g., North America, India, and Iran. There are only few DRI-plants located in Europe. The commercial processes that are most frequently applied are MIDREX and HYL/Energiron. The reported CO_2 intensity of the produced steel varies substantially due to differences in the charging (hot, cold) of DRI into the EAF, regional differences in the CO_2 intensity of the power grid, and the amount of scrap that is co-fed to the EAF. In general, DRI-EAF steel is associated with lower emissions than steel produced by BF-BOF, see Table 2-1. Since CO_2 removal is inherent to the current DRI processes, CCS is comparatively simple. The ULCOS consortium has developed ULCORED[52], a process that uses the syngas from partial oxidation (POX) of natural gas or coal, which could remove half of the emitted CO_2 from the direct reduction of iron ore[33,45].

Carbon direct avoidance. In addition to CO_2 removal or the replacement of fossil carbon with biogenic carbon, technologies have been proposed that avoid the usage of carbon in primary steelmaking. Such carbon direct avoidance (CDA) approaches involve hydrogen direct reduction (H-DR) and electrowinning (EW). The H-DR technology uses hydrogen as the reducing gas for DRI production in a shaft furnace, together with subsequent refinement of the DRI in an EAF[53]. Preferably, from the carbon intensity perspective, the hydrogen is generated by water electrolysis using renewable electricity. Compared to the commonly used DRI-EAF with syngas from natural gas or coal, hydrogen possesses a higher reduction potential, although it reacts endothermically with iron ore. This means that more heat has to be supplied for H-DR than is the case for the usual mode of CO-based reduction, which is exothermic. Three H-DR projects are currently under development in Europe: SALCOS[54] (Salzgitter), SUSTEEL[55] (Voestalpine), and HYBRIT[56] (SSAB, LKAB, Vattenfall). Both ThyssenKrupp[57] and ArcelorMittal[58] have announced their engagement in H-DR development at their Duisburg and Hamburg sites, respectively. H-DR is potentially close to CO_2-free[59] (*cf.* Table 2-1), and may offer flexible production and intermediate storage of hydrogen and DRI, which could be useful in balancing the power loads in an electricity system that is based on intermittent renewable energy[40,53]. The key obstacles to H-DR are its reliance on low-cost renewable electricity, the scalability of the involved technologies[35] (e.g., hydrogen storage, water electrolysis, direct reduction with hydrogen), and uncertain economic viability. If all steel were to be produced through H-DR using hydrogen generated with electricity, the Swedish, Austrian and German power demands would increase by 15 TWh (ca. +10% of present demand)[40], 33 TWh (+47% of present demand)[60], and 237 TWh (+120% of present demand)[59], respectively. Fischedick et al.[61] have estimated the marketability of H-DR as coming into being by Year 2030 or Year 2040 depending on the market conditions. Electrowinning, which has also been developed by the

ULCOS consortium (ULCOWIN, now SIDERWIN), is based on the electrolysis of iron ore fines in aqueous alkaline solutions[42]. It is potentially CO_2-free[62] if run on renewable electricity and is applicable to small-scale decentralized steel production[35]. The market entry of EW is not expected before Year 2040[61].

3 Methodology

This work investigates the interactions between the design and operational factors of a carbon capture unit and a steel mill through modeling. Figure 3-1 illustrates how the carbon capture model interacts with models of the steel mill and the cost estimation. The main part of the work is in the modeling of the carbon capture unit, which is based on CO_2 absorption using a 30 wt.% aqueous monoethanolamine (MEA) solution. The CO_2 absorption model is used in **Paper I** to design a partial capture process that is suitable for a generic case. Thus, as indicated above, the results from this study should be applicable to several process industries and not only to the steel industry. Building on the findings from **Paper I**, the CO_2 absorption process for partial capture is integrated with the reference steel mill, considering the different levels of available excess heat (**Papers II** and **III**), to quantify the efficiency of partial CO_2 capture from the steel mill off-gases. In **Paper A**, the absorption model is used in a study of the dynamic interactions of the capture unit and the steel mill, to consider the effects of plant operation.
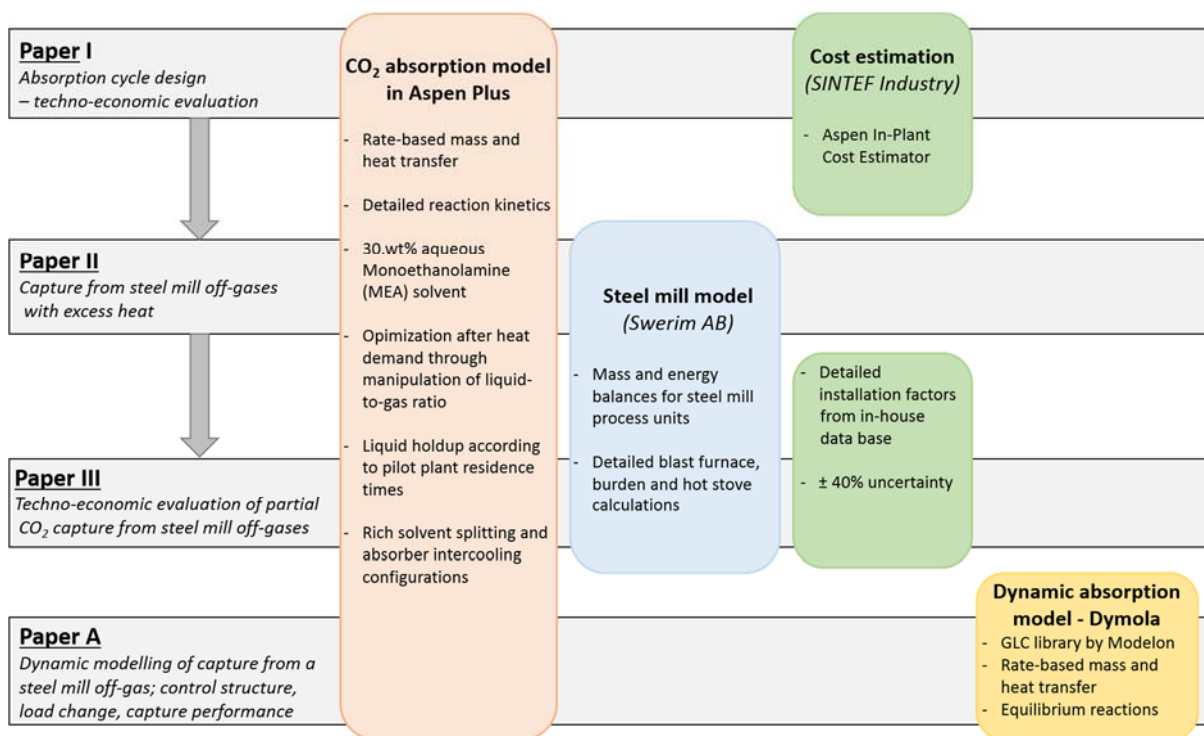

Figure 3-1: Overview of the relationships between the modeling tools applied in this thesis and their linkages to the appended papers.

3.1 Process modeling of partial capture

*The modeling of CO₂ absorption applied to partial capture is described – first for a steady-state model used as fundamental tool in **Papers I-III**, followed by a dynamic model used in **Paper A**.*

3.1.1 The CO₂ absorption model

Figure 3-2a is a schematic of the modeled CO_2 absorption process in the so-called *standard configuration*. The CO_2-rich gas is brought into contact with the liquid absorbent in a structured-packed column, the absorber, where CO_2 is absorbed into the liquid phase. The CO_2-lean gas stream is vented, whereas the CO_2-rich liquid enters the desorber (or stripper). In the desorber, the CO_2 is released by increasing the temperature (to around 120°C) and the solvent is regenerated. The warm CO_2-lean solvent is circulated back to the absorber via a cross-heat exchanger and cooler. The pure CO_2 stream exiting the top of the stripper is compressed for transport and storage.

The process is modeled in the Aspen Plus ver. 8.8 software and based on the built-in property method ELECNRTL used to estimate the properties of the aqueous MEA solution. ELECNTRL is based on the Redlich-Kwong equation of state for gas properties combined with the nonrandom two-liquid (NRTL) activity coefficient model for electrolytes in the liquid phase. Analogous to the work of Garðarsdóttir et al.[63], the model considers reaction rate constants for relevant reactions in the chemical absorption of CO_2 with MEA. The absorber and stripper columns are modeled by estimating the mass transfer rates between the liquid and gas phases using the two-film theory. Since the gas absorption rate is limited on the liquid side, the liquid film is discretized to consider both reactions in the liquid film and the mass transfer resistance[64]. The structured packing in the columns is sized using correlations for mass transfer coefficients, interfacial area, and liquid-hold up, as described by Bravo et al.[65,66]. The heat transfer coefficients are derived from the calculated mass transfer coefficients using the Chilton and Coburn analogy.

All equipment is simulated in design mode, i.e., it is sized to a targeted capture rate. The process is optimized towards minimum specific heat demand by varying the liquid-to-gas ratio at a targeted capture rate. A full capture reference is designed with liquid hold-up (residence times) in line with those of reported pilot[67] and full-scale[68,69] plants. The partial capture designs are derived from the full capture design, either by decreasing the solvent circulation rate, the so-called *separation rate path* (SRP), or the flow rate of the CO_2-rich gas entering the absorber, the so-called *split stream path* (SSP), while maintaining the gas-phase residence time in the absorber packing as well as other design parameters (for details, see the modeling section in **Paper I**). In addition to the standard configuration, three modified process configurations are assessed for their energy and cost efficiencies in the partial capture designs. Rich solvent splitting (RSS; Figure 3-2 b) improves the energy efficiency of the stripper, whereas intercooling of the absorber (ICA; Figure 3-2 c) enhances the absorption of CO_2 into the liquid phase. Furthermore, the combination of RSS and ICA (see Figure 3-2 d) is studied.
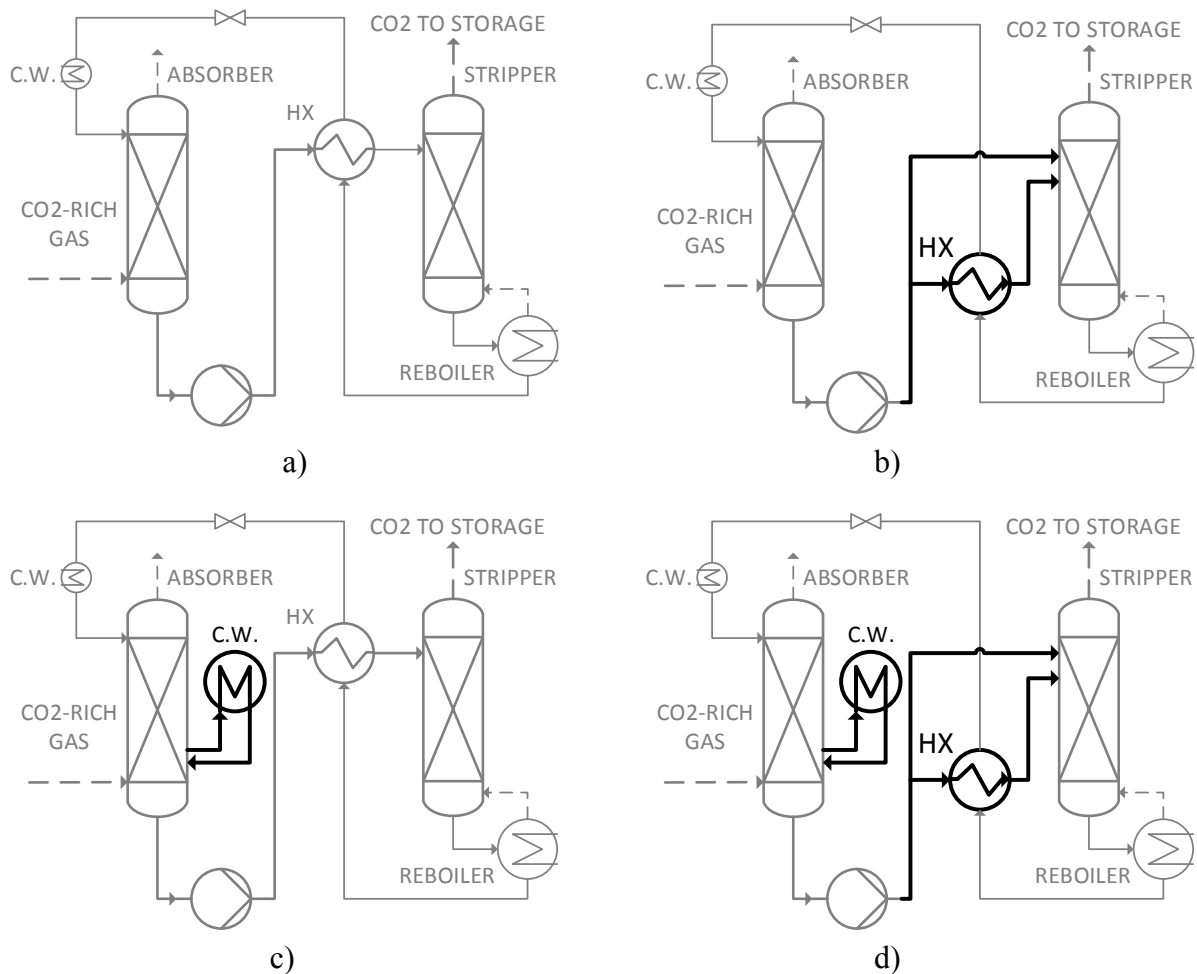
Figure 3-2: Schematic of the CO₂ absorption model and studied process configurations. a) Standard configuration; b) rich solvent splitting (RSS); c) intercooled absorber (ICA); and d) combination of RSS and ICA.

3.1.2 The dynamic absorption model

The dynamic model of the MEA absorption cycle is based on the work of Montañés et al.[70,71] and is described in detail in **Paper A**. The model is written in the modeling language Modelica using the Dymola software with unit operations from the GLC library[72] built by Modelon AB. The unit models of the GLC library have been validated against pilot-plant data by Montañés et al. [70]. The dynamic model calculates rate-based mass and heat transfer and assumes chemical equilibrium for the reactions and enhancement factors for their impacts on mass transfer. The dynamic model describes the standard configuration (*cf.* Figure 3-2) and includes the same units as the steady-state model, apart from the addition of a buffer tank for lean solvent upstream of the absorber. The dynamic model is designed after the steady-state modeled in Aspen Plus with a maximum separation rate of 90% in the absorber, which corresponds to a heat load of 155 MW in the reboiler. This enables partial capture at varying loads according to the separation rate path. Discrepancies between the dynamic and steady-state models are within 1% for the design case and up to 7% for the off-design cases.

3.2 Mapping excess heat with the steel mill model

The steel mill model is an established, in-house spread-sheet-based model of the SSAB integrated iron and steel plant in Luleå, Sweden, which is the reference plant for this work. The steel mill model is a static 1-dimensional model composed of inter-linked mass and energy balances for each unit operation, as described in the work of Hooey et al.[73]. The model considers detailed calculations for the blast furnace, burden and hot stove. In this work, the reference year is 2006 with ca. 3.4 Mt CO_2 emitted and a crude steel production of 2.2 Mt. The steel mill model is applied to quantify the CO_2 emissions from the steel mill's off-gases and to map heat sources that can supply low-pressure steam of ca. 3 bar (~133°C) to drive the MEA solvent regeneration. Table 3-1 lists the five excess heat sources and one heat source using external fuel evaluated in this work. The accumulated level of heat assumes that the technologies will be deployed in sequence – forming *heat levels* (HL) 1–6. The reader is referred to **Paper II** for a description of the recovery technologies, especially for dry coke quenching and dry slag granulation as these are unique to the steel industry.

Table 3-1: Potential heat sources for MEA solvent regeneration, their associated heat recovery technology, recovery efficiency, and heat quantity per emitted kg CO_2 at the reference steel mill (Luleå) without carbon capture. Adapted from Paper III.

Source	Recovery method	Recovery efficiency[a]	Heat (source)[b] MJ/kg CO_2	Accum. Heat (level)[c] MJ/kg CO_2	Heat Level (HL)[d]
CHP plant (excess heat)	Back-pressure operation	63%	0.59	0.59	1
Gas flaring (excess heat)	Steam boiler	93%	0.40	0.99	2
Hot stove flue gas (excess heat)	Heat recovery boiler	91%	0.09	1.07	3
Hot coke (excess heat)	Dry coke quenching + heat recovery boiler	67%	0.11	1.18	4
Hot slag (excess heat)	Dry slag granulation + moving bed heat exchanger +heat recovery boiler	65%	0.24	1.42	5
Additional CHP plant (primary energy)	Biomass fired steam boiler + back-pressure steam turbine	85%[e]	1.08	2.51	6

[a] Potential to convert the excess energy into steam.

[b] Accessible energy from specific source per emitted kg CO_2 at the investigated plant site.

[c] Accumulated accessible energy at the given heat level HL per emitted kg CO_2 at the investigated plant site.

[d] Rating according to level of accessibility (i.e., technology readiness) of the excess energy.

[e] The total efficiency (steam and electricity) is 85% and the electrical efficiency is 22.7%.

3.3 Cost estimation

The cost for implementing partial capture in an extension to an existing integrated iron and steel mill is represented by the annualized investment cost for the capture plant (CAPEX), as well as the operating expenditures (OPEX). Related to the amount of captured CO_2, these costs form what is usually referred to as the (specific) capture cost (€ per tonne CO_2).

The cost estimation method reflects a mature technology ("*n*th-of-a-kind") and uses the Aspen In-Plant Cost Estimator with dimensions obtained from the CO_2 absorption model in Aspen Plus to obtain the cost for each major piece of equipment. The scope of the included equipment is illustrated in Figure 3-3. The equipment cost are multiplied by detailed installation factors retrieved from an in-house database[74] by SINTEF Industry (formerly Tel-Tek), to obtain the installation cost that represents accurately the equipment type and size. In addition, it is assumed that all items of equipment, except for major vessels such as tanks and columns, are placed in non-insulated buildings. A contingency (20%) is included, although the purchase of land, piling, and costs for secondary buildings are not. This method used for CAPEX estimation usually has an uncertainty of ± 40% (80% confidence interval), which is given in parentheses for the estimated cost in the outcome chapters of the present work. The economic parameters used in this work are listed in Table 3-2 and are those commonly applied[74–77]. The plant availability mimics the high availability of major units in the steel mill, and the electricity price reflects the spotprice on the Nordic market Nord Pool, which had an average electricity price of 29 €/MWh in the period 2013 – 2016. In this work the reference currency is €$_{2015}$. The cost of steam is assessed separately in a bottom-up approach for CAPEX and OPEX, following the method described by Ali et al.[78] (see **Paper III** for details).

Table 3-2: Economic parameters assumed for the estimation of capture plant cost in the steel industry

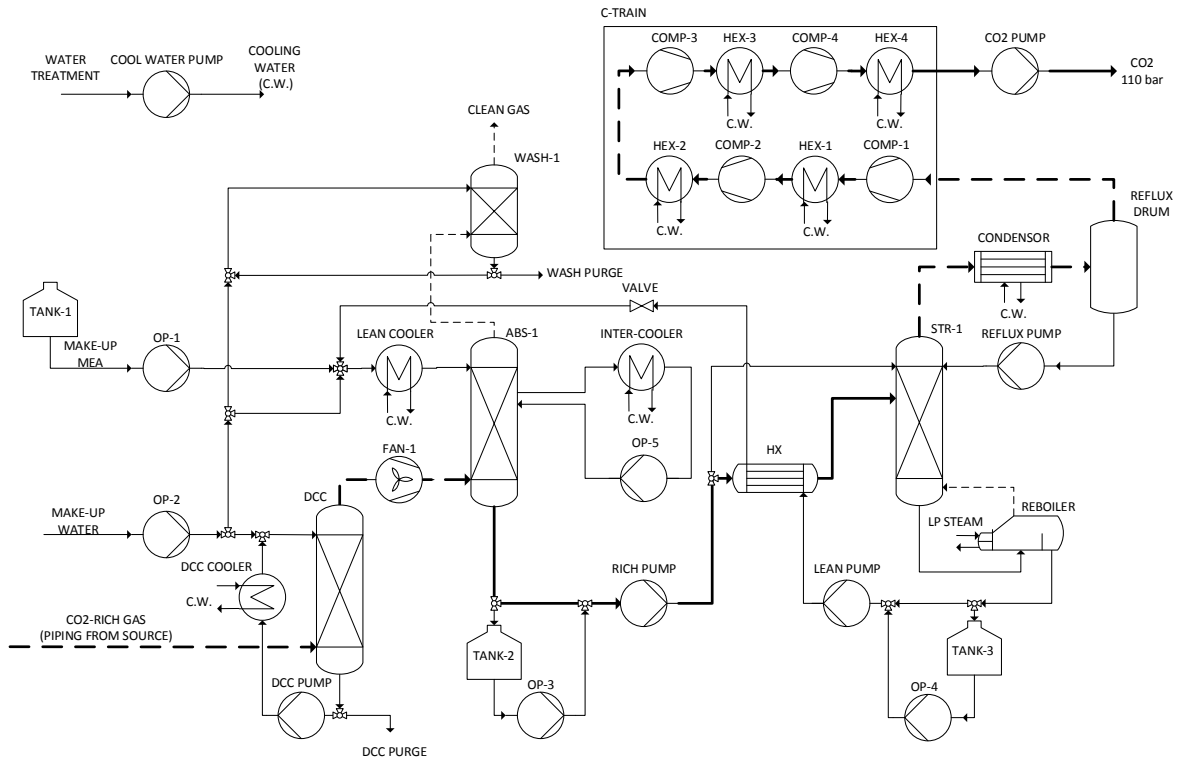|	
Economic plant life time	25 years
Construction time	2 years
Plant availability	95%
Rate of return	7.5%
Annual maintenance cost	4% of investment cost
Annual labor cost	821 k€/annum
Utilities	
MEA make-up	1867 €/m^3
Cooling water	0.022 €/m^3
Electricity	0.030 €/kWh
Steam	assessed separately

Figure 3-3: Scope for the cost estimation of installed equipment in the capture plant. Shown is an exemplary flowsheet for a single-absorber design with rich solvent splitting and absorber intercooling configurations, gas treatment (DCC), and CO₂ compression to 110 bar. Source: Paper III

4 Concept and design of partial capture

4.1 The concept of partial capture

Partial capture of carbon aims, for specific market or site conditions, to capture a small fraction of the CO_2 available onsite. Conditions that may favor the implementation of partial capture in process industry over the implementation of a conventional full capture process, are discussed in this thesis and include (for further conditions, see **Paper I**):

1) Industrial sites that have access to low-cost excess heat[16];
2) Sites with multiple stacks with varying capture costs[34];
3) Market conditions that allow for a continuous optimization of the product portfolio[25] (e.g., volatility of electricity prices or seasonal dependency of district heating); and
4) Carbon capture in combination with other low-carbon technologies, such as fuel switching (from fossil fuel to biomass or natural gas), and improvements in energy efficiency.

These conditions are all valid for today's integrated iron and steel mills. Partial capture is different to full capture in that the lower capture rate confers new technical degrees of freedom that can be used in process optimization, such as gas reallocation (discussed below), and in that it can be designed for market conditions that will vary over time and that value flexibility. As illustrated in Figure 4-1, partial capture sites have the potential to achieve full decarbonization together with co-mitigation measures, and to evolve towards full capture over time. Partial capture is, thus, a low-risk starting point towards the final destination in the "roadmap" for industrial decarbonization.

Carbon capture in general is a technology that allows one to adjust the carbon intensity of industrial products in favor of energy intensity. When applying partial capture that is powered exclusively by excess heat, the carbon intensity of a product can be decreased without increasing the use of primary energy for heat supply, as discussed in Section 5.3.
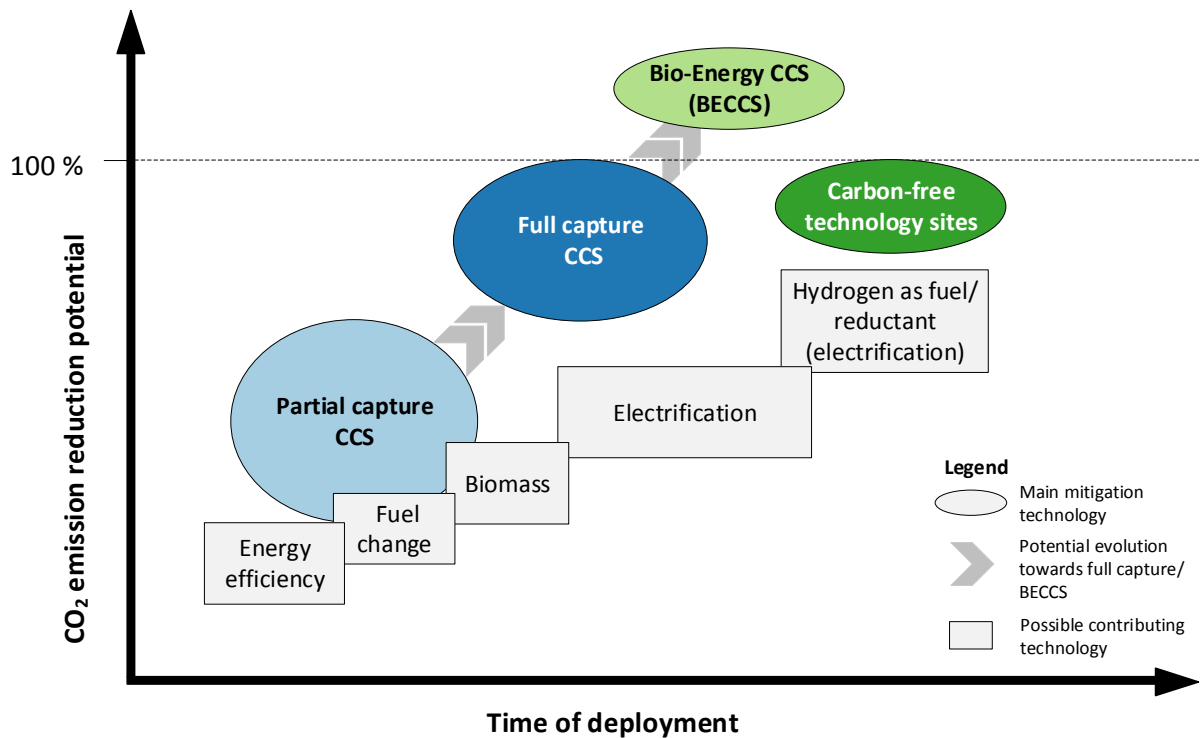
Figure 4-1: Partial capture on an industrial-system level in the context of the decarbonization of process industry over time. Adapted from Paper I

4.2 Partial capture design and implications for steel industry

This section discusses the design of partial capture for process industry and highlights its relevance for implementing partial capture in the steel industry.

Partial capture may be achieved through two pathways, as illustrated in Figure 4-2: 1) the *split stream path* (SSP), in which the capture rate is reduced by bypassing parts of the CO_2-rich gas flow, so that a slipstream is treated at a high separation rate of CO_2 in a downscaled absorber (i.e. ~90%); or 2) the *separation rate path* (SRP), whereby the entire gas flow is treated but a smaller fraction of the CO_2 in the gas flow is separated (i.e., <<90%). The SSP can be interpreted as representing a downscaled full capture design with the same reboiler heat demand per tonne of separated CO_2. The SRP is similar to full capture in terms of the size of the equipment, although it has a lower solvent circulation rate, which means that it separates less CO_2 in the absorber.

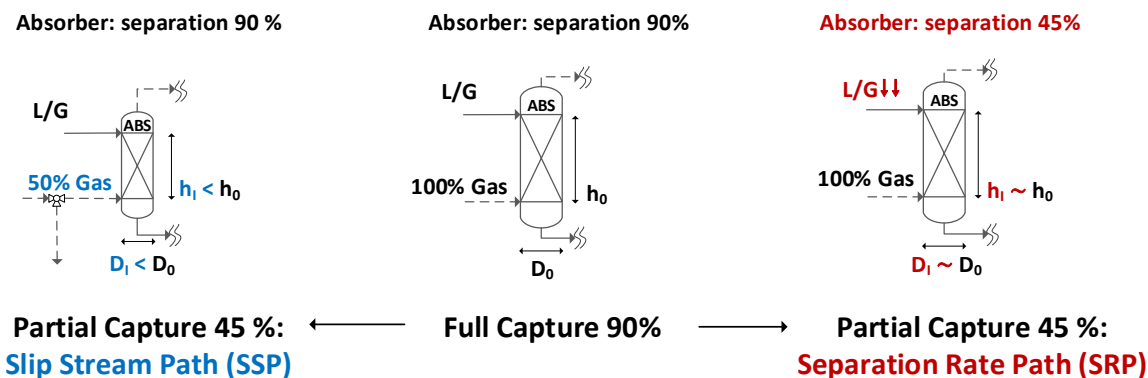**Figure 4-2: The SSP and SRP design pathways for partial capture of 45% of the CO₂, compared to full capture of 90% of the CO₂ in the feed gas. The indices "0" refer to full-capture reference dimensions, and indices "1" refer to partial-capture design dimensions. Adapted from Paper I.**

Figure 4-3 shows the reboiler heat duty (MJ per kg of CO_2) and the estimated capture costs of the two design paths depending on the capture rate for a CO_2-rich gas that is typical for process industry. The specific heat required for the solvent regeneration is lower for the SRP than for the SSP or full capture, as previously described [79]. Depending on the market conditions, this reduction in heat demand may be sufficiently large enough for the specific capture cost (per tonne CO_2) of the SRP to be lower than that for SSP or even full capture. In the literature, the SSP has been identified as the more-cost-effective approach in optimizing the cost for CCS from coal-fired power plants[79–81], which typically have CO_2 concentrations of about 13 vol.%.

The reduced heat demand of the SRP is due to the lower liquid-to-gas (L/G) ratio in the SRP. Figure 4-4a shows how partial CO_2 separation through a lowered L/G ratio reduces the (maximum) temperature in the liquid phase in the absorber [compare how the full capture temperature (■) drops by about 15°C when the L/G ratio is reduced by ca. 3 to achieve partial capture (●)]. At the lower temperature, the CO_2 partial pressure at local phase equilibrium is lower and more CO_2 is absorbed in the same gas-liquid contact area. Thus, the rich loading of the solvent exiting the absorber is increased, which entails a reduced heat demand in the stripper column.

Figure 4-4b demonstrates the reduced heat demands for gases with high CO_2 concentrations when separating CO_2 at lower L/G ratios. The CO_2 concentrations for steel mill off-gases are depicted (areas shaded in blue), revealing that SRP, in terms of energy efficiency, is the preferred design for partial capture from these gases. Note that Figure 4-4 shows the standard process configurations, i.e., without intercooling, which might reduce the heat demand especially for the shown SSP/full capture designs. Importantly, there is no distinct advantage in choosing SRP over SSP/full capture designs for CO_2 concentrations similar to those seen in the flue gases from coal-fired or natural gas-fired power plants, which may be one reason for the previous studies preferring the SSP[79,80,82] as the cost-effective design. In addition, the SRP has the potential to increase the separation rate in the absorber, in case more heat can be made readily available onsite over time. In comparison, the SSP design has a lower capacity to achieve such an expansion of CO_2 separation. In all, the treatment of the entire flow (SRP) of a gas that is typical for process industry requires less heat per tonne CO_2-separated, may, thus, be

even more economic, and is more flexible to load variations and possible extension of capture rates over time, as compared to treating a slip of the gas at a fixed separation rate (SSP).
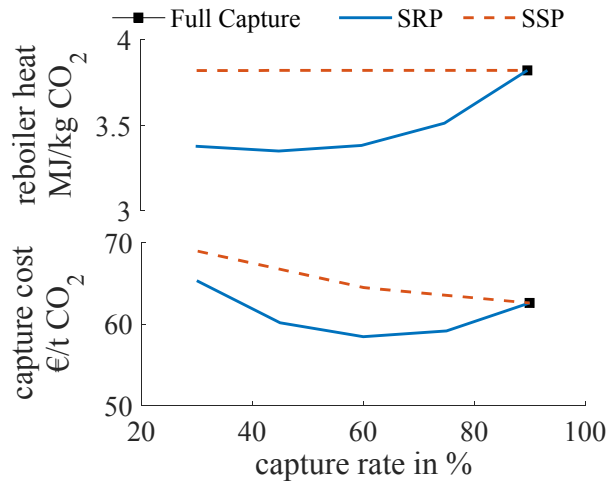

Figure 4-3: Comparison of design paths for partial capture, showing the reboiler heat demand (top) and specific capture cost (bottom) for the separation rate path (SRP) and split stream path (SSP), as compared to a 90% full capture design. The CO_2 concentration in the absorber feed is 20 vol.%. Note that the ordinates do not start from zero. Source: Paper I.

a)   b)
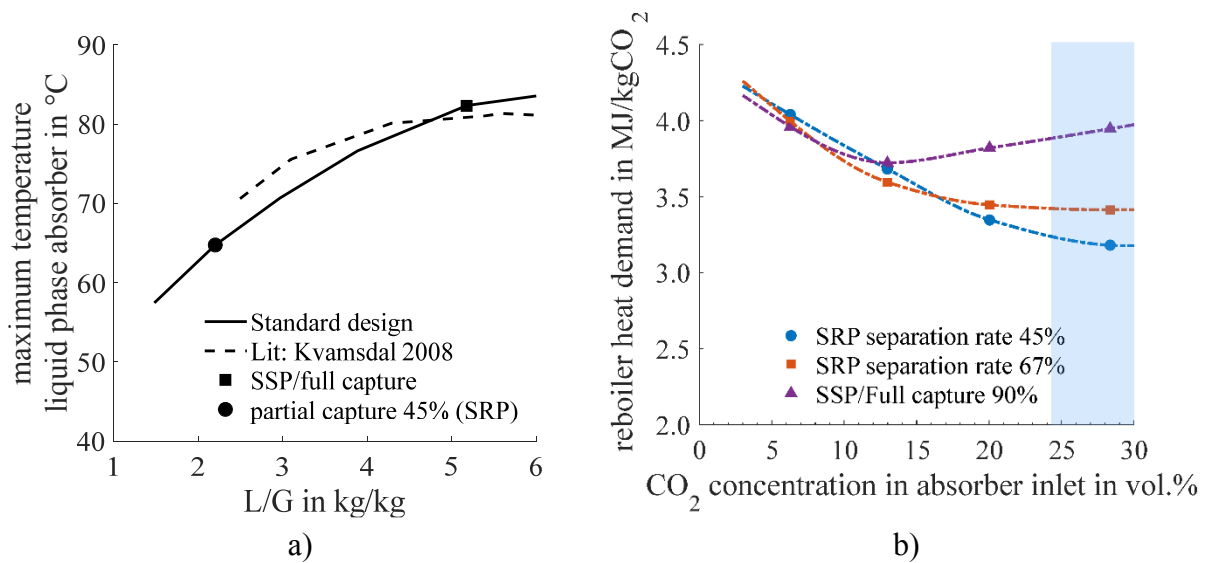
Figure 4-4: Partial capture design choice influenced by feed gas concentration. a) Maximum temperature of the absorber liquid for the separation rate path (SRP) for partial capture (●), as compared to SSP/full capture (■) from a gas with 20 vol.% CO_2; the dashed line refers to Kvamsdal and Rochelle[83]. b) Reboiler heat demand for SRP compared to SSP/full capture with dependence upon the feed gas CO_2 concentration for the standard configuration without intercooling; the blue-shaded area represents the CO_2 concentrations in the steel mill off-gases from blast furnace, CHP plant, and hot stoves. Note that the ordinates do not start at zero. Source: Paper I.

Three modified process configurations (*cf.* Figure 3-2b-d): rich solvent splitting (RSS); intercooling in the absorber (ICA); and the combination of both (ICA+RSS) are analyzed for their applicability to partial capture depending on the separation rates in the absorber. Compared to the standard configuration (*cf.* Figure 3-2 a), the RSS configuration shows lower costs for all separation rates in the range of 45%−90%, while intercooling (ICA) shows no cost-lowering effect for separation rates below 75%. Combining the two process configurations yields a design that cost-effectively reduces the heat demand by 6%–21% compared to the standard

configuration for separation rates of 30%–97%. The interested reader is referred to the detailed analyses visualized in Figures 8, 10 and 14 in **Paper I.** It is noteworthy that for the economic performance of partial capture, the OPEX, which is governed by the steam cost, is found as to predominate over the CAPEX by a factor of 1.5–7.0 depending on the scale (captured CO_2) and the steam cost (*cf.* Figures 17 and 18 in **Paper I**). This result regarding the significance of access to low-value heat is examined and verified for partial capture in the steel industry in **Papers II** and **III**, as explained in the following section.

5 Techno-economic assessment of partial capture in the steel industry

5.1 Technical performance of partial capture of CO_2 from steel mill off-gases

The findings related to partial capture design are tailored to the properties of CO_2-rich gases from the reference steel mill. This section highlights key findings in relation to the performance depending upon the quantity and intermittency of the heat supply to the absorption process. The first section focuses on varying quantities of, mainly, excess heat to fuel partial capture under the assumption of constant heat load, while the second section provides insights into the performance of the CO_2 capture unit while varying both the heat load and gas supply.

5.1.1 Partial capture with excess heat at constant load

The choice of CO_2 source in a steel mill – the three major sources of CO_2 that carry most of the CO_2 emitted at the reference site are included in the investigation: The first two are flue gases from the CHP plant and the hot stoves that emit 59% and 22 % of the CO_2 onsite, respectively. The third is the blast furnace gas that is combusted in, *inter alia*, the hot stoves and the CHP plant and contains 44% of the CO_2 emitted. These gases have a concentration of CO_2 around 25 vol.% or 30 vol.% in case of the flue gas from the CHP plant. Each gas is studied as an exclusive feed to an absorber, thus, no blending of gases is analyzed. The SRP with a combination of the RSS and ICA configurations is applied in all cases. Figure 5-1 shows the specific heat requirements for CO_2 release from the MEA solvent in the stripper for all three sources of CO_2. The reboiler heat demand increases the lower the CO_2 partial pressure becomes in the lean gas, i.e., the desired level of CO_2 removal from the gas. Separation from BFG requires the least heat, since it has an elevated pressure of 1.8 bar, which enhances the physical absorption of CO_2 into the liquid phase leading to a richer loading, which reduces the heat demand in the reboiler. For similar shares of CO_2 separation, end-of-pipe capture from the atmospheric flue gases of the CHP plant or the hot stoves shows a higher heat demand than CO_2 capture from the pressurized blast furnace gas.
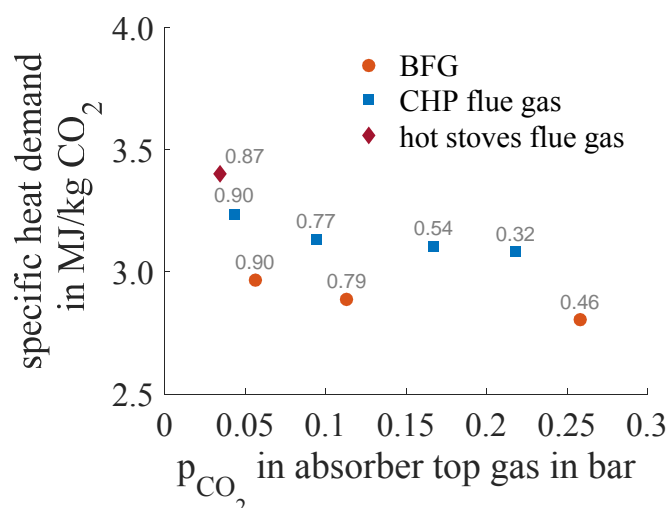
Figure 5-1: Specific heat demand for 30 wt.% MEA absorption from a blast furnace gas (BFG), the flue gas from a combined heat and power (CHP) plant, and the flue gas from the hot stoves with dependence upon the partial pressure of CO_2 in the lean gas exiting the absorber. The data labels indicate the separation rates of CO_2 in the absorber (in %).

The choice of high- versus low-level of integration affects the steel mill. The quantities of CO_2 that may be captured by utilizing the various sources of excess heat mapped in Table 3-1 are determined for the CHP plant flue gases (low-level of integration of CCS) or blast furnace gases (high-level of integration of CCS). Figure 5-2 matches each level of excess heat (HL) with an achievable reduction of emissions depending on the source of CO_2. The difference in the slopes of the *two curves* (red and blue) reflect the above mentioned difference in heat demand between BFG and CHP capture (*cf.* Figure 5-1). If more heat can be retrieved beyond a CO_2 separation rate of about 95% in the BFG, capture from the flue gases of the CHP plant is preferred. In the case of CO_2 capture from the BFG, the amount of available heat in HL2–5 is increased through the CO_2 capture. As more CO_2 is removed from the BFG, the heating value of the BFG increases and the BFG can be used more extensively in the hot stoves than in the CHP, which releases coke oven gas to the CHP instead (*cf.* Figure 10 in **Paper II**). Although less energy is allocated to the CHP in this way, the amount of excess heat from the hot stove flue gases are increased (HL2–5) [compare the slope of the *steps* in Figure 5-2]. Thus, overall the steel mill uses less energy at the same production rate (*cf.* Table 4 and Figure 11 in **Paper II**), also by avoiding cooling in the condensing turbine stages. Overall, a high-level integration of CCS through CO_2 capture from BFG is more energy-efficient, i.e., more CO_2 can be captured for the amount of retrieved heat, and potentially allows for a more flexible gas allocation and thereby energy management of the steel mill.

Table 5-1 summarizes the capture scenarios investigated in **Papers II** and **III,** and describes the achievable reductions in emissions from a single CO_2 source (Scenarios 1–3) that is fueled exclusively by excess heat. For hot stove flue gases and the blast furnace gas, the limit is set by achieving full capture from the respective source, i.e., 90% separation of CO_2 in the absorber. For CHP plant flue gases, the limit is set by the amount of retrievable excess heat (i.e., 90% separation is not reached). Overall, the CHP flue gas carries the largest quantity of CO_2 in a single stream onsite, which implies a 43% reduction in site emissions if all the excess heat was to be extracted.
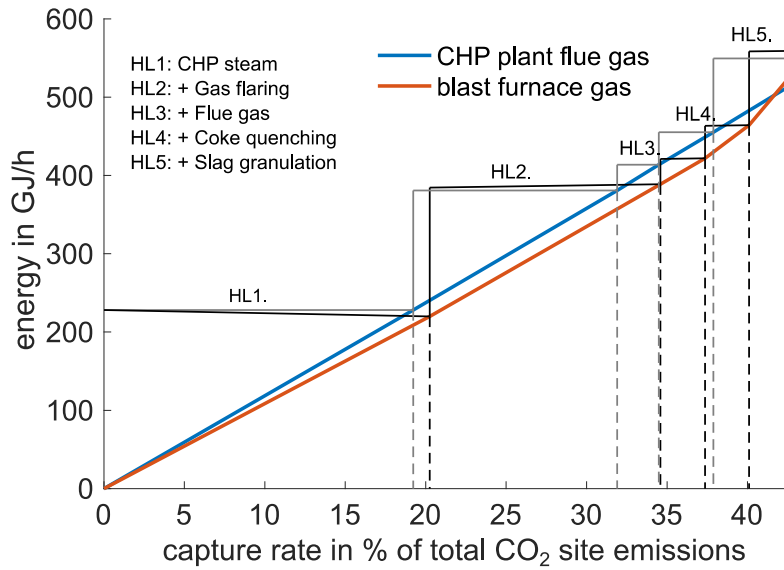
Figure 5-2: Levels of retrievable excess heat (HL1–5) and the resulting emissions reduction for capture from the blast furnace gas compared to end-of-pipe capture from the CHP plant flue gas. Source: Paper II.

Table 5-1: Investigated capture scenarios and their maximum achievable emissions reductions, as well as their additional fuel and power requirements. The percentages given in parentheses for 'extra fuel' represent the share of the total heat supply to the reboiler. FGHR, flue gas heat recovery; CDQ, coke dry quenching; DSG, dry slag granulation; Bio-CHP, biomass-fired CHP plant. Adapted from Paper III.

Capture scenario	CO_2 source	Applied heat sources	Max. site reduction % CO_2	Extra fuel MJ/kg$_{CO2}$	Net power import MJ/kg$_{CO2}$
1	hot stoves flue gas	FGHR; back-pressure	19.0	0	-0.01
2	blast furnace gas	Back-pressure; flare gases; FGHR; CDQ	38.8	0	+0.07
3	CHP plant flue gas	Back-pressure; flare gases; FGHR; CDQ; DSG	43.2	0	+0.09
4	blast furnace gas + hot stoves flue gas	Back-pressure; flare gases; FGHR; CDQ; DSG; Bio-CHP	51.0	0.28 (11%)	+0.06
5 *(full capture)*	blast furnace gas + hot stoves flue gas + CHP plant flue gas	Back-pressure; flare gases; FGHR; CDQ; DSG; Bio-CHP	76.3	1.66 (43%)	-0.15

5.1.2 The impacts of seasonal and hourly variations on capture performance

The impact of the above assumption of a constant, yearly- averaged heat load is highlighted in this section. For this, the assumption is released and, instead, seasonal and hourly variations are considered. Based on a steady-state design (SRP) in the standard process configuration, the behavior of the MEA absorption cycle in terms of response time and capture performance is simulated when there are variations in feed gas flow (BFG) and heat supply. As the heat source, steam from the CHP plant operated in back-pressure mode and from the combustion of flare gases is applied. District heating causes large seasonal differences in the amounts of available heat from the CHP plant averaging 0 MW and 110 MW in the winter and summer, respectively (*cf.* Figure 4 in **Paper A**). Variations in the BFG flow and gas flaring occur frequently throughout the year, although they often last for only 2 hours.

Figure 5-3 compares the capture performance of a dynamic plant following actual variations to a steady-plant that uses the same averaged amount of heat during a 2-week period in summer. The dynamic plant, in fact, captures 1% more CO_2 than the steady-state plant over the shown time period. Implementing a feedback control strategy that controls the stripper bottom temperature by manipulating the solvent circulation rate increases by an additional 1.2 % the amount of captured CO_2. The reason why the dynamic plant performs so well is the non-linearity of the response to changes in heat load. Figure 5-4 shows the absorbed (absorber) and released (stripper) CO_2 for a periodic variation in heat load (±30 MW) depending on the duration of one cycle. This demonstrates that the increase in CO_2 production in response to a heat increase is both faster and of greater magnitude than the drop in CO_2 production caused by a decrease in heat of the same magnitude. The figure also reveals a buffering capacity for the solvent between the absorber and stripper, which allows for temporary CO_2 release from the stripper even when no gas enters the absorber. The buffering capacity is a function of the size and location of the solvent buffer tank and the solvent circulation rate. It affects the response time of the plant, which, for example, is slower in winter due to lower solvent circulation (lower heat load). **Paper A** concludes that the dynamic MEA capture plant copes well with the described variations within the reference steel mill and can deliver a capture performance similar to that of a steady-state plant, as assumed in **Papers II** and **III**. The prerequisite for this is that the absorption process is designed to be sufficiently large to cope with the entire span of the experienced seasonal variations of the heat load. A first estimation gives an increased cost of 6 (±2) € per tonne CO_2 for increasing the heat load from 61 MW (yearly average) to 155 MW (maximum heat load) for the same amount of captured CO_2 from back-pressure steam per annum. This cost difference may be optimized through a trade-off between the annual capture rate and the cost-efficiency of the design, and requires further investigation. Alternatively, heat storage facilities may be an attractive option, although have not been assessed here. Furthermore, the flare gases may require a buffer storage or a boiler that is sufficiently large to manage their variations. The consequences of varying the loads of the capture unit that are experienced downstream of the unit, e.g., for the CO_2 compressor and sizing of CO_2 handling facilities, have not been assessed and warrant further investigation.
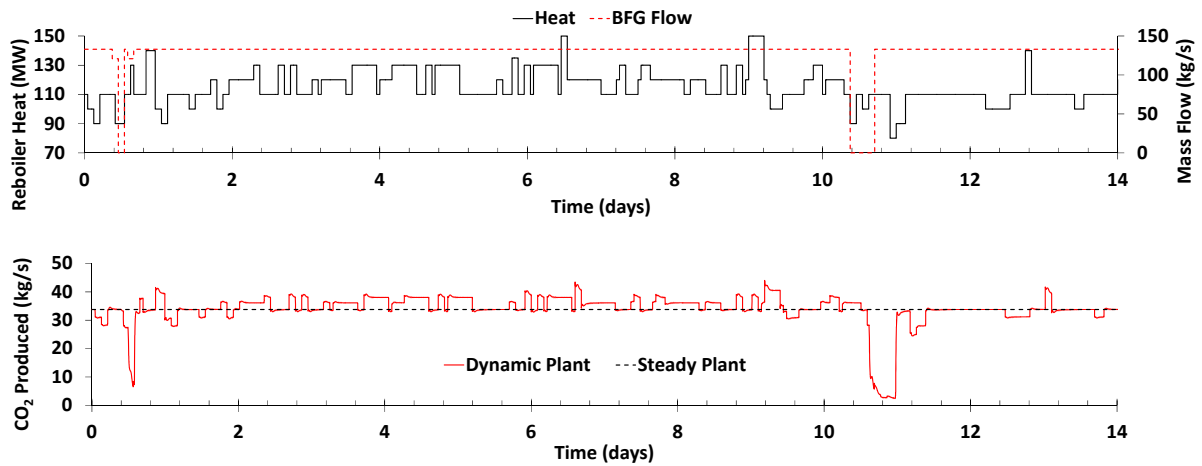
Figure 5-3: Capture performance of a blast furnace gas (BFG) during a 2-week period. Upper panel: Historic variations in the BFG flow and available heat from back-pressure operation and flare gases. Lower panel: Transient responses in CO_2 production to variations for a dynamic plant, as compared to a steady-state plant utilizing the same average heat at constant load. For details as to the origins of the historic data, see Paper A. Source: Paper A.

Figure 5-4: Relative amplitudes of CO_2-produced (stripper) and CO_2-absorbed (absorber) depending on the period of sinusoidal variation (±30MW around the 110-MW baseline) in the reboiler heat duty. The maximum (Increase) and minimum (Decrease) values of the responses are plotted separately. Source: Paper A.

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5.2 Economic performance of partial capture in the steel industry

This section quantifies the value of partial capture in the steel industry compared to full capture, and identifies low-cost captur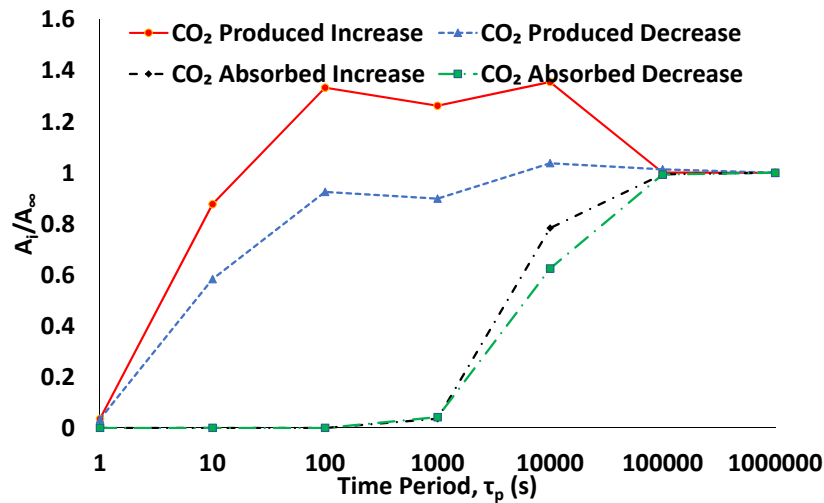e scenarios. A detailed study based of the capture scenarios listed in Table 5-1 is conducted (for a detailed description, see **Paper III**). The underlying approach is to apply energy-efficient designs for partial capture together with low-value excess heat to minimize the specific capture cost. The partial capture scenarios (Scenarios 1–3) are powered by up to five sources of excess heat and are compared to Scenarios 4 and 5 that require an additional supply of primary energy, which is assumed to be low-grade biomass (HL6 in Table 3-1), as a supplement to excess heat. Scenario 4 combines CO_2 capture from BFG and hot stoves in a common capture unit with two absorbers and one stripper, targeting a CO_2 separation rate of 90% in each absorber. Scenario 5 represents the full capture benchmark scenario by adding another capture unit to separate 90% of the CO_2 in the CHP plant flue gas, which results in a 76% reduction of CO_2 emissions onsite.

5.2.1 Cost of heat supply

A bottom-up analysis of the production cost of low-pressure steam is conducted and considers the investment costs and operating costs. The OPEX includes power, cooling water, additional fuel (biomass), and loss of sales revenues from power generation in the CHP plant when switching from condensing mode to back-pressure operation. Besides steam piping, the equipment considered varies with the heat-supplying technology used, see the Appendix of **Paper III** for more details. Figure 5-5 provides the steam cost depending on the quantity of
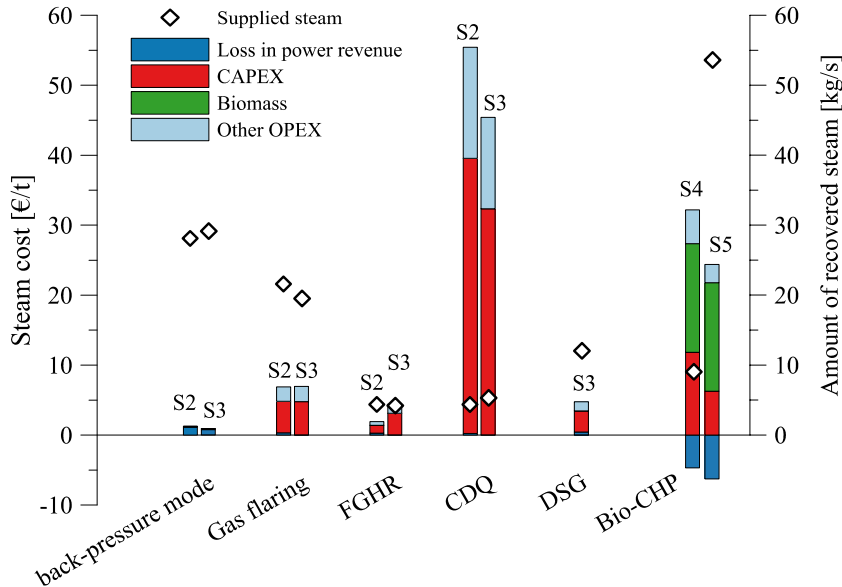

Figure 5-5: Production cost for steam at 3 bar from the heat-supplying technologies investigated in this work. S1–S5 correspond to capture Scenarios 1–5 in this work. FGHR, flue gas heat recovery; CDQ, coke dry quenching; DSG, dry slag granulation; Bio-CHP, biomass-fired CHP plant. Source: Paper III.

steam and the applied recovery technology. Back-pressure operation, heat recovery from hot stove flue gases (FGHR), and utilization of flare gases for steam production are comparatively low-cost options for the heat supply with the cost of steam in the range of 1 ($\pm$0.1) € to 7 ($\pm$2) € per tonne of steam. In comparison to these sources of excess heat[A], the cost of steam from primary energy supply (biomass in this case) is expensive, being in the range of 18 ($\pm$3) €/tonne and 28 ($\pm$5) €/tonne for Scenarios 4 and 5 ("Bio-CHP"), respectively. Note that the price for steam would be around 14 €/tonne due to scaling effects if all the heat was supplied through biomass, instead of the only 43% in Scenario 5 (the rest is excess heat), assuming the price for low-grade biomass, such as bark, is 16 €/MWh. Applying prices for refined wood fuels[84] (e.g., pellets, ~25-30 €/MWh) leads to costs of 23–28 €/tonne steam if all the heat was to be supplied through biomass. **Paper III** highlights the scale required for supplying CO_2-neutral biomass and the competition from other sectors for this scarce resource as potential challenges when using biomass to achieve full capture. Under present market conditions, fossil fuels may be more economic than biomass, but they are not considered as a viable option given that their use entails additional direct CO_2 emissions onsite. In all, steam from excess heat is cost-efficient compared to steam from the combustion of additional fuels.

5.2.2 Absolute and specific costs for partial and full capture in a steel mill

The economic efficacy for full capture and partial capture is quantified through both absolute and specific costs under the influence of scale and steam cost. The CAPEX and OPEX for Scenarios 1–5 are estimated as described in Section 3.3. The installation costs are in the range of 93 ($\pm$37) M€ to 307 ($\pm$122) M€ for the smallest and largest annual capture capacities of 0.6 Mt CO_2/annum and 2.6 Mt CO_2/annum, respectively (Figure 6 in **Paper III**). The corresponding annualized absolute cost including CAPEX and OPEX are 20.6 ($\pm$4.1) M€ and 99.5 ($\pm$12.1) M€. The compressor, cross heat exchanger, reboiler and gas piping are the costliest items of equipment. The equipment cost is affected by the differences in scale, the CO_2 source, and the design of the capture unit (number of units, two-absorber-one-stripper design), which is considered in the cost estimation. As examples, the reboiler size is affected by the specific heat demand, the piping depends on the location and scale of the CO_2 source, and the number of columns varies in certain cases (e.g., two-absorber-one-stripper design in Scenario 4, no direct contact cooler for BFG). Perhaps self-explanatory but important, the estimated cost show that partial capture reduces significantly the absolute installation cost and annualized cost and thereby can lower the threshold for investment.

Figure 5-6 illustrates the annual cost for the studied Scenarios 1–5 with regards to the heat source and the achieved reductions in CO_2 emissions. The blast furnace gas gives the lowest capture costs – on average, the annual costs are 3–5 €/tonne CO_2 lower than end-of-pipe capture. The cost span is mainly attributed to the increased pressure of the BFG, which leads to a lower heat demand (OPEX) and lower CAPEX as the diameters of the columns and piping become smaller compared to those at atmospheric pressure. Although the difference of 3–5 €/tonne CO_2

[A]The other two sources of excess heat, coke dry quenching (CDQ) and dry slag granulation (DSG), are deemed either too expensive or not commercially available yet – see discussions in **Paper III**

between the capture from the BFG and the flue gases is in the order of the magnitude of the uncertainty for the cost estimation, it is substantial considering the magnitude of the emissions and the present EU ETS prices. When supplied with back-pressure steam from the CHP plant and the steam accrued from flue gas heat recovery and flare gases, capture from BFG achieves an emissions reduction of 36% (ca. 1.2 Mt CO_2/annum) at a minimum annual cost of 28 (±4) €/tonne CO_2-captured. In comparison, full capture from all three CO_2 sources can achieve a reduction in site emissions of 76% (ca. 2.6 Mt CO_2/annum) at 39 (±5) €/tonne CO_2-captured. It should be noted that these full capture costs rely on the availability of low-value biomass and excess heat. They would be in the range of 41–50 or 48–57 €/tonne CO_2-captured if wood pellet prices are applied or the entire heat is supplied by natural gas or coal (extra emissions not considered), respectively. Figure 5-7 attributes this large difference in economic performance to the steam cost, which dominates the CAPEX in the full capture scenario. This illustrates that cost savings may overcome the effects of economy-of-scale favoring higher capture rates - if excess heat is used. Note the higher share of CAPEX for partial capture compared to full capture – this motivates having the focus on low-cost and simple process configurations (e.g., RSS and ICA) for partial capture. In all, this analysis shows that full capture is more reliant on primary energy than partial capture that is powered through excess heat, and, therefore, has a cost structure that is more sensitive to external risks, such as changes in the energy system and market volatility.
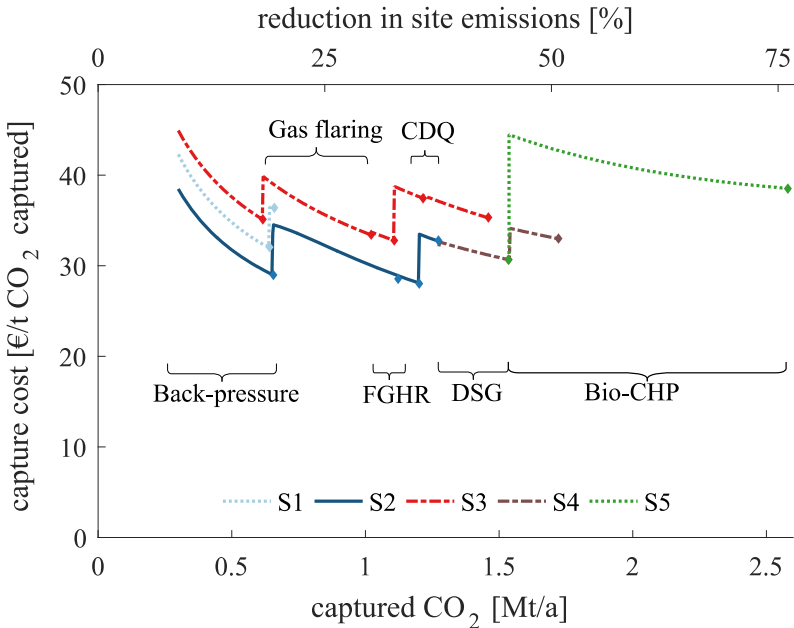
Figure 5-6: Annualized capture cost in relation to heat source and achieved capture rate at the site level. S1–S5 correspond to capture Scenarios 1–5 in this work. FGHR, flue gas heat recovery; CDQ, coke dry quenching; DSG, dry slag granulation; Bio-CHP, biomass-fired CHP plant. Source: Paper III.
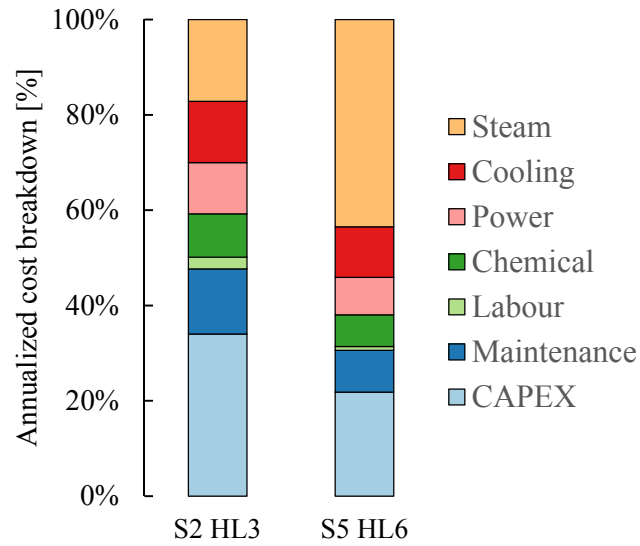
Figure 5-7: Breakdown of the annualized costs for partial capture (S2 HL3) and full capture (S5 HL6) in a Swedish steel mill. Partial capture is from BFG with excess heat from back-pressure operation, flue gas heat recovery and flare gases. Full capture is from BFG, CHP and hot stove flue gases with all sources of excess heat and additional fuel combustion. Source: Paper III.

5.3 The impact of partial capture on the carbon and energy intensities of primary steel

Over the last few decades, the blast furnace route (BF-BOF) has improved its energy efficiency and has reached a technical minimum of CO_2 emissions close to the theoretical limit of the chemical reduction of iron oxides to iron in the blast furnace[39] *when the applying best-available-technology (BAT).* Figure 5-8 shows the carbon and energy intensities of primary steel production and illustrates how applying CCS or direct-reduced iron production using natural gas (DRI-EAF) affects these relations. Three CCS options are shown. Top-gas recycling with CCS (TGRBF-CCS) could improve energy efficiency beyond the BAT-levels and simultaneously reduce the carbon footprint of BF-BOF. Partial capture that is powered exclusively by excess heat reduces the CO_2 intensity while only marginally affecting the energy intensity of primary steel. Full capture, however, requires more energy per avoided carbon and therefore deviates from the current BAT-levels. Despite the enlarged energy footprint due to a reliance on additional necessary fuel, full capture may achieve large reductions in CO_2 intensity. While the carbon intensity of DRI-EAF is similar to that of partial or full capture, it varies considerably with the CO_2 intensity of the power grid, as exemplified in Figure 5-8 for the values of 0 g CO_2/kWh and 500 g CO_2/kWh representing a grid with 100% renewables (CO_2-free) and a grid similar to that in Germany[59], respectively.
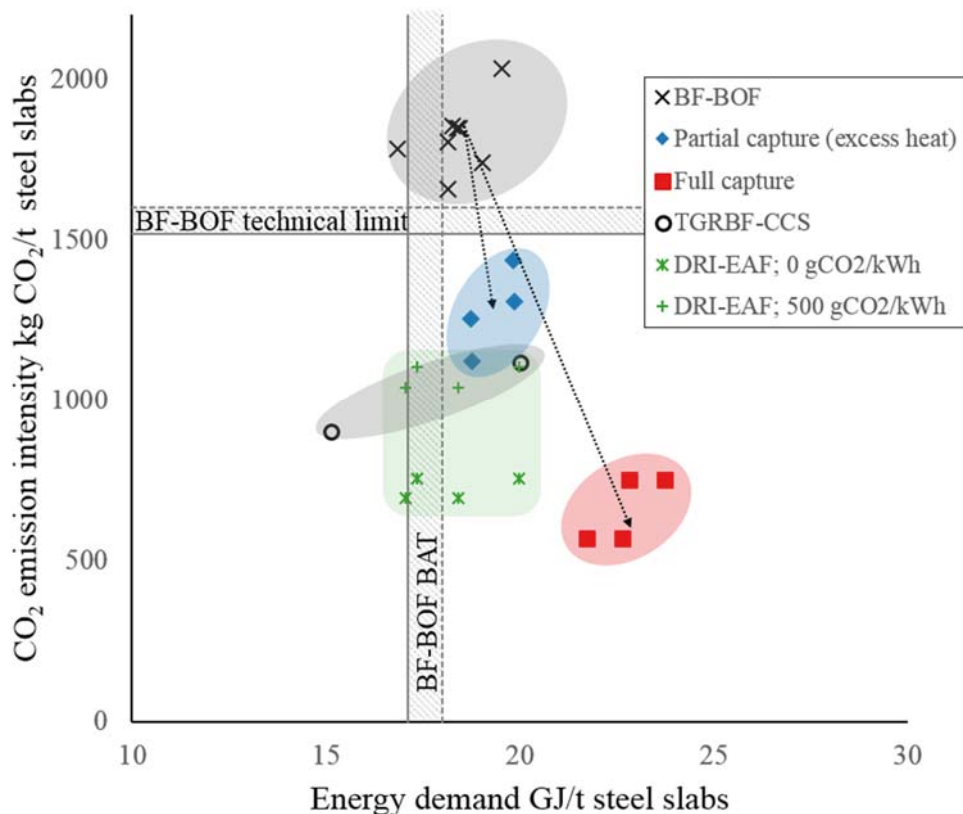
Figure 5-8: The relationship between the carbon and energy intensities of primary steel slabs produced using: blast furnaces without CCS (BF-BOF); blast furnaces with CCS (partial and full CO_2 capture), oxygen-blown blast furnaces with top gas recycling and CCS (TGRBF-CCS); and direct reduction with natural gas (DRI-EAF). Shown are the results for: partial CO_2 capture from either BFG or CHP exclusively powered with excess heat; and full capture from BFG, CHP and hot stoves (combined) with excess heat and external energy (CO_2-emissions neutral). Based on data from Paper III and from references[15,39,85–87]**.**

6 Partial capture – a window of opportunity for the steelmaking industry

Conditions for near-term implementation - the estimated cost for partial capture (€/tonne CO_2-captured) for the reference steel mill is combined with the transport and storage costs and discussed in relation to projections of the price of carbon. Figure 6-1 shows the full CCS-chain cost, i.e., the capture, transport and storage cost, in relation to the three carbon price projections: (i) a sustainable development scenario *WEO 2°C* in line with the 2°C target according to IEA[6]; (ii) a moderate development scenario *WEO&NEPP*[6,88]; and (iii) a market-oriented EUA forecast (EU ETS forecast) by Refinitiv[89]. The figure illustrates that early implementation of partial capture in the steel industry in the 2020s would be economically viable for ambitious and moderate carbon price developments throughout its entire economic lifetime of around 25 years. The market-oriented forecast of EU ETS does not foresee carbon prices being sufficiently high to trigger an implementation before Year 2030, nor until Year 2040 if extrapolated. In light of a carbon-restricted economy, partial capture deployed later than Year 2030 would not be sufficient to reach the EU emissions targets for the period 2040–2050. Instead, full capture or alternative production pathways would be required to achieve close to 100% decarbonization.
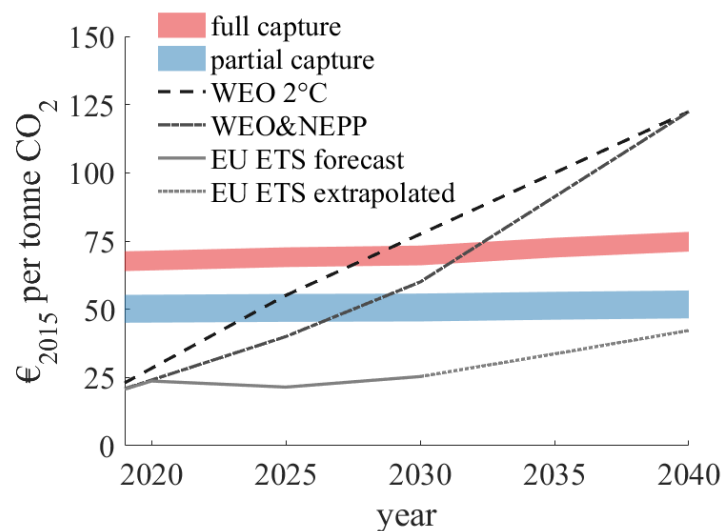

Figure 6-1: Abatement cost for the steel industry based on partial capture from the blast furnace gas with excess heat from back-pressure operation, flue gas heat recovery, and flare gases compared for three carbon price scenarios: a sustainable development scenario (WEO 2°C)[6], a moderate development scenario (WEO&NEPP), and a carbon-market forecast (EU ETS forecast) by Refinitiv[89]. The carbon price for EU ETS has been extrapolated for the period 2030–2040. Electricity cost for Sweden according to NEPP[88,90] scenarios; the shown full capture is from BFG, CHP plant and hot stove flue gases (combined) using excess heat and biomass combustion; the biomass price is set equal to the electricity price. Ship transport to a storage site in the Baltic Sea assumed[91], see Paper III for details. Adapted from Paper III.

Discussion as to the role of partial capture in the transition to CO_2-neutral steel production - the implementation of partial capture should consider the projected operation of the blast furnace in terms of economic and technical lifetimes. The following discussion describes the possible decarbonization pathways for a steel mill with a blast furnace that reaches the end of its lifetime by 2040–2045 and is replaced by hydrogen direct reduction (H-DR) based on 100%

renewable energy (electricity & biomass). The pathways assume that investment decisions are made in the 2020s. Possible decarbonization pathways that assume initiation of operation in Year 2025, are illustrated for the reference steel mill in Figure 6-2. Note that the figure does not include the full capture scenario, top gas recycling with CCS (maintaining the BF-BOF technology), alternatives such as HIsarna (smelting reduction with CCS), or even EW, as these technologies are unlikely to be implemented for a sufficiently long period prior to the transition to H-DR in Year 2040/45. However, they represent alternatives to the implementation of H-DR. As indicated in Figure 4-1, partial capture could also be a catalyst for the implementation of many of these technologies.

In Figure 6-2, the business-as-usual (BAU) scenario assumes a renewal of the refractory lining (required every 15–20 years) inside the blast furnace, which would allow continued BF operation until the end of lifetime in 2040/2045. It should be noted that end-of-pipe capture and, most likely, BFG capture will not have to be synced with the relining/overhaul of the BF and just coincide in the figure. In comparison, the DRI/EAF pathway assumes a shutdown of BF-BOF production already in 2025 and a switch to direct reduction, requiring the respective natural gas infrastructure.
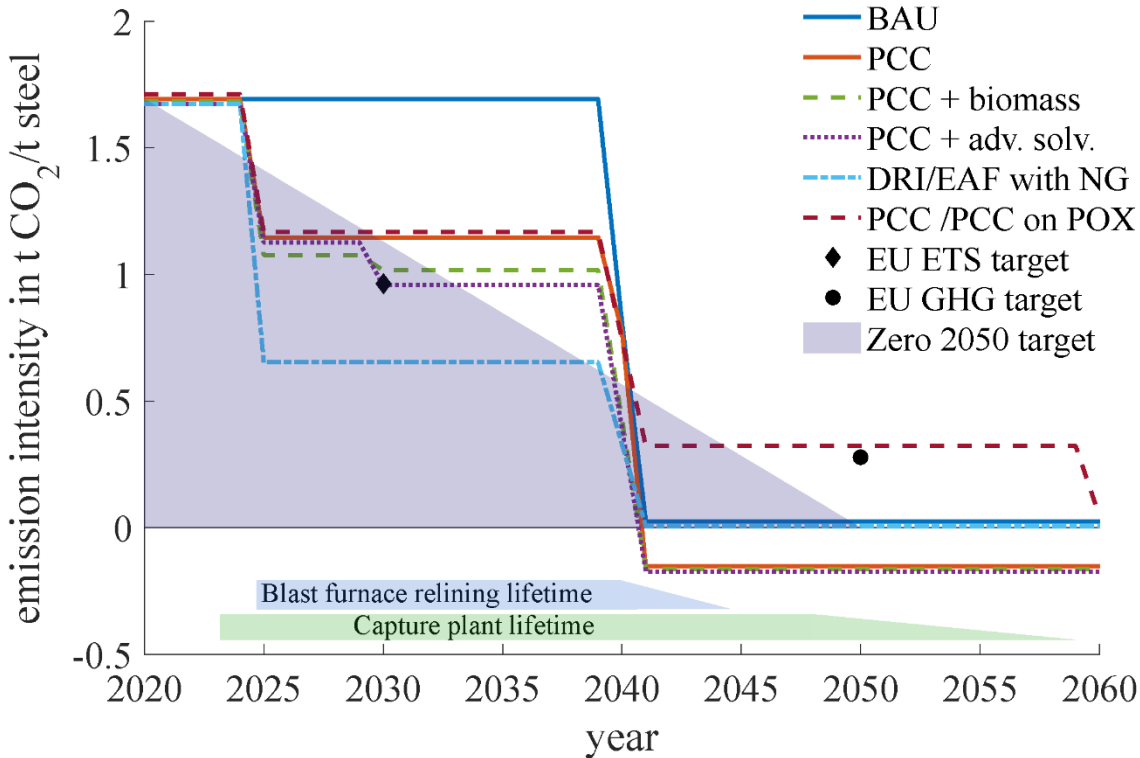

Figure 6-2: Timelines of the decarbonisation pathways for BF-BOF steelmaking in the transition to hydrogen reduction by Year 2040 based on a reference steel mill (Luleå) with 2.2 Mt of produced steel per annum. BAU, Business-as-usual BF-BOF, then hydrogen direct reduction (H-DR) with hydrogen from renewable energy (RE), assumes as 0g CO_{2eq}/kWh$_{el}$; **PCC**, partial carbon capture from BFG with excess heat, then capture of fossil and biogenic CO_2 from H-DR with RE; **PCC + biomass,** bio-coal injection into BF replacing first 10wt.% then 20wt.% of fossil pulverized coal; **PCC + adv. solv. (advanced solvent)**, assumes 2.5 MJ/kg CO_2; **DRI/EAF with NG**, direct reduced iron/electric arc furnace with natural gas (NG), assumes 50 g CO_{2eq}/kWh$_{el}$; **PCC /PCC on POX,** 70% of the hydrogen is supplied via partial oxidation (POX) of coal (or NG) with PCC and 30% via electrolysis with RE, assumes 50 g CO_{2eq}/kWh$_{el}$. Based on own calculations, data in **Paper III** (capture from BFG with excess heat - HL3), and data from the literature[30,40,59,92].

The remaining four scenarios illustrate the roles that partial capture could play if operated from Year 2025 onwards with 1.2 Mt CO_2/annum captured (resembles CO_2 capture from BFG,

HL3 *cf.* Table 3-1) during the lifetime of the BF. After the transition to H-DR, the invested CCS infrastructure could be used to capture the remaining fossil and biogenic emissions, which would amount to 55,000 and 370,000 tonne CO_2 per annum, respectively, according to the HYBRIT concept[40], and thereby achieve net-negative CO_2 emissions. Biomass could also be used earlier as a co-mitigation measure with partial capture; the corresponding curve in Figure 6-2 assumes the stepwise introduction of bio coal into the blast furnace, replacing first 10 wt.% and then 20 wt.% of the injected pulverized coal. The shown emission reduction foresees a credit being granted for negative emissions; see the description of the allocation method in the previous work by the author[49]. The use of advanced solvents (2.5 MJ/kg CO_2) for partial capture by Year 2030 could be sufficient to attain the EU ETS reduction goal of 43%, as compared to Year 2005.

Furthermore, an early initiation and a parallel track for large-scale emissions reduction would decrease the risk of the hydrogen based concept. For example, the CCS infrastructure could be used for supply of carbon-neutral hydrogen ("blue hydrogen") from fossil feedstocks. Scenarios when this will be needed are easy to imagine and may, for example, include that: (i) the carbon-free electricity generation is not realized, meaning that the carbon footprint of the steel would increase (sensitivity: 0–500 g/kWh correspond to ca. 25–1580 kg CO_2/t steel); and (ii) the required electrolysis, power generation, or hydrogen storage capacities are not realized due to difficulties to scale-up in time or to achieve economic viability. One potential fossil feedstock is coal, for which an infrastructure is already in place, or natural gas, which is common for hydrogen generation today. In this respect, partial oxidation (POX) from coal could be a possible technology[B]. Alternatively, steam methane reforming with CCS could be applied as well, possibly using chemical looping technologies[93]. In the case shown in Figure 6-2 (POX), the 1.2 Mt CO_2/annum possible to capture with the partial capture unit proposed would be able to produce ca. 70% of hydrogen needed from fossil sources (90% separation) ("blue"), while the remaining 30% could be supplied from renewable ("green") hydrogen. A synergy may be achieved between the oxygen accumulated as a by-product of water electrolysis[40] and the required oxygen supply to the gasifier.

In addition, partial capture could mitigate to some extent the risk for carbon leakage and thereby possibly prevent primary steelmaking industry from migrating to other countries. While free allocation of EUAs is most likely to cover the majority of the emissions for the period until Year 2030, steel producers may face lower shares of free allocation after Year 2030. Implementing partial capture would facilitate closing the emerging gap or confer the flexibility to sell-off superfluous EUAs.

In summary, partial CO_2 capture is a mature and low-cost technology that has a time-window for implementation in the coming 10–15 years (or within one more investment cycle for the BF refractory lining), after which the lifetime of the capture unit will most likely be too short before policies calling for 100% decarbonization will favor full capture technologies or alternative

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[B] This has traditionally been achieved through acid gas removal of both CO_2 and sulfur with Rectisol or Selexol or MDEA/DEA solvents, and at high pressures. Utilizing the same partial capture infrastructure would likely require retrofitting or de-pressurizing of the acid gases, which would affect the process efficiency. However, the CO_2 compression and intermediate storage facilities and the CO_2 shipping terminal could be used un-altered.

production paths, such as H-DR. However, partial capture can play a role in mitigating the risk of the late arrival of, for example, H-DR, and create synergies in the H-DR concepts that are proposed. Moreover, partial capture could augment CO_2 capture performance through co-mitigation with, for example, biomass, and the use of advanced solvents.

7 Conclusions

This thesis evaluates partial CO_2 capture as a CCS concept that aims to reduce the CO_2 capture cost with the example given of the iron and steel industry, and to enable a decarbonization of process industry timely with regard to the goals for climate change mitigation. The focus is on amine absorption cycles and their cost-effective design for partial capture (**Paper I**), as well as the applications of such processes to reduce the CO_2 emissions from an integrated steel mill powered by a constant (**Papers II** and **III**) or varying (**Paper A**) load of excess heat. The work is performed through process modeling of the carbon capture process integrated to a reference steel mill. From the results it can be concluded that:

- Amine absorption cycles for partial capture can be designed for CO_2 separation rates in the absorber well below 90% and for an energy-efficient operation that may lower specific capture cost (€/tonne CO_2-captured), as compared to the separation rates that are typical for full capture ($\geq$90%). The potential savings in energy and, both absolute and specific, cost may be particularly prominent for industries with concentrations of CO_2 above 20 vol.%. The thesis confirms this for partial CO_2 capture from steel mill off-gases that is powered exclusively by excess heat, as compared to full capture processes, which are (partly) powered by external energy supply.

- Variations in heat and gas supply experienced in a steel mill can be coped with by an appropriate design of the amine absorption plant without penalties to the capture performance.

- CO_2 capture from the pressurized blast furnace gas outperforms end-of-pipe capture from flue gases in terms of energy-efficiency and CO_2 capture cost. The lowest capture cost of 28 ($\pm$4) €/tonne CO_2-captured, which corresponds to a reduction of site emissions by 36%, is found for CO_2 separation from the blast furnace gas powered by excess heat from the onsite CHP plant, flue gas heat recovery from hot stoves, and the utilization of flare gases.

- Low-value excess heat significantly influences the cost structures for full and partial capture at the reference mill. For partial capture powered by excess heat, the CAPEX predominates over the steam cost. In contrast, the steam cost dominates the CAPEX for full capture owing to its reliance on external energy supply.

- Current technical limits for the CO_2 intensity of primary steel produced in blast and basic oxygen furnaces can be overcome if CCS is applied. While partial capture powered by excess heat has a lower impact on the carbon intensity, it entails a considerably lower energy penalty than full capture.

Furthermore, this thesis reveals a window of opportunity for the implementation of partial capture within the next 10–15 years or within one more investment cycle (for blast furnace relining) before full capture or carbon-free production technologies, such as hydrogen direct reduction, are required to fulfill the reduction targets envisioned for establishing the carbon-

constrained economies by Year 2050. Implementation of partial capture in the steel industry in the 2020s will be economically viable if long-term policies beyond Year 2030 ensure average carbon prices of 40–60 €/tonne CO_2 throughout the economic lifetime of the partial capture unit. In addition, partial capture may be applied together with other mitigation options (e.g., biomass), or extended to full capture in time or applied in synergy with hydrogen direct reduction in case biogenic carbon is used to deliver process heat.

To conclude, this thesis finds that partial capture of CO_2 is a readily available and potentially economically viable mitigation option for the iron and steel. Implementation before Year 2030 would likely allow for EU reduction targets for Year 2030 to be achieved, and would provide a response to the required initiation of large-scale emissions reductions in line with the targeted 1.5°C global warming limit.

7.1 Considerations for future research

The topics explored in this thesis can be extended to answer new research questions. These include:

- The discussed energy and carbon intensities of partial CO_2 capture in combination with and in comparison to other mitigation options for primary steelmaking, which could be extended to include the economic dimension. In a cost-optimizing approach, different mitigation pathways could be analyzed based on the existing plant infrastructure, investment cycles, and market conditions over time;

- A focus on process industries, which should be explored further by analyzing industries that have similar preconditions and an inherent reliance on carbon, such as petroleum refineries or cement production plants. The technical minima for carbon intensity may be overcome cost-effectively by partial capture when considering excess heat, CO_2 sources onsite, and hourly and seasonal variations;

- The placement of partial CO_2 capture from relevant process industries in the context of the energy system. The conditions under which partial CO_2 capture may become implementable from a systems perspective, and the potential effects it may have on the deployment of power-supplying technologies could be analyzed;

- The cost-efficient design of partial capture from industrial CHP plants with respect to seasonal variations and the balancing of a product portfolio that includes power, district heating, and CO_2 emissions reductions. The relationship between a design for maximum heat load (maximized CO_2 capture) and a design that captures the most cost-effective CO_2 could be explored. The entire CCS-chain onsite should be investigated, including capture, compression, and CO_2 handling (intermediate storage to buffer to ship transport). Furthermore, heat storage options to cope with seasonal variations should be investigated.

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