# Towards Sustainable Iron- and Steelmaking with Economic Optimization

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Doctor of Technology Thesis Thermal and Flow Engineering Laboratory Department of Chemical Engineering Åbo Akademi University

Turku/Åbo, Finland 2014

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## PREFACE

The work reported in this thesis was done during the years 2008-2013 at the Thermal and Flow Engineering Laboratory at Åbo Akademi University. The work was funded by Academy of Finland's Sustainable Energy "SusEn" programme and SYMBIOSIS-project. The financial support received from the Academy of Finland is gratefully acknowledged.

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I would like to thank all co-authors. Especially I want to thank Dr. Mikko Helle for all his help and support that I have received from him. I would also like to thank all my co-workers at the Thermal and Flow Engineering Laboratory at Åbo Akademi University, especially Vivéca Sundberg and Alf Hermanson for all their help. The Ruukki Steel Works at Raahe is also acknowledged for providing valuable blast furnace and steel works data.

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Hannu Helle

# SAMMANFATTNING

Järn och stålframställningsindustrin är bland de största källorna för antropogena emissioner av koldioxid i världen. Den stigande CO<sub>2</sub>-halten i atmosfären och den globala oron för växthuseffekten och klimatförändringen har lett till omfattande undersökningar om hur man kan minska energiintensiteten och CO<sub>2</sub>-utsläppen inom denna industrisektor. Denna avhandling studerar tre olika tänkbara lösningar på problemet genom matematisk modellering och optimering av ett integrerat stålverk.

I den första delen av studien undersöks möjligheten att använda biomassa som ett hjälpreduktionsmedel i masugnen. Genom att förbehandla biomassan kan dess värmevärde och kolhalt ökas samtidigt som syrehalten minskar vilket gör den bättre lämpad som reduktionsmedel. Eftersom tryckhållfastheten hos den förbehandlade biomassan är avsevärt lägre än koksens, kan biomassan inte ersätta koks i masugnsbeskickningen i stora mängder. Därför antas biomassan injiceras via formorna.

Avskiljning och lagring av koldioxid (Carbon Capture and Storage, CCS) ses idag som en möjlighet att minska utsläppen från kraftverk, men tekniken kan också användas för att minska CO<sub>2</sub>-utsläppen från ett integrerat stålverk. För en masugn kan nyttan av CCS ytterligare ökas genom att återanvända toppgasen i processen efter koldioxidstrippning. CCS påverkar dock ekonomin för det integrerade stålverket, eftersom mängden av toppgaser tillgängliga för t.ex. kraftoch värmeproduktion då minskar.

Högklassiga råvaror är en förutsättning för en jämn masugnsdrift. Speciellt kol av hög kvalitet behövs för att producera koks med önskade egenskaper för att säkra tillräcklig gaspermeabilitet och jämn masugnsdrift. Kol av lägre kvalitet samt naturgas, som vissa länder har i stora mängder, kan utnyttjas i olika direktoch smältreduktionsprocesser. Det producerade direktreducerade järnet (Direct Reduced Iron, DRI) kan i sin tur chargeras i en masugn, syrgaskonverter eller ljusbågsugn. Också processenhetens storlek och investeringskostnaderna för de alternativa framställningsprocesserna kan vara lägre än för en masugn.

I den här avhandlingen studerades ekonomin hos ett integrerat stålverk med hjälp av simulering och optimering. Systemet beskrivs av linjära modeller för enhetsprocesserna från koksverk till ståltillverkningsenheter, med en mer detaljerad termodynamisk modell för masugnen. Resultaten från studien av masugnsdrift med injektering av biomassa visade tydligt betydelsen av en ordentlig förbehandling av biomassan eftersom sammansättning, värmevärde samt utbyte påverkas av t. ex. pyrolystemperaturen. När det gäller återvinning av masugnens toppgas efter CO<sub>2</sub>-strippning, uppnås avsevärda minskningar i utsläppsnivåer om den avskilda koldioxiden kan lagras, men den optimala återvinningsgraden tillsammans med andra driftsparametrar är starkt beroende av kostnaderna för CO<sub>2</sub>-utsläpp och strippning/lagring. Vad beträffar ekonomin bakom användning av direktreducerat järn i masugn spelar prisförhållandet mellan DRI-pellets och masugnspellets en stor roll. Den höga energiåtgången i ugnen med roterande härd (Rotary Hearth Furnace, RHF) för att reducera järnmalmen leder till ökade CO<sub>2</sub>-utsläpp för stålverket som helhet, varför miljövänligheten hos konceptet främst ligger i återvinning av interna biprodukter från produktionen.

## ABSTRACT

The iron and steelmaking industry is among the major contributors to the anthropogenic emissions of carbon dioxide in the world. The rising levels of  $CO_2$  in the atmosphere and the global concern about the greenhouse effect and climate change have brought about considerable investigations on how to reduce the energy intensity and  $CO_2$  emissions of this industrial sector. In this thesis the problem is tackled by mathematical modeling and optimization using three different approaches.

The possibility to use biomass in the integrated steel plant, particularly as an auxiliary reductant in the blast furnace, is investigated. By pre-processing the biomass its heating value and carbon content can be increased at the same time as the oxygen content is decreased. As the compression strength of the pre-processed biomass is lower than that of coke, it is not suitable for replacing a major part of the coke in the blast furnace burden. Therefore the biomass is assumed to be injected at the tuyere level of the blast furnace.

Carbon capture and storage is, nowadays, mostly associated with power plants but it can also be used to reduce the  $CO_2$  emissions of an integrated steel plant. In the case of a blast furnace, the effect of CCS can be further increased by recycling the carbon dioxide stripped top gas back into the process. However, this affects the economy of the integrated steel plant, as the amount of top gases available, e.g., for power and heat production is decreased.

High quality raw materials are a prerequisite for smooth blast furnace operation. High quality coal is especially needed to produce coke with sufficient properties to ensure proper gas permeability and smooth burden descent. Lower quality coals as well as natural gas, which some countries have in great volumes, can be utilized with various direct and smelting reduction processes. The DRI produced with a direct reduction process can be utilized as a feed material for blast furnace, basic oxygen furnace or electric arc furnace. The liquid hot metal from a smelting reduction process can in turn be used in basic oxygen furnace or electric arc furnace. The unit sizes and investment costs of an alternative ironmaking process are also lower than those of a blast furnace.

In this study, the economy of an integrated steel plant is investigated by simulation and optimization. The studied system consists of linearly described unit processes from coke plant to steel making units, with a more detailed thermodynamical model of the blast furnace. The results from the blast furnace operation with biomass injection revealed the importance of proper pre-

processing of the raw biomass as the composition of the biomass as well as the heating value and the yield are all affected by the pyrolysis temperature. As for recycling of  $CO_2$  stripped blast furnace top gas, substantial reductions in the emission rates are achieved if the stripped  $CO_2$  can be stored. However, the optimal recycling degree together with other operation conditions is heavily dependent on the cost structure of  $CO_2$  emissions and stripping/storage. The economical feasibility related to the use of DRI in the blast furnace depends on the price ratio between the DRI pellets and the BF pellets. The high amount of energy needed in the rotary hearth furnace to reduce the iron ore leads to increased  $CO_2$  emissions.

# LIST OF PUBLICATIONS AND CONTRIBUTION OF THE AUTHOR

The thesis consists of this summary and the following papers as appendices:

- I. Helle, H., Helle, M., Saxén, H. and Pettersson, F., (2009) "Mathematical optimization of ironmaking with biomass as auxiliary reductant in the blast furnace", *ISIJ International* **49**, 1316-1324.
- II. Helle, H., Helle, M., Saxén, H. and Pettersson, F., (2010) "Optimization of top gas recycling conditions under high oxygen enrichment in the blast furnace", *ISIJ International* 50, 931-938.
- III. Helle, H., Helle, M., Pettersson, F. and Saxén, H., (2010) "Multiobjective optimization of ironmaking in the blast furnace with top gas recycling", *ISIJ International* 50, 1380-1387.
- IV. Helle, H., Helle, M. and Saxén, H., (2011) "Nonlinear optimization of steel production using traditional and novel blast furnace operation strategies", *Chemical Engineering Science* 66, 6470–6481.
- V. Huitu, K., Helle, H., Helle, M., Kekkonen, M. and Saxén, H., (2013) "Optimization of steelmaking using Fastmet direct reduced iron in the blast furnace", *ISIJ International* 53, 2038-2046.

The author of the thesis is the main contributor of Papers I-IV. He has been developing the models used in the papers and performed the described simulations and optimizations and written most of the papers. For Paper V the author was responsible for modeling and performing the computer simulations and interpreting the results. This paper will also be part of another dissertation (KH).

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#### **1 INTRODUCTION**

The iron and steelmaking industry is among the most energy intensive industrial sectors in the world and due to the fact that the dominating iron and steelmaking processes are still mainly coal-based and highly dependent on fossil fuels, substantial amounts of fossil carbon dioxide emissions are released. During the last decade much research has been devoted to finding potential ways to increase the efficiency and to decrease the use of fossil reductants and rate of emissions of the processes. Regardless of all the research done on the traditional iron and steelmaking units and the development of alternative ironmaking processes, this industrial sector still contributes by about 5-7 % of the world's anthropogenic  $CO_2$  emissions and the unit that consumes most energy in the chain of steelmaking, has already been largely optimized: the most efficient blast furnaces in the world are considered to operate only 5 % above the theoretical minimum in terms of reductant consumption (Kim et al. 2002; Lüngen et al. 2004; OECD/IEA 2013).

As steel is one of the most vital materials or products of today's society, it can be assumed that the global production of steel will continue its strong growth in the future. The global concern about the effects of greenhouse gases in the atmosphere has made it extremely important to explore possible methods to reduce the emissions from the steelmaking while still keeping the production of steel economically profitable. In this thesis, three different methods have been studied: the use of biomass as an auxiliary reductant in the blast furnace, blast furnace operation with top gas recycling and carbon capture and storage and the effect of using direct reduced iron in the blast furnace. In particular, their potential to reduce the  $CO_2$  emissions and the effect on the economy of the integrated steel works have been investigated with the help of mathematical simulation and optimization.

#### 2 IRONMAKING AND STEELMAKING

#### 2.1 Global situation

Global crude steel production has been rising steadily during the last ten years, excluding the decrease in 2008 and 2009 induced by the global financial crisis. In 2013, the global production was 1582 million tonnes, representing an increase of 86 % compared to 2000. The largest crude steel producer in the world is China, with a share of 49 % and a production of 779 million tonnes in 2013. As a comparison, the total crude steel produced 110 million tonnes and the USA, Russia, India and South Korea 87, 69, 81 and 69 million tonnes, respectively (World Steel Association 2014).

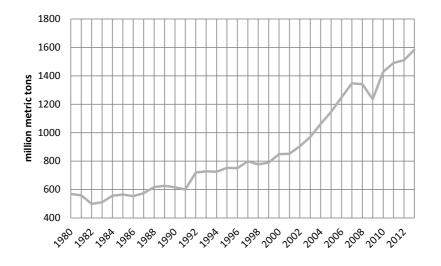


Fig. 1 World crude steel production 1980-2013.

Global scrap use in steelmaking has been around 500 million tonnes per annum during recent years. Scrap is mainly used in electric arc furnaces (EAF), the share of which of the total crude steel production was 29 % (in 2012) with a production of 452 million tonnes. Scrap is also used in basic oxygen furnaces (BOF) where it, in addition to being a source of ferrous material, works as a cooling agent. Total production of crude steel from the BOF in 2012 was 1074 million tonnes. Basic oxygen furnaces are mainly charged with the blast furnace iron (BFI), also called as pig iron, liquid iron and hot metal. The production of pig iron in 2012 was 1100 million tonnes. The alternative product of the iron ore reduction

process is called direct reduced iron (DRI); DRI production in 2012 was 74.0 million tonnes showing a small increase from the 73.3 million tonnes produced in 2011 (BIR 2013; World steel Association 2014; Midrex 2013).

#### **2.2 BF** – **BOF** route

Despite the fact that many alternative methods to reduce iron ores have been developed, some of which are also commercially available, the blast furnace continues to be the dominant process to produce iron. This is due to the cost and energy efficiency of it, partly as a result of the large size and high production rate combined with a high and uniform quality of the hot metal. The blast furnace is a combined counter current heat exchanger and chemical reactor, where the reduction of iron oxides takes place. The main product of the blast furnace is hot metal, which typically consist of about 94 % Fe, 4.5 % C, 0.5 % Si and small amounts of Mn, P, S and other metals. The temperature of the tapped hot metal is usually between 1400 °C and 1500 °C. Other main outputs from the process are slag and top gas. Slag consists of oxides of Ca, Mg, Al and Si, and its temperature is 50-100 °C higher than that of the hot metal. Slag can be used by other industries, for example, in cement making. A typical composition of the blast furnace top gas is 20-25 % CO<sub>2</sub>, 20-25 % CO, 50 % N<sub>2</sub>, 1-4 % H<sub>2</sub> and some  $H_2O$ . With a relatively low heating value (of about 3.5 MJ/m<sup>3</sup>) a part of the top gas is commonly used to preheat the blast at the hot stoves while the remaining portion is used to produce heat and power at the power plant.

After tapping, the hot metal is transferred to a mixer, where desulphurization is done. Desulphurized iron is then fed to the basic oxygen furnace, where the carbon content of the hot metal is reduced by blowing pure oxygen onto the bath. This is called primary steelmaking. Liquid steel produced in the basic oxygen furnace can be further refined to meet the desired quality in terms of chemistry and temperature, which is known as secondary steelmaking. Secondary steel processing includes for instance further desulphurization, degasification, reduction of harmful non-metallic inclusions and adjustments of composition and temperature. It can also assist the production of ultra-low-carbon steels. Once the desired composition of the steel is reached the steel is typically continuously cast into slabs, blooms or billets. After casting the steel products are taken to rolling mills. Different heat treatments can be used to improve the properties of steels as a final production stage after rolling (Ghosh et al. 2008).

#### 2.3 Biomass and charcoal utilization in the blast furnace

Biomass-derived fuels are traditionally considered as renewable fuels due to the short carbon cycle; for organic matter the carbon cycle varies from a few weeks to a few decades whereas for non-renewable fossil fuels the carbon cycle is in the order of millions of years. As the carbon released from the renewable fuels can be captured by photosynthesis in real time into new biomass, the use of biomass-derived fuels is typically assumed to be carbon neutral. However, this can only be true with sustainably produced and managed biomass sources. It is important to remember that biomass fuels are not always carbon neutral (Johnson 2009; Mathieson et al. 2012).

The use of charcoal in iron production is not a new invention; it was used as the only fuel and reducing agent in iron making before the insufficiency of wood in some countries resulted in the development of coke, first as an alternative to and later as a substitute for charcoal. Nowadays, when the mitigation of  $CO_2$  emissions is a global theme and challenge in primary steelmaking, the interest in charcoal use is rising, as it is not a fossil but a renewable source of carbon and energy.

At the moment charcoal is commonly used in blast furnaces in Brazil, where charcoal fines are injected into the blast furnace tuyeres as a substitute for pulverized coal injection. Charcoal injection rates reported are between 100 and 150 kg/tHM. The benefits of charcoal injection compared to pulverized coal are also related to the very low sulphur and phosphorous content of charcoal due to low ash content, which leads to improved hot metal quality. The specific surface area of charcoal is 60-350 times greater than the area of pulverized coal, making charcoal a highly reactive carbonaceous material. In Brazil there are also small blast furnaces where charcoal is charged from the top of the blast furnace replacing major part of the coke, but since the compression strength of the charcoal is much lower than that of coke, the height of the blast furnaces operating with partial charcoal burden is limited (Babich et al. 2010).

Suopajärvi et al. (2012a) have studied the effects of biomass use in integrated steel plant. In this simulation study the blast furnace was operated with pellets as the iron bearing material and the influence of charcoal as tuyere injectant to replace the heavy oil in the process was evaluated. Two cases, where the charcoal was either produced outside the system boundaries (Case 1) or from biomass within the system boundaries (Case 2), were simulated and compared with the reference case. The results from the investigation showed that the fossil  $CO_2$  emissions decreased by 15.4 % for Case 1 and by 26.4 % for Case 2. Other observations in the study were the lower energy content of the blast furnace top gas when charcoal was injected lowering the production of the power plant. However, the pyrolysis process where charcoal is produced creates valuable by-

products, which can be utilized in the power plant in Case 2, where the pyrolysis unit is within the system boundaries.

In Finland, the largest steel producer is Ruukki which has two blast furnaces and a total yearly maximum production of 2.6 million tonnes of hot metal. Replacing the heavy oil used as a tuyere injectant by charcoal with an injection rate of 150 kg/thm would require, depending on the moisture of the green wood, density of the wood and the pyrolysis yield, around 3 Mm<sup>3</sup>/a green biomass. In comparison in 2010 the use of forest chips in heat and power plants in Finland was 6.2 Mm<sup>3</sup> (Suopajärvi et al. 2012b).

Alternative methods to the use of charcoal in blast furnaces have also been investigated. The use of carbon iron ore composite has been suggested to increase the burden reactivity, which, in turn, controls the thermal reserve zone temperature. Experimental studies have been made where the reaction and reduction behavior of the carbon iron ore composite with biomass char has been investigated. The results showed, e.g., that the reduction of the composite with biomass char begins at lower temperatures compared to composite with coke leading to a lower temperature of the thermal reserve zone. This opens up possibilities to increase the CO utilization and decrease heat losses (Ueda et al. 2009).

In Australia, the possibility to use biomass in the iron and steel industry has been investigated by The Australian Steel Industry  $CO_2$  Breakthrough Program. Lifecycle analysis, techno-economic studies and technology reviews have been introduced for several technologies and applications including

- Biomass supply
- Pyrolysis technologies
- Sintering solid fuel
- Coke making blend component
- Steelmaking recarburization
- Blast furnace tuyere injection
- Nut coke replacement
- Reductant for pre-reduced feeds
- Replacement for charge carbon, foaming agent and steel recarburiser in EAF

For the integrated steelmaking route, consisting of blast furnace and basic oxygen furnace the  $CO_2$  emissions reduction potential is reported to be up to 32 - 58 %, with the largest potential in the replacement of the fossil tuyere injectant. However, it is stated that the biomass-derived materials are assumed to be produced in a sustainable way and the calculations are simplified in that only material substitutions on the basis of carbon content is considered. The physical properties of the charcoal as well as the effect on the operation efficiencies have been ignored (Mathieson et al. 2011).

# 2.4 Carbon dioxide capture and storage in general and in the blast furnace

The ULCOS program, which aims to reduce the  $CO_2$  emissions in iron making of today's best routes by at least 50 %, has selected four key technologies which could lead to aimed  $CO_2$  emission reductions:

- Top Gas Recycling Blast Furnace with CO<sub>2</sub> Capture and Storage (CCS)
- HIsarna with CCS
- ULCORED with CCS
- Electrolysis

Apart from the electrolysis, which requires the availability of large quantities of  $CO_2$ -free electricity, all the chosen technologies require a carbon capture and storage technology to reach the target (Meijer et al. 2009; ULCOS 2013).

## 2.4.1 Carbon dioxide capture

Carbon dioxide capture and storage (CCS) means the process, where  $CO_2$  is separated from industrial or energy-related sources, transported to a storage location and stabilized or long-term isolated from the atmosphere. The target in  $CO_2$  capture is to produce a high pressure stream with high concentration of  $CO_2$ that can be transported to a storage site. Today there are processes for separation of  $CO_2$  in large industrial scale. However,  $CO_2$  is typically separated to purify other industrial gas streams and in most cases the separated  $CO_2$  is not stored but only emitted to the atmosphere. Carbon dioxide capture systems are typically divided into the following processes (IPCC 2005):

- Post-combustion
- Pre-combustion
- Oxyfuel

Operating principles for the separation technologies are presented in Fig. 2. All the presented main capturing processes include a step, where  $CO_2$ ,  $H_2$  or  $O_2$  is separated from the bulk gas, such as flue gas, air or raw natural gas. The separation can be carried out by means of physical or chemical solvents, membranes, solid sorbents, or by cryogenic separation depending on the process conditions (IPCC 2005).

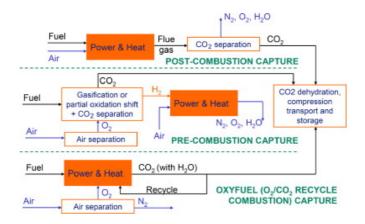


Fig. 2 Principles of three main CO<sub>2</sub> capture options (Gibbins et al. 2008).

In post-combustion systems the primary fuel is combusted with air and the  $CO_2$ is separated from the flue gases. The main component in this kind of exhaust gas is nitrogen, originating from the air, and the  $CO_2$  concentration is typically low (3) -15 % by volume). Therefore the post-combustion capture system that can be found, for example, on a modern pulverized coal or natural gas combined cycle power plants, is typically an absorption process employing powerful organic solvents such as watery solution of monoethanolamine (MEA) as the absorbent. The advantages of the post-combustion systems are related to the possibility to fit the separation unit to an existing power plant. The disadvantages of this capture method derive from the low partial pressure of CO<sub>2</sub> (13 kPa for a coal fired power plant) in the flue gas which together with the impurities  $(NO_x, SO_x and$ O<sub>2</sub>) lead to relatively high absorbent consumption and high energy requirements to regenerate the solvent. The low concentrations of CO<sub>2</sub> in the flue gases also imply that large volumes of flue gases have to be processed, which, in turn, leads to large size equipment and high capital costs. Knudsen et al. (2009) have reported that for a 1 ton/h CO<sub>2</sub> absorption pilot plant, which used 30 % MEA, the steam requirement for solvent regeneration was 3.7 GJ/ton CO<sub>2</sub> at 90 % removal and the MEA consumption was 1.4 kg/ton CO<sub>2</sub> (Pires et al. 2011; IPCC 2005; Olajire 2010).

The performance of amine absorption to separate  $CO_2$  from the conventional blast furnace top gas and nitrogen free blast furnace top gas (with shaft injection of recycled gas) has been studied by Tobiesen et al. (2007). The blast furnace gas has some important differences compared to exhaust gases from a power plant. First of all, it does not contain oxygen, which decreases the degradation of the amine absorbent. Secondly, the partial pressure of  $CO_2$  in the blast furnace gas is about 100 kPa. Therefore, amines which will not work properly at low  $CO_2$ partial pressures can be used to separate  $CO_2$  from the blast furnace gas. Typically three main classes of amines are used as chemical solvents: primary, secondary and tertiary amines. Primary amines, such as MEA, react strongly and quickly and work at low partial pressures of CO<sub>2</sub>. Secondary (e.g. diethanolamine, DEA) and tertiary (e.g. methyldiethanolamine, MDEA) amines have lower reactivity but, for instance, MDEA has typically much lower regeneration cost, lower corrosion rate and lower degradation rate compared to primary and secondary amines. Tobiesen et al. (2007) tested by simulation three different amine absorbents and found that with the best amine (50 wt% 2-amino-2-methyl-propanol), a heat requirement of 2.2 GJ/ton CO<sub>2</sub> was achieved for the treatment of the conventional blast furnace top gas with a CO<sub>2</sub> removal around 85 %. In the case of the nitrogen free top gas the CO<sub>2</sub> removal rates were higher, about 95 %, and the lowest thermal energy requirement in the regeneration unit was 2.4 GJ/ton for the solution consisting of 35 wt% MDEA/ 15 wt% piperazine. In the pre-combustion capture systems the aim is to produce a gas mixture of CO<sub>2</sub> and H<sub>2</sub>. This is achieved by processing the primary fuel in a reactor with steam and oxygen or air, resulting in a mixture consisting of mainly carbon monoxide and hydrogen, called synthetic gas ("syngas"). This process is known as gasification, partial oxidation or reforming. Syngas is next allowed to react with more steam in a catalytic reactor, called a shift reactor or converter, resulting in CO<sub>2</sub> and more H<sub>2</sub>. The CO<sub>2</sub> can then be separated from the H<sub>2</sub>, which can be used as a fuel in a gas turbine combined cycle plant. The separation can be done by absorption, adsorption or membranes. The advantage of the precombustion separation compared to the post-combustion separation lies in the higher CO<sub>2</sub> concentration and pressure in the output stream, which allows the use of different solvents with lower energy requirements for the regeneration. Also the capture equipment is smaller due to syngas conditions, but in spite of this a disadvantage of the pre-combustion separation is its high investment costs (Pires et al. 2011; IPCC 2005; Olajire 2010).

Oxyfuel combustion can be seen as a modified post-combustion separation method. In the oxyfuel combustion the fuel is combusted with pure oxygen instead of air, keeping the nitrogen out of the system, resulting in  $CO_2$  concentration exceeding 80 % in the flue gas, the rest being mainly H<sub>2</sub>O. Burning the fuel with pure oxygen leads to excessively high flame temperatures in the combustor, so some  $CO_2$  rich flue gas is typically circulated back to the combustor to decrease the temperatures. The water vapour is separated from the  $CO_2$  by cooling and compressing the flue gas stream. Advantages of the oxyfuel combustion depend on the high concentration of  $CO_2$  in the flue gas, which leads to a relatively simple separation process, minimal  $NO_x$  formation and decreased amount of flue gas that needs to be desulphurized. Furthermore, since the processes for  $O_2$  production and  $CO_2$  separation mainly rely on physical separation, chemical reagents and/or solvents are only needed in the gas desulphurization contributing to lower operational costs and environmental load.

However, cryogenic distillation, which is the main process used in oxygen production, is very expensive and energy consuming process. To decrease the energy required in the air separation unit, Burdyny and Struchtrup (2010) have proposed a hybrid system based on  $O_2/N_2$  permeable membrane followed by cryogenic distillation of this oxygen enriched air (Pires et al. 2011; IPCC 2005; Olajire 2010, Burdyny et al. 2010). In a blast furnace with top gas recycling and oxygen blast, the conditions resemble the oxyfuel combustion and CO<sub>2</sub> capture.

In the CCS-chain the  $CO_2$  capture stage is the most expensive component accounting for up to two-thirds of the total cost of a CCS project. Carbon capture also requires energy thus increasing the total emissions of the plant operating with CCS when compared to a similar plant operating without CCS. Two generally used measures for the cost of  $CO_2$  capture and storage are

$$LC_{\text{avoided}} = \frac{COE_{\text{capture}} - COE_{\text{ref}}}{E_{\text{CO2,ref}} - E_{\text{CO2,capture}}}$$
(1)

$$LC_{\text{capture}} = \frac{COE_{\text{capture}} - COE_{\text{ref}}}{CO_{2,\text{capture}}}$$
(2)

where  $LC_{avoided}$  and  $LC_{capture}$  are levelized costs of avoided or captured CO<sub>2</sub> emissions ( $\notin$ /tCO<sub>2</sub>),  $COE_{capture}$  and  $COE_{ref}$  are costs of electricity for plants with and without CCS ( $\notin$ /kWh),  $E_{CO2,ref}$  and  $E_{CO2,capture}$  are the emission rates of the reference plant and the capture plant (tCO<sub>2</sub>/kWh) and  $CO_{2,capture}$  is the amount of CO<sub>2</sub> captured (tCO<sub>2</sub>/kWh). The difference between the measures derives from the masses of CO<sub>2</sub> avoided and CO<sub>2</sub> captured. As Fig. 3 shows, the amount of CO<sub>2</sub> avoided is typically lower than the amount of CO<sub>2</sub> captured since the energy required to operate the CO<sub>2</sub> capture system increases the emissions. Therefore, numerically, the cost of CO<sub>2</sub> avoided is higher than the cost of CO<sub>2</sub> captured (IPCC 2005; Ogden et al. 2010).

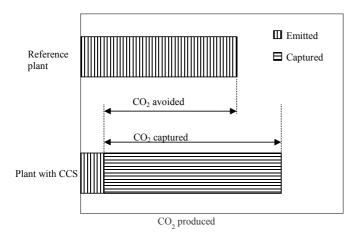


Fig. 3 CO<sub>2</sub> avoided vs. CO<sub>2</sub> captured.

#### 2.4.2 Transportation of captured carbon dioxide

Transportation of  $CO_2$  by pipeline or by ship or by tanker trucks is typically needed since commonly  $CO_2$  is not stored at the same place where it is captured.  $CO_2$  transportation is a mature technology; since the early 1970s tens of millions of tons of  $CO_2$  have been yearly transported in pipelines from natural and anthropogenic sources to sites mainly in Texas, where  $CO_2$  has been used for enhanced oil recovery (EOR). The transport in pipelines, where  $CO_2$  is typically in the form of supercritical liquid at 80-150 bar, is considered to be the most cost-efficient method to transport large volumes. Transportation with tanker ships, which can be the most economically attractive method in some situations, requires the liquefaction of  $CO_2$  and the conditions of 7 bar and -54°C (Pires et al. 2011; IPCC 2005; Haugen et al. 2009; Parfomak et al. 2008).

Costs of both pipeline and marine transportation of  $CO_2$  depend heavily on the transportation distances and quantities. In the case of pipelines, many other factors such as whether the pipeline is onshore or offshore, topography of the pipeline route and steel price, have also strong effect on the costs. In IPCC (2005) the cost of pipeline transport of  $CO_2$  for a nominal distance of 250 km was approximated to be between 1 and 8 US\$/tCO<sub>2</sub>. In the same report it is demonstrated that the shipping method becomes cheaper than pipelines for distances greater than 1000 km and for amounts smaller than few million tonnes of  $CO_2$  per year (IPCC 2005).

In Finland, the ground seems to allow no domestic carbon sequestration. This causes distances over 1000 km to the closest operational CO<sub>2</sub> storage sites, located off-shore in the North Sea and Barents Sea. Because of the long distances together with the non-existing pipeline network and the fact that most industrial sources of CO<sub>2</sub> are located around the Gulf of Finland and the Gulf of Bothnia, the pipeline transportation would be considerably more expensive than ship transportation in Finland. Kujanpää et al. (2011) approximated the costs of shipping the captured CO<sub>2</sub> to the geological formations under the North Sea to amount to  $11.8 \notin/tCO_2$ , excluding the costs for liquefaction and harbor fees.

#### 2.4.3 Carbon dioxide storage

CO<sub>2</sub> storage options are typically grouped in:

- Geological storage
- Ocean storage
- Mineral carbonation

The geological  $CO_2$  storage sites can further be divided into three alternatives: oil and gas reservoirs, which are even depleted or in combination with enhanced oil or gas recovery; saline aquifers; and unminable coal beds combined with coal bed methane recovery (IPCC 2005).

Several projects, where a geological storage of  $CO_2$  is utilized, are operated around the world. The world's first commercial CO<sub>2</sub> storage project was Sleipner in Norway, where since 1996 about 1 MtCO<sub>2</sub>/a from natural gas processing at the same site has been captured by using amine technology and stored in a deep saline reservoir, called the Utsira formation, located in the North Sea at around 900 m below the sea floor. There is no sign of  $CO_2$  leakage and the cost of injection is reported to be \$17/tCO<sub>2</sub>. In Algeria 1.2 MtCO<sub>2</sub>/a, originating from gas processing from the In Salah oil field, has been stored since 2004 at a depth of 1800 m in a depleted gas reservoir located near the gas processing plant. In this formation, which is reported to have a storage capacity of around 17 million tonnes, no CO<sub>2</sub> leakage has occurred and the injection costs are around \$6/tCO<sub>2</sub>. In total around 10 MtCO<sub>2</sub>/a is stored in geological storages at the moment. In addition around 30 MtCO<sub>2</sub> is used annually for enhanced oil recovery. For instance the Weyburn project in Canada, operated since 2000, injects 1MtCO<sub>2</sub>/a. The  $CO_2$  used for EOR is produced in the USA as a byproduct in a coal gasification and piped around 330 km to the EOR site. The EOR is assumed to extend the life time of the Weyburn field by 25 years, increasing the oil production by an additional 120-130 million barrels and ultimately 20 million tonnes of CO<sub>2</sub> is expected to be stored. However, in 2011 a report was released about the possible CO<sub>2</sub> leakage at the Weyburn field and it is still under

investigation if the leaking  $CO_2$  stems from the injected  $CO_2$  (IPCC 2005; CC&ST 2013).

 $CO_2$  can also be injected and stored in unminable coal beds. Advantages of this approach are that in addition to storing the  $CO_2$  also the amount of methane recovered from the coal bed is increased. However, this technology is still in the demonstration phase and the global storage capacity is relatively low compared to other geological storage options. Estimated storage capacities of different geological storage options are presented in Table 1 (IPCC 2005).

Table 1 Storage capacity for geological	storage options including uneconomical storage
options (IPCC 2005)	

Reservoir type	Lower estimate of storage capacity (GtCO <sub>2</sub> )	Upper estimate of storage capacity (GtCO <sub>2</sub> )	
Oil and gas fields	675	900	
Unminable coal seams (ECBM)	3-15	200	
Deep saline formations	1000	Uncertain, but possibly 10000	

Storing  $CO_2$  in the ocean by direct injection has not been demonstrated in industrial scale but it has been investigated in laboratories for 30 years and it is possible in theory. However, storing  $CO_2$  in the ocean was recently prohibited by law in Europe but especially in Japan ocean storage is still seen as a possible method. The ocean storage of  $CO_2$  involves injecting  $CO_2$  into the deep ocean at depths of more than 1000 m, where it dissolves or forms hydrates or heavier-than water plumes. According to analysis and models, the injected  $CO_2$  will be isolated from the atmosphere for hundreds of years. Since  $CO_2$  is soluble in water, there is a natural transfer of  $CO_2$  between the atmosphere and the oceans. By injecting  $CO_2$  into the water this natural uptake of around 2  $GtCO_2/a$  is accelerated. At the moment the ocean contains approximately 40000 GtC compared with 750 GtC in the atmosphere and 2200 GtC in the terrestrial biosphere. Consequently, the ocean concentration would change by less than 2 % by the amount of carbon that would double the atmospheric concentration (IPCC 2005; Pires et al. 2011; Khoo et al. 2006; Teir et al. 2011).

Mineral carbonation involves the conversion of  $CO_2$  to solid inorganic carbonates using chemical reactions. This process occurs in nature but it is very slow and therefore it needs to be accelerated considerably to make it a viable option to store the anthropogenic  $CO_2$ . Mineralization offers permanent and safe storage of  $CO_2$  which is available worldwide and has a high capacity. The main disadvantages of this method are the high costs, the slow reactions and the large amounts of raw material needed and products produced (IPCC 2005; Pires et al. 2011).

#### 2.4.4 Top gas recycling in the blast furnace

In iron and steelmaking the  $CO_2$  capture and storage would typically be connected to the idea of recycling the blast furnace top gas. The top gas recycling itself is an old idea; already in 1994 Tseitlin et al. (1994) reported results from 13 industrial-scale campaigns done at Toulachermet in Russia in the years 1985-1990. At the trials monoethanolamine was used to remove the carbon dioxide from the top gas and the  $CO_2$ -stripped top gas was heated and injected through the tuyeres together with highly oxygen enriched (cold) blast. At that time the stripped  $CO_2$  was not planned to be stored and the process was validated by the complete utilization of the carbon introduced into the process which would lead to lower coke and reductant rates.

After that the top gas recycling has been studied in many papers. Austin et al. (1998) investigated three top gas recycling methods using a mathematical model to predict the furnace performance. He found that simple blast replacement, where part of the blast is replaced with recycled top gas so that the tuyere gas volume remained constant, and oxygen enriched blast replacement, where, in addition, the tuyere gas oxygen volume is kept at constant level by enriching the remaining blast, both decrease the production rate and increase the fuel rate. However the third method, where the  $CO_2$  is stripped from the recycled top gas and both the recycled gas and the oxygen enriched blast are heated to the blast temperature, was found to be the most promising with an increase in the production of up to 25 % and a reduction in the reductant rate of 20 %.

The same two-dimensional multi-fluid blast furnace model has also been used by Nogami et al. (2005), who investigated top gas recycling with plastic injection. In this study about one third of the top gas was recycled and after  $CO_2$  removal injected into the blast furnace at the tuyere level together with pure oxygen at 25 °C under constant tuyere level conditions. This resulted in a production increase of 6 %. Other benefits of the system were the absence of hot stoves and the high caloric value of the top gas due to higher contents of hydrogen and carbon monoxide. However the coke rate and the reducing agent rate were higher compared to standard operation. This was mainly due to the constant tuyere level conditions. The relaxation of these conditions, for instance lowering the raceway temperature or increasing the bosh gas volume by higher oxygen injection rate was supposed to decrease the coke consumption and the reducing agent rate.

Nogami et al. (2006) also investigated the material and energy balances of an iron making system consisting of hot stoves, coke oven with coke dry quenching, sintering and blast furnace. In this study the blast consisting of pure oxygen and  $CO_2$ -stripped, recycled blast furnace top gas was heated in the hot stoves. It was concluded that the stripped  $CO_2$  should be stored in order to decrease the emissions from the system.

Babich et al. (2002) studied the effect of hot reducing gas injection and concluded that injection of both pulverized coal and hot reducing gas with simultaneous oxygen enrichment of the blast is an effective technology to increase the production rate and to lower the coke rate.

The effect of hot reducing gas injection was also investigated by Desai et al. (2008). In this study the reducing gas was produced either by low grade coal gasification or by recycling blast furnace top gas after  $CO_2$  and  $N_2$  removal. Theoretical investigation with a mathematical model showed that injection of hot reducing gas in the form of  $CO_2$  and  $N_2$  stripped blast furnace top gas is more effective than the injection of reducing gas generated from coal gasification and increase in productivity up to 25 % together with substantial decrease in the coke rate was reported.

In addition to the tuyere injection of the CO<sub>2</sub>-stripped blast furnace top gas, part of the gas could also be injected in the shaft. Murai et al. (2004) investigated simultaneous tuyere and shaft injection of the recycled top gas in combination with massive plastics injection and found the concept promising in terms of lowering the CO<sub>2</sub> emissions. Recently, Chu et al. (2010) numerically evaluated the blast furnace operation under top gas recycling and lower temperature operation achieved by carbon composite agglomerate charging. The simulation results showed that simultaneous shaft and tuyere injection of hot, recycled top gas increased the heat supply and enriched the reduction atmosphere in the shaft zone, improved the reduction of iron burden and enhanced the efficiency of the shaft zone. This results in a highly efficient low-carbon blast furnace with increased productivity and lower reducing agent rate. Lately, however, some concern has been expressed that the injected shaft gas would not penetrate deeply enough into the furnace. The reason for this would be that the furnace diameter is maximal at the lower shaft, and that no cohesive zone acts as a distributor of this gas.

Within the ULCOS program the recycling of hot top gas after  $CO_2$  stripping was demonstrated in practice at the experimental pilot blast furnace in Luleå, Sweden. To remove the unwanted nitrogen from the top gas the blast consisted of pure oxygen instead of preheated air and at the trials a vacuum pressure swing adsorption (VPSA) was adopted for  $CO_2$  removal. Two different top gas recycling approaches presented in Fig. 4 were tested:

- Injection of recycled gas only through main tuyeres at 1250 °C (version 3)
- Injection of recycled gas through main tuyeres at 1250 °C and through additional tuyeres located in the lower stack at 900 °C (version 4)

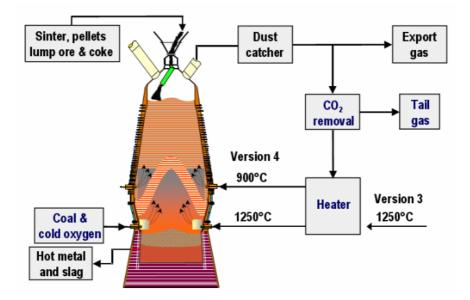


Fig. 4 Process flow of the two versions (Zuo et al. 2009).

The results from the trials were encouraging and in good agreement with earlier theoretical estimates. The blast furnace operation was stable with a smooth burden descent and good hot metal quality. The operation of the VPSA plant was also smooth providing a  $CO_2$  content of 2.67 % in the injected gas. The reductant rate decreased compared to the reference case and the  $CO_2$  emissions decreased by 76 % if the extracted  $CO_2$  is supposed to be stored (Danloy et al. 2009; Zuo et al. 2009).

Recently, Ervasto et al. (2013) have studied post combustion capture of  $CO_2$  at an integrated steel mill. In the case study, which is based on Ruukki Metals Ltd's Raahe steel plant in Finland, the effects of different capture amounts, different solvents and different heat supply options for solvent regeneration on the energy balance and emissions of the steel mill were investigated. Solvents investigated in this study were conventional MEA with regeneration energy of 3.4 MJ/kg  $CO_2$ , more advanced amino acid based solvent with regeneration energy of 2.7 MJ/kg and an imaginary future solvent which can be regenerated at a significantly lower temperature than the MEA. The results of the study showed that with the studied post combustion  $CO_2$  capture technologies it is possible to significantly cut down the greenhouse gas emissions from an integrated steel mill.

Tsupari et al. (2013) continued the study by investigating the ecomical feasibility of the post combustion  $CO_2$  capture technologies presented by Arasto et al. (2013). With an electricity price of 100  $\notin$ /MWh, a break-even price of 72  $\notin$ /ton

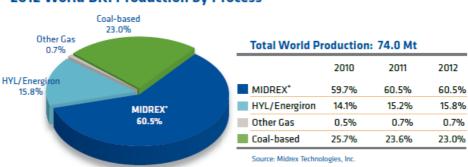
 $CO_2$  was found for  $CO_2$  emissions, where post-combustion capture with MEA became more profitable than an operation without CCS.

# 2.5 Alternative ironmaking processes

The classical steelmaking route, where iron ore is reduced in the blast furnace followed by the basic oxygen furnace where the hot metal is converted into steel, requires pre-processing (agglomeration) of both iron ore and coal. Installation costs of an integrated steel plant including pre-processing units, blast furnace and converters are high, which in turn leads to high production capacities. To overcome these problems alternative ironmaking processes have been developed (Huitu et al. 2009).

Alternative ironmaking processes are typically divided into direct reduction and smelting reduction processes. In direct reduction processes the iron ore is reduced at lower temperatures, compared to blast furnace, and in solid state. As there is no need to melt the iron ore, the carbon content of the direct reduced iron (DRI) is much lower compared to the hot metal produced in the blast furnace. On the other hand, as the reduction takes place in solid state, no slag is formed to remove the gangue materials of the charge (Huitu et al. 2009).

Direct reduction processes are typically either gas-based or coal-based. Most of the gas-based direct reduction processes use natural gas as a reducing agent which makes the processes attractive to sites which have access to cheap natural gas. A common feature for gas-based processes is the reforming of natural gas to reducing gas before it is fed into the reactor or shaft furnace, where the iron ore reduction takes place. Coal-based processes, in turn, are popular in areas with an excessive supply of cheap coal. With coal-based processes the reduction of iron ore takes place either in a rotary kiln or in a rotary hearth furnace. As seen in Fig. 5, the most dominating direct reduction process is the gas-based process Midrex and the share of coal based processes is 23 % (Huitu et al. 2009).



#### 2012 World DRI Production by Process

Fig. 5 World direct reduction production by process in 2012 (Midrex 2013).

Smelting reduction processes offer an alternative to traditional blast furnace iron making. Typically, a smelting reduction process consists of two different stages (cf. Fig. 6). In the pre-reduction unit the iron ore is partly reduced in a solid state by hot reducing gas originating from the smelting reduction vessel, where the final reduction of the ore together with the melting takes place. The investment cost of a smelting reduction process is lower compared to a blast furnace with ancillary units and it can use coal instead of coke as reductant and often also fine ores as feed material. The unit size and cost of the reactor are lower due to high temperatures required of liquid phase reactions and high smelting rates. The most popular smelting reduction process is Corex, which is currently in operation in South Africa, South Korea, India and China with an annual capacity around 5 Mt. A further developed variant is the FINEX process, which is operated by POSCO in South Korea. In this process, the prereduction takes place in fluidized beds, and the DRI is fed as hot briquetted iron into the Corex smelter-gasifier (Huitu et al. 2009; Nill 2003; Chatterjee 2010).

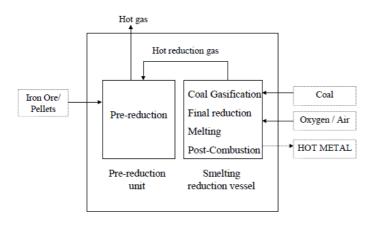


Fig. 6 Schematic of smelting reduction process (Nill 2003).

#### **3 WORK IN THIS THESIS**

This thesis consists of five papers, where the effect of alternative raw materials and novel operation strategies on the economy of an integrated steel plant is investigated by means of simulation and optimization. The papers are:

- I. Helle, H., Helle, M., Saxén, H. and Pettersson, F., (2009) "Mathematical optimization of ironmaking with biomass as auxiliary reductant in the blast furnace", ISIJ International 49, 1316-1324.
- II. Helle, H., Helle, M., Saxén, H. and Pettersson, F., (2010)
   "Optimization of top gas recycling conditions under high oxygen enrichment in the blast furnace", ISIJ International 50, 931-938.
- III. Helle, H., Helle, M., Pettersson, F. and Saxén, H., (2010) "Multiobjective optimization of ironmaking in the blast furnace with top gas recycling", ISIJ International 50, 1380-1387.
- IV. Helle, H., Helle, M. and Saxén, H., (2011) "Nonlinear optimization of steel production using traditional and novel blast furnace operation strategies", Chemical Engineering Science 66, 6470-6481.
- V. Huitu, K., Helle, H., Helle, M., Kekkonen, M. and Saxén, H., (2013) "Optimization of steelmaking using Fastmet direct reduced iron in the blast furnace", ISIJ International 53, 2038-2046.

In Paper I, the economy of the blast furnace operation with biomass injection as an auxiliary reductant is investigated. Paper II concentrates on optimization of the blast furnace operation with top gas recycling under high oxygen enrichment. Paper III continues the study presented in Paper II, but in this case the task is formulated as a multi-objective optimization problem. In Paper IV the studied system, which is slightly modified from that of Papers II and III in relation to the gas preheating, is optimized with nonlinear programming. Here, the study is not limited to only high oxygen enrichment. Finally, Paper V studies the potential of using direct reduced iron from the FASTMET process as a partial substitute for pellets in a blast furnace.

## 3.1 Motivation of work

Despite the fact that the specific energy consumption and  $CO_2$  emissions of steelmaking have been halved since 1970, the production of one ton of primary steel still requires almost 20 GJ of energy and causes at least 1.8 t of  $CO_2$  emissions. In the production chain from iron ore to primary steel the blast furnace is still the clearly dominating unit used to reduce the ore and to produce

the hot metal needed in steelmaking. Since the blast furnace and other unit processes around it, such as the coke oven and sintermaking plant, are well established and operate very close to their thermodynamic limits, the only possibility to substantially reduce the energy demand and the emissions from the steel plants in operation around the world is to find fundamentally new and innovative ways to operate the existing processes.

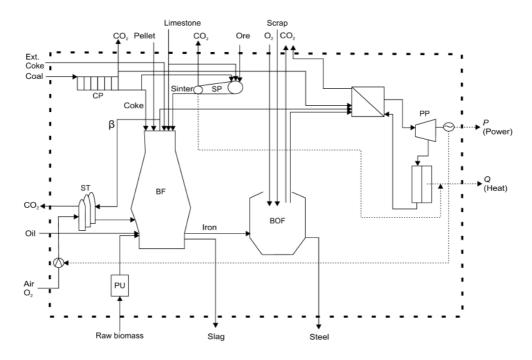
Many alternative ways to operate the blast furnace for lowering the emission rates have been suggested, e.g., by plastics injection, use of high reactivity coke to lower the reserve zone temperature, and replacing part of the coal by non-fossil reductants such as biomass. In addition, alternative methods for carbon dioxide capture and storage in steelmaking have been actively investigated. In these investigations the starting point has often been the oxygen blast furnace or highly oxygen enriched blast so that the nitrogen content of the blast furnace top gas is reduced to make carbon dioxide capturing economically and technically more attractive. Recycling this CO<sub>2</sub>-stripped top gas back to the blast furnace with tuyere or shaft injection has also often been proposed in these studies.

In the papers reported in this thesis, the effect of innovative ways of operation on the economy and emissions of an integrated steel works is studied.

#### **3.2** Flows and units for the system studied (Papers I – V)

The unit processes of the studied system are for the most part same in all the papers including a sintermaking plant (except Paper V), cokemaking plant, hot stove, blast furnace, basic oxygen furnace and power plant. In addition, in the system studied in Paper I there is a biomass pyrolysis unit for the pre-processing of the biomass to be injected, in Papers II, III and IV there is a unit for blast furnace top gas  $CO_2$  stripping and in Paper V a Fastmet-unit is included in the system to produce direct reduced iron as a raw material for the blast furnace. In Paper V also an oxygen plant and units for steel casting and rolling are included in the system boundaries but the sintermaking plant is excluded.

The material flows between the units are also in many respects the same in all the papers excluding the operation of the hot stoves: in Papers I and V a part of the blast furnace top gas is used to heat the stoves where-as in Papers II and III coke oven gas is used (because the strong recycling limits the top gas availability). In the system studied in Paper IV blast furnace top gas is used in the case of traditional blast furnace operation and coke oven gas in the cases of hot gas recycling. In all the papers the remaining parts of the blast furnace top gas and coke oven gas, together with a predetermined share of the basic oxygen furnace gas, is consumed at the power plant to produce power and heat. The systems



studied in Papers I, II and III, IV and V are presented in Fig. 7, Fig. 8, Fig. 9, and Fig. 10.

Fig. 7 System with biomass use studied in Paper I. CP: cokemaking plant, SP: sintermaking plant, ST: hot stoves, BF: blast furnace, BOF: basic oxygen furnace, PP: power plant, and PU: biomass pyrolysis unit.

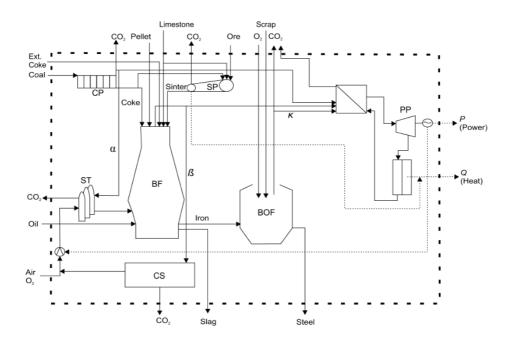


Fig. 8 System with top gas recycling and highly oxygen-enriched blast studied in Papers II and III. CS: CO<sub>2</sub> stripping unit.

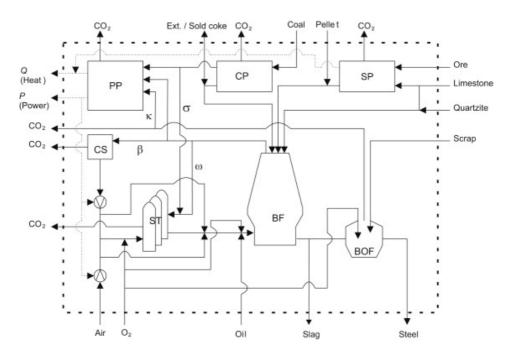


Fig. 9 System considering traditional BF to top gas BF studied in Paper IV.

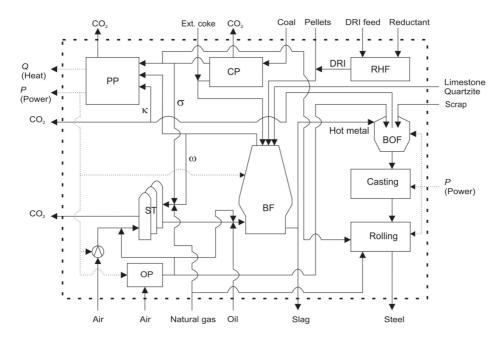


Fig. 10 System with DRI use studied in Paper V. OP: Oxygen plant, RHF: Rotary hearth furnace.

Most of the unit models are based on simple linear relationship between the inputs and the outputs of the unit in question. The relations are derived from a process data from a Finnish integrated steel works that was used as a reference for the studies. The blast furnace, in turn, is modeled in more detail.

#### 3.2.1 Coke plant

Coke has three important roles in the blast furnace: as the coke is burned in the combustion zones it supplies energy and reduction potential for the chemical reactions to occur. It also supports the burden and ensures the permeability for the gas and liquid flows, especially in the bosh, where melting of the charged ore takes place and finally it carburizes the liquid hot metal to meet the requirements of the steelmaking (Ghosh et al. 2008).

The coke used in the blast furnace is produced in coke ovens, where carbonization of the coal takes place under controlled conditions. The quality of the coke, which is affected by the quality of the chosen coal blend and its behavior during the carbonization together with the actual carbonization conditions, has a major effect on the coke rate and productivity of a blast furnace. The following characteristics are important for the coke suitable for blast furnace (Ghosh et al. 2008):

- Large enough particles not to be fluidized and carried out by the top gas
- Strong enough not to break during handling prior to charging
- Strong enough not to fall apart under the pressure created by the burden in the blast furnace
- Sufficiently inert in the upper part not to react (and degrade) at lower temperatures, but reactive enough to produce carbon monoxide at higher temperatures during its descent in the lower part of the blast furnace
- Sufficiently strong after the reaction with carbon dioxide to assure permeability in the lower parts of the blast furnace.

In addition to the coke, some valuable liquid and gaseous by-products are produced during the cokemaking. Production of one tonne of coke yields approximately (Ghosh et al. 2008):

- 750-800 kg of coke for blast furnace
- 45-90 kg of coke breeze (< 6 mm in size)
- 285-345 m<sup>3</sup>n of coke oven gas
- 27-34 litres of tar
- 55-135 litres of ammonia liquor
- 8-12.5 litres of light oil.

In all the papers (I-V) presented in this thesis the cokemaking plant is modeled with linear relations between the mass flow rate of feed coal ( $\dot{m}_{coal}$ ) and the mass flow rate of produced coke ( $\dot{m}_{coke}$ ) and the volume flow rate of purified coke oven gas ( $\dot{V}_{COG}$ ):

$$\dot{m}_{\rm coke} = 0.695 \dot{m}_{\rm coal} \tag{3}$$

$$\dot{V}_{\rm COG} = 319.7 \, \frac{\mathrm{m}^3 \mathrm{n}}{\mathrm{t}} \cdot \dot{m}_{\rm coal} \tag{4}$$

Since part of the produced coke is consumed in the sinter plant, the internal flow rate of the coke available for the blast furnace is:

$$\dot{m}_{\rm coke,\,int} = \dot{m}_{\rm coke} - \dot{m}_{\rm coke,\,sint}$$
 (5)

The cokemaking plant was given a maximum capacity of 55 t/h and if more coke is needed in the system, external coke can be bought at a fixed price. External coke is produced outside the balance boundary and thus it does not increase the  $CO_2$  emissions of the system. In Papers I, II, III and V the cokemaking plant is operated to produce only the coke needed in the system. In Paper IV the cokemaking plant is operated at maximum capacity and in cases, where all the produced coke is not used within the system, the excessive coke can be sold. This constraint was included to mimic the conditions encountered in many steelmaking plants.

#### 3.2.2 Sinter plant

The quality of the raw materials charged into the blast furnace has a major influence on the productivity and energy consumption of the furnace. Therefore iron ore is usually prepared before charging into the furnace.

Nowadays, the iron ore is normally charged in the blast furnace as a mixture of sinter, pellets and lump iron ore. Sintering and pelletizing are processes used to agglomerate iron ore fines into larger pieces. In the mixture charged into the blast furnace the share of sinter is typically at least 50 %, pellets account for around 30 % of the charge while the share of lump ore is often less than 10 %. The benefits of the sinter and pellets compared to the lump iron ore are the higher iron content, size consistency (especially with pellets) and the utilization of iron ore fines and other smaller size iron ore fractions (Ghosh et al. 2008; Biswas 1987).

In Papers I-IV the blast furnace is charged with both sinter and pellets and in Paper V only with pellets. (This reflects the transition of the reference plant from mainly sinter charging to 100 % pellet charging.) In the studies pellets are supposed to be produced outside the studied systems, while sinter is produced in the sinter plant, which is located inside the balance boundary. In the sinter plant model linear relations between the produced sinter and iron ore, coke and limestone feed are considered. The mass flow rate of produced sinter ( $\dot{m}_{sint}$ ) and the feed rates of coke and lime are expressed as

$$\dot{m}_{\rm sint} = 1.042 \dot{m}_{\rm ore} \tag{6}$$

$$\dot{m}_{\rm coke,\,sint} = 0.0460 \dot{m}_{\rm sint} \tag{7}$$

$$\dot{m}_{\rm lime,\,sint} = 0.0714 \dot{m}_{\rm sint} \tag{8}$$

In addition, a heat flow, assumed proportional to the mass flow of produced sinter, is recovered in the sinter plant:

$$\dot{Q}_{\rm sint} = 85.12 \,\mathrm{MJ/t} \cdot \dot{m}_{\rm sint}$$
 (9)

#### 3.2.3 Biomass pyrolysis unit

In Paper I, the effect of biomass injection on the blast furnace operation was studied. Biomass in the form of wood chips or logging residue could offer a non-fossil source of energy and reduction potential in the blast furnace. However, earlier simulation studies (Ueda et al. 2008; Helle et al. 2008) have shown that injection of dry wood chips, which have high oxygen content and low heating value, would result in a low flame temperature together with decreased productivity and low coke replacement ratio in the furnace. Therefore, a biomass pretreatment unit is needed, where the oxygen content of the biomass can be decreased by partial pyrolysis. This will result in increased heating value and higher carbon content, as well as a decrease in yield.

Figure 11 shows a typical dependence of yield and biomass composition on the pyrolysis temperature. The asterisks (\*) depict values for certain pyrolysis temperatures obtained from literature (Ranta 1994); the curves for the composition and yield depicted in Fig. 11 are created with piecewise cubic interpolation. The heating value of the biomass is approximated as a function of the composition as presented by Cordero et al. (2001).

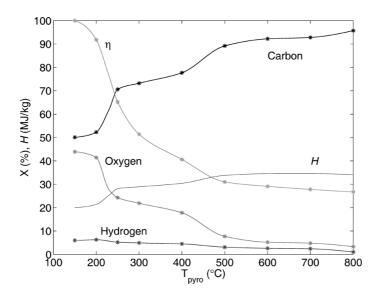


Fig. 11 Temperature dependence of key variables (yield, carbon, oxygen, and hydrogen content and heating value, *H*) in biomass pyrolysis.

The pyrolysis unit was given a maximum capacity of 15 t/h. The value is based on a gross estimate of the biomass available in the region close to the steel plant. For the sake of simplicity, no credit was given from the production of valuable by-products during the pyrolysis. Instead, the energy from the by-products was taken to be used to maintain the process conditions in the pyrolyzer.

### 3.2.4 CO<sub>2</sub> stripping unit

In the studies of Papers II, III and IV the blast furnace is operated with blast furnace top gas recycling. In these studies it is assumed that the (case-specific) part,  $\beta$ , of the top gas that is recycled, first goes to a stripping unit, where 95 % of the carbon dioxide is separated. Thus, the volume flow rate of recycled gas is  $\dot{V}_{rg} = \beta \dot{V}_{BF}$  and the mass flow rate of stripped carbon dioxide is

$$\dot{m}_{\rm CO_2,CCS} = 0.95 \dot{V}_{\rm rg} Y_{\rm CO_2,BF} \frac{44}{22.4} \frac{\rm t}{\rm m^3 n}$$
(10)

where  $Y_{CO_{2},BF}$  is the molar ratio of carbon dioxide in the blast furnace top gas.

### 3.2.5 Blast furnace

In the studies of this thesis the blast furnace unit is described in more detail by a thermodynamic first-principles model (Saxén et al. 2001; Thorn 2002) which is based on the approximations introduced by Rist and Meysson (1964). In the BF model, which is especially useful in the evaluation of the use of different injected reducing agents with a known composition and heating value, the furnace is divided into two main control volumes, the preparation zone (PZ) and elaboration zone (EZ), with the reserve zone on the boundary between these (cf. Fig. 12). The BF model is based on mass and energy balance equations and the assumption that a thermal and chemical reserve zone, where the gas and solids have practically the same temperature, exists in the furnace shaft. The temperature of the gas and solids in the reserve zone is assumed to be known, and the composition of the ascending gas is assumed to approach equilibrium with the descending iron oxides that are taken to be in the form of wüstite (FeO<sub>1.05</sub>). Furthermore, in the combustion zone in front of the tuyeres the model assumes adiabatic conditions and that the carbon and hydrogen entering the zone leave it only as CO and H<sub>2</sub>. With given blast parameters and specific injection rates of auxiliary reductants the temperature and flow of the combustion gases, i.e., the adiabatic flame temperature and the bosh gas volume and composition can be determined.

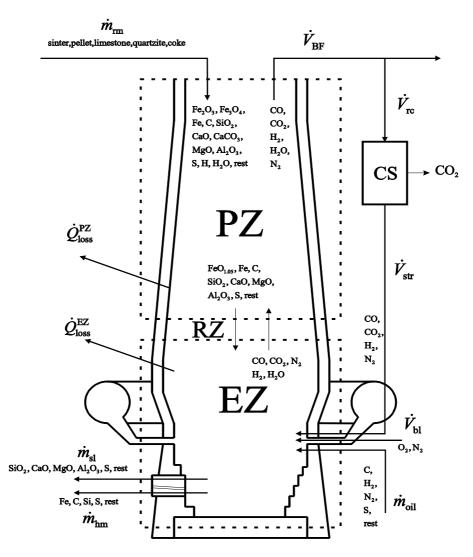


Fig. 12 Flows and components considered in the BF model.

The accuracy of the blast furnace model has been evaluated by Thorn (2002). A comparison between the practical data from the steel plant and the results from the blast furnace model showed good correlation.

### 3.2.6 Hot stoves

Cowper stoves, together with turbo-blowers constitute the hot stove unit, where the blast is heated before it enters the blast furnace. The stove is made of bricks, it has a cylindrical form with a height around 30 m and a diameter of around 6-8 m. Stoves are operated in a cyclic manner: during the heating cycle, the blast

furnace gas or some other gas from the steel plant is burnt with air in the stove combustion chamber, from where the hot gases are led through hundreds of channels formed between the bricks to release most of their heat to the checkerwork. Within 1-2 hours the bricks are heated with the hot flue gases, the combustion is stopped, and compressed air is blown through the stove in the opposite direction and becomes heated through the contact with the hot bricks. This cooling cycle of the stove takes about 1 hour and because of this a minimum of three stoves is required in the system (Ghosh et al. 2008).

In the studies presented in this thesis the cyclic operation of the hot stoves is neglected and the stoves are assumed to operate as a single continuous counter current heat exchanger in steady state, with a performance mimicking that of the stove system.

The pressure of the blast is first increased in compressors to exceed the pressure prevailing in the furnace at the tuyere level. In the compressor model, the blast is approximated as an ideal gas, and the true power requirement is given by

$$P_{\rm compr} = \dot{n}_{\rm bl} (H_{m,1} - H_{m,0}) = \dot{n}_{\rm bl} (H'_{m,1} - H_{m,0}) / e \tag{11}$$

with

$$H'_{m,1} = H_m(p_{\rm bl}, S_{m,0}); \ T'_1 = T(p_{\rm bl}, H_{m,1}); \ S_{m,0}$$
  
=  $S_m(p_0, T_0)$  (12)

After isentropic compression from ambient conditions  $(T_0, p_0)$  to the blast pressure  $(p_{bl})$ , the molar enthalpy  $(H'_{m,1})$  can be determined. The true power requirement  $(P_{compr})$  can be calculated by using an efficiency factor (e) and finally the gas temperature after compression  $(T'_1)$  is solved. This gas is next mixed with the added oxygen and the temperature of the mixture  $(T_1)$  is solved from an energy balance equation. The heat transfer in the stoves is expressed as

$$\dot{Q}_{\rm st} = G_{\rm st} \Delta T_{\rm ln} \tag{13}$$

where  $G_{st}$  is the heat conductance of the stoves and  $\Delta T_{ln}$  is the logarithmic mean temperature difference from the "hot" to the "cold" side

$$\Delta T_{\rm ln} = (\Delta T_1 - \Delta T_2) / \ln(\Delta T_1 / \Delta T_2) \tag{14}$$

with  $\Delta T_1 = T_{g1} - T_1$  and  $\Delta T_2 = T_{g2} - T_2$  referring to the temperature difference at the two ends of the stoves, and subscript g refers to the combustion gas. The (combustion) gas inlet temperature ( $T_{g2}$ ) depends on the composition of the combustion gas and on the air excess ratio. The temperatures  $(T_{g1}, T_2)$  of the exiting gases and the heat flow  $(\dot{Q}_{st})$  are solved from the energy balance equations for the heat accepting and heat releasing sides together with the transport equation (13). By tuning the heat conductivity  $(G_{st})$  of the hot stove system the overall performance of the model is adjusted in agreement with observed values of the hot blast temperature  $(T_2)$  and the off-gas temperature  $(T_{g1})$  in the steel works used as reference in the study under traditional blast furnace operation.

In the studies of the papers of the thesis, the preheating of the blast in the hot stoves is assumed to be undertaken with different gases. In Paper I, the hot stoves are operated in the traditional way with blast furnace top gas. In Papers II and III, where the blast furnace is operated with top gas recycling, the amount of blast furnace top gas available for the stoves is not necessarily sufficient, and hence the stoves are assumed to be heated with coke oven gas. In Paper IV, depending on the conditions of the studied case, both blast furnace top gas and coke oven gas is assumed to be available for the stoves.

In Papers II and III it is also assumed that both the recycled blast furnace top gas after  $CO_2$  stripping and the strongly oxygen enriched blast are mixed, compressed and preheated in hot stoves even though in practice the compression and preheating should be performed separately requiring two set of hot stoves and compressors. Therefore in Paper IV two alternative preheating methods were studied, which both would require only one set of hot stoves: either the blast is heated and the recycled top gas is added without heating or the recycled top gas is heated and only cold oxygen is injected, in which case the blast oxygen content is fixed at 99 vol-%. The amount of top gas, coke oven gas or the mixture of these needed for the combustion in the stoves is determined so as to reach the desired blast temperature.

### 3.2.7 Power plant

Co-generation, also called combined heat and power production, refers to the simultaneous generation of both power and useful heat. Higher energy efficiency of the co-generation systems leads to savings of 10-40 % in the fuel consumption (Madlener et al. 2003).

In all the papers power  $(P_{pp})$  and heat  $(\dot{Q}_{pp})$  are produced in the combined heat and power plant. The power plant unit is modeled to combust a mixture of the remaining blast furnace top gas and coke oven gas together with a part ( $\kappa$ ) of the basic oxygen furnace gas with a given air excess. This assumption is made to get a realistic image of the CO<sub>2</sub> emissions; in reality the gases are often used in other parts of the integrated plant, e.g., in slab reheating furnaces and for ladle preheating. The generated high temperature heat is used to produce steam at high pressure for a turbine, with given efficiency factors for the turbine and the generator. The low pressure steam is condensed and the released heat is used for district heat production. In all the papers the power requirements for raw material transportation, cooling water pumping, etc. are neglected and the net power production of the studied system is

$$P = P_{\rm pp} - \sum_{i} P_{\rm cons, i}$$
<sup>(15)</sup>

where *i* is the other consumers of the electricity in the steel plant. The power and heat from the power plant are simply determined as

$$P_{\rm pp} = \alpha \gamma \dot{E}_{\rm pp} \tag{16}$$

$$\dot{Q}_{\rm pp} = (1 - \alpha) \gamma \dot{E}_{\rm pp} \tag{17}$$

with

$$\dot{E}_{\rm pp} = \dot{V}_{\rm COG, pp} h_{\rm COG} + \dot{V}_{\rm BF, pp} h_{\rm BF} + \kappa \dot{V}_{\rm BOF} h_{\rm BOF}$$
(18)

In the equations  $\alpha$  (= 0.2) is the distribution factor of energy between power and heat,  $\gamma$  (=0.83) is the overall efficiency of the power plant,  $h_{COG}$ ,  $h_{BF}$ , and  $h_{BOF}$ are the effective heating values of the coke oven gas, the blast furnace top gas and the basic oxygen furnace gas,  $\dot{V}_{COG,pp}$  and  $\dot{V}_{BF,pp}$  are the remaining parts of the coke oven gas and blast furnace top gas available in the power plant, and  $\kappa$  (= 0.5) describes the part of the basic oxygen furnace gas recovered to the power plant. For the case of simplicity, the values of  $\alpha$ ,  $\gamma$  and  $\kappa$  have been assumed constant in this work.

### 3.2.8 Basic oxygen furnace

In the basic oxygen furnace the hot metal produced in the blast furnace is converted into liquid steel (expressed with the mass flow rate symbol  $\dot{m}_{\rm ls}$  in the thesis), which is the main output from the studied system in Papers I-IV. For the sake of simplicity, all additives except scrap are neglected and the scrap is assumed to be pure Fe. Simple linear relations are used to describe the mass

flow rate of liquid steel and the volume flow rates of consumed oxygen and produced off-gas:

$$\dot{m}_{\rm ls} = 0.8953\dot{m}_{\rm hm} + \dot{m}_{\rm scrap} \tag{19}$$

$$\dot{V}_{O_2,\text{BOF}} = 45.62 \frac{\text{m}^3 \text{n}}{\text{t}} \cdot \dot{m}_{\text{hm}}$$
 (20)

$$\dot{V}_{\rm BOF} = 41.48 \frac{{\rm m}^3 {\rm n}}{{\rm t}} \cdot \dot{m}_{\rm hm} \tag{21}$$

In all the papers the use of scrap is fixed at  $\dot{m}_{\rm scrap} = 0.25 \dot{m}_{\rm hm}$ .

### 3.2.9 Casting and rolling

In Paper V, the liquid steel from the basic oxygen furnace is also cast and rolled. In the casting unit the steel losses are estimated to be 5 % and in rolling 6 %. The carbon dioxide emissions from the casting and rolling are perceived by the carbon balance of the system (26). The final product from the system in Paper V is thus rolled steel, described by  $\dot{m}_{steel}$  in the thesis.

#### 3.2.10 Oxygen plant

In oxygen production the oxygen is separated from air typically with either distillation, adsorption or membranes, from which the distillation is the most mature technology allowing both high purities and large scale productions (Burdyny et al. 2010).

In papers I-IV the oxygen used in the system was assumed to be bought. In Paper V an oxygen plant is included in the system, where the oxygen needed in the blast furnace and in the basic oxygen furnace is produced. The power consumption in the oxygen plant is described as

$$P_{\rm op} = 0.3 \frac{\rm MWh}{\rm t_{0_2}} \dot{m}_{0_2} \tag{22}$$

where the energy requirement 0.3 MWh/t  $O_2$  is in line with the values presented by Burdyny and Struchtrup (2010) for a medium sized oxygen production by cryogenic distillation.

#### 3.2.11 CO<sub>2</sub> emissions from the studied system

Carbon dioxide emissions from the alternative systems presented in the papers can be expressed as a function of the difference between the input and the output of carbon of the system. In Paper I, the elements entering the system which contain carbon are coking coal, limestone, oil for tuyere injection, possibly the external coke and biomass whereas the only carbon output besides the carbon dioxide is the carbon in the liquid steel after the basic oxygen furnace. Thus, the total mass outflow rate of the carbon dioxide can be expressed as

$$\dot{m}_{\rm CO_2} = \frac{44}{12} \left( \dot{m}_{\rm coal} X_{\rm C,coal} + \dot{m}_{\rm lime} X_{\rm C,lime} + \dot{m}_{\rm oil} X_{\rm C,oil} + \dot{m}_{\rm coke,ext} X_{\rm C,coke} + \dot{m}_{\rm bio} X_{\rm C,bio} - \dot{m}_{\rm ls} X_{\rm C,ls} \right)$$
(23)

where  $X_{C,i}$  expresses the mass fraction of carbon in component *i*. The total inflow of limestone is the sum of the limestone used in the sinter making and the limestone charged directly into the blast furnace.

In Papers II-V the carbon dioxide emissions from the system are calculated in a similar way. In the studies of Papers II and III the blast furnace is operated with the possibility of top gas recycling and without biomass injection. Thus the carbon dioxide emissions from the systems in Papers II and III can be estimated by a balance equation

$$\dot{m}_{\rm CO_2} = \frac{44}{12} \left( \dot{m}_{\rm coal} X_{\rm C,coal} + \dot{m}_{\rm lime} X_{\rm C,lime} + \dot{m}_{\rm oil} X_{\rm C,oil} + \dot{m}_{\rm coke,ext} X_{\rm C,coke} - \dot{m}_{\rm ls} X_{\rm C,ls} \right) - \dot{m}_{\rm CO_2,str}$$
(24)

where  $\dot{m}_{CO_2,str}$  describes the amount of carbon dioxide which is stripped and stored.

By contrast to the treatment in these papers, Paper IV assumes that the coke plant always operates at full capacity, occasionally creating situations where more coke is produced than is needed in the blast furnace and in the sintermaking. In these cases the excess coke is assumed to be sold, which creates one more carbon outflow compared to Papers I-III:

$$\dot{m}_{\rm CO_2} = \frac{44}{12} \left( \dot{m}_{\rm coal} X_{\rm C,coal} + \dot{m}_{\rm lime} X_{\rm C,lime} + \dot{m}_{\rm oil} X_{\rm C,oil} \right. \\ \left. + \dot{m}_{\rm coke,ext} X_{\rm C,coke} - \dot{m}_{\rm coke,exp} X_{\rm C,coke} \right.$$

$$\left. - \dot{m}_{\rm ls} X_{\rm C,ls} \right) - \dot{m}_{\rm CO_2,str}$$

$$(25)$$

For the case of simplicity, the carbon content of the bought and sold coke is assumed to be equal.

In Paper V the total mass outflow rate of carbon dioxide is expressed as

$$\dot{m}_{\rm CO_2} = \frac{44}{12} \left( \dot{m}_{\rm coal} X_{\rm C,coal} + \dot{m}_{\rm lime} X_{\rm C,lime} + \dot{m}_{\rm oil} X_{\rm C,oil} \right. \\ \left. + \dot{m}_{\rm coke,ext} X_{\rm C,coke} + \dot{m}_{\rm red} X_{\rm C,red} + \dot{m}_{\rm ng} X_{\rm C,ng} \right.$$
(26)  
$$\left. - \dot{m}_{\rm steel} X_{\rm C,ls} \right)$$

where  $\dot{m}_{red}$  describes the mass flow of the reductant used in a rotary hearth furnace and  $\dot{m}_{ng}$  is the mass flow of natural gas used in the rolling unit.

#### 3.2.12 Linearization of the blast furnace model

The blast furnace model, described in detail in the Appendix of Paper IV, is in Papers I-III linearized in order to simplify the treatment and to speed up the solution of the optimization problem. The more detailed blast furnace model was run extensively under a large number (>300 000) of input combinations, with input variables uniformly distributed within their admissible regions. Independent variation of model inputs in the simulation gives rise to simulated cases with infeasible furnace states such as excessive flame temperatures or negative top gas temperatures. Infeasible solutions were removed from the data set and the remaining feasible solutions and corresponding input combinations were used to develop the linearized blast furnace model.

The selection of the input variables of the linearized model is based on the study presented by Thorn (2002), where the blast furnace model run under normal operation, i.e. without biomass injection or top gas recycling, was linearized. In the study the presented  $R^2$ -values showed that the output values predicted by the linearized model correspond well with the values calculated with the blast furnace model.

In Paper I, the linear model is expressed as a function of eight input variables:

$$y_{i} = K_{i,0} + K_{i,1} \frac{\dot{V}_{air}}{km^{3}n/h} + K_{i,2} \frac{\dot{V}_{O_{2},BF}}{km^{3}n/h} + K_{i,3} \frac{m_{oil}}{kg/t hm} + K_{i,4} \frac{T_{bl}}{^{\circ}C} + K_{i,5} \frac{m_{pel}}{kg/t hm} + K_{i,6} \frac{m_{lime,BF}}{kg/t hm} + K_{i,7} \frac{O_{bio}}{kg/t hm} + K_{i,8} \frac{E_{bio}}{MJ/t hm}$$

$$i = 1, ..., 13$$
(27)

Table 2 and Table 3 present all the input quantities of the system in Paper I together with the 13 central output variables, y, predicted by the linearized model and the limits for the variables.

Table 2 Input quantities and feasible regions for Paper I.

Variable	Symbol	Unit	Range
Blast volume	$\dot{V}_{\rm bl}$	km <sup>3</sup> n/h	0-140
Oxygen volume	$\dot{V}_{O_2,\mathrm{BF}}$	km <sup>3</sup> n/h	0-40
Specific oil rate	$m_{ m oil}$	kg/t hm	0-122
Blast temperature	$T_{\rm bl}$	°C	850-1100
Specific pellet rate	$m_{ m pel}$	kg/t hm	0-600
Specific limestone rate	$m_{ m lime.BF}$	kg/t hm	0-100
Specific biomass injection	$m_{ m bio}$	kg/t hm	0-120
rate			
Blast oxygen	$Y_{O_2,bl}$	%	21-32
Sinter	$\dot{m}_{ m sint}$	t/h	0-160
Pyrolysis temperature	T <sub>pyro</sub>	°C	150-800
Own coke	$\dot{m}_{\rm coke,int}$	t/h	0-55
Biomass feed rate	$\dot{m}_{ m bio}$	t/h	0-15

Table 3 Predicted output variables and their constraints for Paper I.

Variable	Symbol	Unit	Range
Production rate	$\dot{m}_{ m hm}$	t hm/h	120-160
Specific coke rate	$m_{ m coke.BF}$	kg/t hm	$\geq 0$
Flame temperature	$T_{\mathrm{fl}}$	°C	2000-2300
Top gas temperature	T <sub>BF</sub>	°C	100-250
Bosh gas volume	$\dot{V}_{ m bg}$	km <sup>3</sup> n/h	170-200
Residence time of solids	τ	h	6.0-9.0
Slag basicity, $X_{Ca0}/X_{Si0_2}$	В	-	1.00-1.2
Slag rate	$m_{ m slag}$	kg/t hm	$\geq 0$
Top gas volume	$\dot{V}_{ m BF}$	km <sup>3</sup> n/h	$\geq 0$
Top gas CO content	Y <sub>CO,BF</sub>	%	$\geq 0$
Top gas CO <sub>2</sub> content	$Y_{\rm CO_2,BF}$	%	$\geq 0$
Top gas H <sub>2</sub> content	$Y_{\rm H_2,BF}$	%	$\geq 0$
Heating value of top gas	H <sub>BF</sub>	MJ/m <sup>3</sup> n	$\geq 0$

The last two variables of the linear model,

$$O_{\rm bio} = m_{\rm bio} X_{\rm O,bio} \tag{28}$$

$$E_{\rm bio} = m_{\rm bio} H_{\rm bio} \tag{29}$$

express the specific inflows of oxygen and energy with the biomass. These variables are used as inputs in the model instead of pyrolysis temperature since they characterize the undesired and desired properties of the biomass and

represent asymptotic features having a value of zero when no biomass is used. That said, the pyrolysis temperature influences the model implicitly through its effect on the oxygen content, the yield and the heating value of the biomass. The  $R^2$ -values comparing the values predicted by the linearized model and the values calculated by the blast furnace model are very high ( $R^2>99$  %) for all the other output variables except the top gas temperature ( $R^2=96.2$  %). In Papers II and III the linearized blast furnace model is

$$y_{i} = K_{i,0} + K_{i,1} \frac{\dot{V}_{rg}}{km^{3}n/h} + K_{i,2} \frac{\dot{V}_{bl}}{km^{3}n/h} + K_{i,3} \frac{Y_{O_{2},bl}}{\%} + K_{i,4} \frac{m_{oil}}{kg/t hm} + K_{i,5} \frac{T_{bl}}{\circ C} + K_{i,6} \frac{m_{pel}}{kg/t hm} + K_{i,7} \frac{m_{lime,BF}}{kg/t hm} \quad i = 1, ..., 14$$
(30)

Table 4 and Table 5 show the 7 input variables and the 14 output variables of the linearized model and their constraints together with the two other constraints. The  $R^2$ -values are on a similar level as in the study in Paper I.

Table 4 Input variables and their constraints for Papers II and III.

Variable	Symbol	Unit	Range
Recycled top gas	<i>V</i> <sub>rg</sub>	km <sup>3</sup> n/h	70-250
Blast volume	, V <sub>bl</sub>	km <sup>3</sup> n/h	0-140
Blast oxygen	$Y_{0_2,bl}$	%	80-99
Specific oil rate	$m_{ m oil}$	kg/t hm	0-200
Blast temperature	$T_{\rm bl}$	°C	850-1100
Specific pellet rate	$m_{ m pel}$	kg/t hm	0-600
Specific limestone rate	$m_{ m lime,BF}$	kg/t hm	0-100

Variable	Symbol	Unit	Range
Production rate	$\dot{m}_{ m ls}$	t ls/h	150-196
Specific coke rate	$m_{\rm coke.BF}$	kg/t hm	$\geq 0$
Flame temperature	$T_{\rm fl}$	°C	1900-2300
Top gas temperature	$T_{\rm BF}$	°C	100-250
Bosh gas volume	$\dot{V}_{ m bg}$	km <sup>3</sup> n/h	150-200
Residence time of solids	τ	h	6.0-9.0
Slag basicity, $X_{CaO}/X_{SiO_2}$	В	-	1.00-1.2
Slag rate	$m_{ m slag}$	kg/t hm	$\geq 0$
Top gas volume	, V <sub>BF</sub>	km <sup>3</sup> n/h	$\geq 0$
Top gas CO content	Y <sub>CO,BF</sub>	%	$\geq 0$
Top gas CO <sub>2</sub> content	$Y_{\rm CO_2,BF}$	%	$\geq 0$
Top gas H <sub>2</sub> content	Y <sub>H2,BF</sub>	%	$\geq 0$
Top gas N <sub>2</sub> content	Y <sub>N2,BF</sub>	%	$\geq 0$
Heating value of top gas	H <sub>BF</sub>	MJ/m <sup>3</sup> n	$\geq 0$
Sinter	$\dot{m}_{ m sint}$	t/h	0-160
Own coke	$\dot{m}_{ m coke,int}$	t/h	0-55

Table 5 Predicted output variables and their constraints, as well as sinter and coke mass production rate constraints for Papers II and III.

# 3.2.13 Optimization problem and objective function

In the analysis with a single objective (Papers I, II, IV and V) the optimization problem of finding the optimal state of operation of the system is tackled by minimizing the specific costs of liquid steel production, where the emissions costs are included. In Paper III, the conflicting goals of reducing both the production costs and the emissions are formulated as a multi-objective optimization problem. In all the papers the economic objective function is expressed in specific terms, i.e. per ton of (liquid) steel.

In Papers I and II, where the blast furnace model is linearized, the minimization is performed with respect to the input variables of the linearized model by nonlinear programming (Floudas 1995). In paper I the specific costs of producing liquid steel are

$$\frac{F}{\mathbf{E}/\mathrm{t}\,\mathrm{ls}} = \left(\frac{\dot{m}_{\mathrm{ore}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{ore}}}{\mathbf{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{pel}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{pel}}}{\mathbf{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{coal}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{coal}}}{\mathbf{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{coal}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{coal}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{coal}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{oil}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} + \frac{\dot{m}_{\mathrm{coal}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{oil}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{oil}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} + \frac{\dot{m}_{\mathrm{lime}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{bio}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{bio}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{bio}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{bio}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{bio}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{bio}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{bio}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{bio}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{bio}}}{\mathrm{E}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{bio}}}{\mathrm{t}/\mathrm{t}} + \frac{\dot{m}_{\mathrm{bio}}}}{\mathrm{t}/\mathrm{t}} +$$

In the objective function  $c_i$ , presented in Table 6, express cost terms per unit. It should be noted that the last two terms, power and district heat, have negative signs since instead of being costs they have a decreasing effect on the overall costs. However, no credit is given to the production of slag, and no operation or investment costs are included in the objective function. The total inflow of pure oxygen takes into account both the oxygen used for the oxygen enrichment of the blast and the oxygen used in the basic oxygen furnace. Furthermore, the outflow of district heat includes both the district heat from the power plant as well as from the sinter plant:

$$\dot{V}_{0_2} = \dot{V}_{0_2,\text{BF}} + \dot{V}_{0_2,\text{BOF}} \tag{32}$$

$$\dot{Q}_{\rm dh} = \dot{Q}_{\rm PP} + \dot{Q}_{\rm sint} \tag{33}$$

To determine the share of carbon dioxide emissions originating from fossil fuels, a factor  $\psi$  is calculated from the carbon inputs:

$$\psi = \frac{\dot{m}_{\text{coal}} X_{\text{C,coal}} + \dot{m}_{\text{coke,ext}} X_{\text{C,coke}} + \dot{m}_{\text{lime}} X_{\text{C,lime}} + \dot{m}_{\text{oil}} X_{\text{C,oil}}}{\dot{m}_{\text{coal}} X_{\text{C,coal}} + \dot{m}_{\text{coke,ext}} X_{\text{C,coke}} + \dot{m}_{\text{lime}} X_{\text{C,lime}} + \dot{m}_{\text{oil}} X_{\text{C,oil}} + \dot{m}_{\text{bio}} X_{\text{C,bio}}}$$

In Paper II there are only few differences in the objective function compared to Paper I:

- The terms  $\frac{\dot{m}_{bio}}{t/h} \cdot \frac{c_{bio}}{\epsilon/t}$  and factor  $\psi$  have been removed
- Cost term  $\frac{\dot{m}_{CO_2,str}}{t/h} \cdot \frac{c_{str}}{\epsilon/t}$  has been added

Thus, the objective function in Paper II can be presented as:

$$\frac{F}{\not{\epsilon}/t \, \text{ls}} = \left(\sum_{i} \dot{m}_{i} \, c_{i} + y\right) / \frac{\dot{m}_{\text{ls}}}{t \, \text{ls/h}}$$

$$i = \{\text{ore, pel, coal, ext. coke, oil, lime, scrap, CO_{2}, CO_{2} \, \text{str}} \}$$
(34)

(-, r, r, ..., r, ..

where  $y = \dot{V}_{0_2}c_{0_2} - P \cdot c_{el} - Q_{dh}c_{heat}$ , which are the common terms present in all the objective functions.

In Paper III the problem is tackled as a multi-objective optimization problem. The objective function  $F_1$  is similar to the objective function in Paper II but the cost term created by carbon dioxide emissions is omitted. The second objective function  $F_2$ , is simply the specific carbon dioxide emissions.

$$\frac{F_1}{\epsilon/t \, \text{ls}} = \left(\sum_i \dot{m}_i c_i + y\right) / \frac{\dot{m}_{\text{ls}}}{t \, \text{ls/h}}$$
(35)  
$$i = \{\text{ore pel coal ext coke oil lime scrape CO1 str}\}$$

 $i = \{ \text{ore, pel, coal, ext. coke, oil, lime, scrap, CO<sub>2</sub> str } \}$ 

 $F_2 = \dot{m}_{\rm CO_2} / \dot{m}_{\rm ls} \tag{36}$ 

By formulating the optimization problem as a multi-objective problem it is possible to study a system with two conflicting objectives, such as the specific steel production costs and the specific emission rate, separately. The nondominated solutions for the problem can be presented as a Pareto-optimal frontier, which displays the solutions where it is not possible to improve one objective without worsening the other. In this case the frontier is determined with the  $\varepsilon$ -constraint method, where the multi-objective optimization problem is reformulated so that one of the objectives is optimized while the other objective is restricted to only take certain values. In other words, the existing optimization problem is reformulated as

$$\min F_1 \tag{37}$$
  
$$s. t. F_2 \le \varepsilon$$

where  $\varepsilon$  is the largest allowed value for  $F_2$ . The procedure to form the Pareto frontier begins by choosing such a large value for  $\varepsilon$  that  $F_2$  is not restricted at all, which means that the problem is equal to only minimizing  $F_1$ . Non-dominated points along the frontier are obtained by gradually decreasing the value of  $\varepsilon$  and solving the next optimization problem. This results in a set of slightly improved values of  $F_2$  together with increased value of  $F_1$ . The procedure is repeated until  $\varepsilon$  reaches such a low value that new feasible solutions cannot be found and the Pareto frontier is formed from the collected solutions.

Cost terms  $c_i$  used in objective functions in Papers I-III are identical and are reported in Table 6.

	Papers I, II and III	Paper IV	Paper V
Core	80 €/t		-
c <sub>pel</sub>	100 €/t	120 €/t	
C <sub>coal</sub>	145 €/t		
C <sub>coke,ext</sub>	300 €/t		
C <sub>coke,exp</sub>	-	250 €/t	-
C <sub>oil</sub>	150 €/t		
$c_{\rm lime}$	30 €/t		
<i>C</i> <sub>quartz</sub>	-	30 €/t	
<i>c</i> <sub>02</sub>	50 €/km <sup>3</sup> n		-
C <sub>scrap</sub>	100 €/t		
$c_{\rm el}$	50 €/MWh		
C <sub>heat</sub>	10 €/MWh		
Cng	-		200 €/km <sup>3</sup> n
Cred	-		90/200/343 €/t

Table 6 Costs in the objective functions in Papers I-V.

Price levels of the biomass ( $c_{bio}$  in Paper I) and the carbon dioxide emission ( $c_{CO_2}$  in Papers I and II) together with the costs from carbon dioxide stripping and storing ( $c_{str}$  in Papers II and III) are varied to study the effect of the changing costs on the specific cost of steel in the optimal solutions.

In Papers IV and V the optimization problem is tackled by nonlinear programming (Floudas 1995). In Paper IV the objective function is almost identical with the objective function of Paper II; the only differences are the cost term  $\dot{m}_{\rm quartz}$  due to consumption of quartzite in the process and the credit term  $\dot{m}_{\rm coke,exp}$  resulting from the operation of the coke plant at full capacity, which leads to situations where excess coke is produced.

$$\frac{F}{\notin/\mathrm{t}\,\mathrm{ls}} = \left(\sum_{i} \dot{m}_{i}\,c_{i} + y - \frac{\dot{m}_{\mathrm{coke,exp}}}{\mathrm{t}/\mathrm{h}} \cdot \frac{c_{\mathrm{coke,exp}}}{\notin/\mathrm{t}}\right) / \frac{\dot{m}_{\mathrm{ls}}}{\mathrm{t}\,\mathrm{ls}/\mathrm{h}}$$

$$i$$

$$= \{\mathrm{ore, pel, coal, ext. coke, oil, lime, scrap, quatz, CO_{2}, CO_{2}, \mathrm{str}\}}$$
(38)

The cost terms  $c_i$  used in Paper IV are mostly the same as in previous papers but the price of pellets has been updated (cf. Table 6). The optimization problem is solved for a given hot metal production rate with respect to the remaining input

variables of the blast furnace model reported in Table 7.

Input/Output	Variable	Symbol	Unit	Range
Ι	Production rate	$\dot{m}_{ m ls}$	t <sub>ls</sub> /h	150-190
Ι	Recycled top gas	<i>V</i> <sub>rg</sub>	km <sup>3</sup> n/h	$\leq$ 220
Ι	Blast oxygen	$Y_{O_2,bl}$	vol-%	21-99
Ι	Specific oil rate	$m_{ m oil}$	kg/t hm	0-120
Ι	Blast temperature	$T_{\rm bl}$	°C	250-1200
Ι	Specific pellet rate	$m_{ m pel}$	kg/t hm	0-600
0	Blast volume	$\dot{V}_{\rm bl}$	km <sup>3</sup> n/h	$\geq 0$
0	Specific coke rate	$m_{\rm coke.BF}$	kg/t hm	$\geq 0$
0	Flame temperature	$T_{\rm fl}$	°C	1900-2300
0	Top gas temperature	$T_{\rm BF}$	°C	115-250
0	Bosh gas volume	, V <sub>bg</sub>	km <sup>3</sup> n/h	150-250
0	Solid residence time	τ	h	6.0-9.5
0	Slag rate	$m_{ m slag}$	kg/t hm	$\geq 0$
0	Top gas volume	$\dot{V}_{\rm BF}$	km <sup>3</sup> n/h	$\geq 0$
0	Top gas CO content	Y <sub>CO,BF</sub>	vol-%	0-100
0	Top gas CO <sub>2</sub> content	$Y_{\rm CO_2,BF}$	vol-%	0-100
0	Top gas H <sub>2</sub> content	$Y_{\rm H_2,BF}$	vol-%	0-100
0	Top gas N <sub>2</sub> content	$Y_{\rm N_2,BF}$	vol-%	0-100
0	Heating value of top gas	H <sub>BF</sub>	MJ/m <sup>3</sup> n	$\geq 0$
0	Gas compression power	Pcompr	MW	$\geq 0$

Table 7 Blast furnace input variables, some output variables and their constraints as presented in Paper IV.

In Paper V the objective function is expressed as

$$\frac{F}{\notin/\text{t steel}} = \left(\sum_{i} \dot{m}_{i} c_{i} + y + \frac{\dot{V}_{\text{ng}}}{\text{km}^{3}\text{n/h}} \cdot \frac{c_{\text{ng}}}{\notin/\text{km}^{3}\text{n}}\right) / \frac{\dot{m}_{\text{steel}}}{t_{\text{steel}}/\text{h}}$$

$$i = \{\text{pel, coal, ext. coke, oil, lime, scrap, quatz, CO2, DRIf, red}\}$$
(39)

where DRIf refers to the DRI feed material and red to the reductant used in the rotary hearth furnace. In this paper the specific operation cost of rolled steel production is optimized at different operation rates, emission allowance costs and DRI feed costs with respect to the inputs of the blast furnace model presented in Table 8.

Input/Output	Variable	Symbol	Unit	Range
Ι	BF production rate	$\dot{m}_{ m hm}$	t <sub>hm</sub> /h	127-166
Ι	Specific DRI rate	$m_{ m DRI}$	kg/t <sub>hm</sub>	0-400
Ι	Blast oxygen	$Y_{O_2,bl}$	vol-%	21-32
Ι	Specific oil rate	$m_{ m oil}$	kg/t <sub>hm</sub>	0-120
Ι	Blast temperature	$T_{\rm bl}$	°C	850-1200
0	Specific coke rate	$m_{\rm coke.BF}$	kg/t <sub>hm</sub>	$\geq 0$
0	Flame temperature	$T_{\rm fl}$	°C	1850-2300
0	Top gas temperature	$T_{\rm BF}$	°C	115-250
0	Bosh gas volume	$V_{\rm bg}$	km <sup>3</sup> n/t <sub>hm</sub>	0.90-1.97
0	Solid residence time	τ	h	6.0-9.5
0	Slag rate	$m_{ m slag}$	kg/t <sub>hm</sub>	≥175

Table 8 Blast furnace input variables, some output variables and their constraints as presented in Paper V.

## **4 OPTIMIZATION RESULTS**

# 4.1 Biomass injection

In the study of biomass injection (Paper I) the target of the optimization task is to analyze the effect of biomass injection on the economy of the production of liquid steel in an integrated steel works. The composition of the biomass together with the yield and the heating value were found to be very temperature dependent; therefore, also the effect of pyrolysis temperature on the system is investigated to find the optimal pre-processing conditions. The system is optimized for steel production rates between 140 and 185 t ls/h. The latter value was found to be close to the upper feasibility limit of the studied system, since for higher production rates no feasible solutions within the constraints could be found. The uncertainty in the future biomass and emission prices was taken into account by optimizing the system using different price levels of the raw biomass ( $c_{\text{Dio}} = 20-110 \text{ €/t}$ ) and emissions ( $c_{\text{CO}_2} = 0-60 \text{ €/t}$ ).

The optimization of the system revealed local minima in the price of steel at three different levels of biomass pre-processing: two intermediate temperatures close to 250°C and 425°C and the temperature of 150°C corresponding to the case with no pyrolysis but only heating and drying of the biomass. In addition, the investigation revealed that the solution at the global optimum always requires either pre-processing the biomass until it reaches the intermediate temperature or operation without biomass injection. High heating value and carbon content achieved by pre-processing the biomass at the maximum temperature of 800°C could not compensate for the high loss in yield to make that a solution.

The minimum costs of steel production for the investigated biomass and emissions price combinations as well as for the operation without biomass injection are shown in Fig. 13. In the figure the solid lines represent the operation with biomass injection and the dashed lines depict biomass-free operation. It can be noticed that for most of the presented cases the steel production cost reaches a minimum at a steel production rate around 150 t ls/h when the blast furnace is operated with biomass injection. For operation without biomass injection the steel production cost increases steadily over the whole production rate range. The reason for the increasing steel production costs is the changes in the optimal blast furnace operation. Especially the increase in the pellet rate along the increasing steel production rate raises the steel production costs.

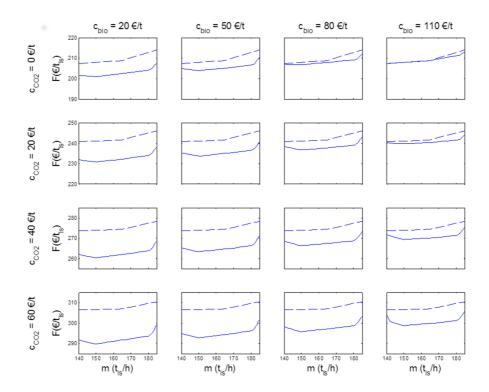


Fig. 13 Optimal steel production cost for steel production rates in the range 140-185 t/h for different price levels of raw biomass and emissions. Solid lines: steel production cost with biomass injection. Dashed lines: price for biomassfree production.

Figure 13 illustrates that cases with low biomass price combined with high emission cost show the largest economic advantage in biomass injection, but also some less extreme combinations seem economically feasible. However, for the cases with high biomass price combined with low cost of emissions, there is some economic advantage only at the higher production rates of steel while at the low production rates the economic benefit from the biomass injection is close to zero.

For most cases the optimal biomass feed rate to the pyrolysis process corresponding to the optimal solutions shown in Fig. 13 is at maximum. This leads after yield loss to specific biomass injection rates with decreasing trend from 82 kg/t hm at 140 t ls/h to around 63 kg/t hm at a steel production rate of 180 t ls/h. As the highest production rates are reached the specific injection rate typically drops due to either the lower feed rate or a higher pyrolysis temperature followed by lower yield.

The biomass injection is typically economically most beneficial at higher production rates; for instance, the case with a moderate biomass price of 80 €/t and a carbon dioxide emission cost of 20 €/t leads to a price advantage of 6.1 €/t ls for biomass injection at a production rate of 180 t ls/h. Therefore it is useful to investigate the optimization results by keeping the production rate fixed at 180 t ls/h and varying the biomass price and emission costs.

The results from this investigation are shown in Fig. 14, which illustrates the optimal feed rate of biomass, the pyrolysis temperature and the price difference in the liquid steel production costs for the operation without and with biomass injection. From the figure it can be seen that whenever it is optimal to inject biomass the pyrolysis temperature is always around 250°C. Quite naturally, when the biomass price is low, it is always optimal to use the highest allowed biomass feed rate (15 t/h), which after the yield loss in the pre-processing gives a specific biomass injection rate of approximately 62 kg/t hm. When the price of biomass is increased, a threshold value is encountered, where the optimal feed rate of biomass suddenly drops to around 10.5 t/h. This is the point of operation, where the use of coke from the own coke plant becomes more economical than the biomass injection and the coke plant starts to operate at maximum capacity. When the biomass price is further increased by 35-50 €/t the optimal biomass injection rate drops to zero giving the point of operation where the use of external coke becomes more economical than the use of biomass injection. Values for other process variables around the threshold values of biomass price for an emission price of  $c_{CO_2} = 20 \notin t$  and for  $\dot{m}_{ls} = 180 t/h$  can be seen in Table 10. The above-mentioned critical threshold values can be approximated as

$$c_{\text{bio}}^{*} = \begin{cases} 0.603 \, c_{\text{coal}} + 1.632 \, c_{\text{CO2}} + 0.009 \, \dot{m}_{\text{ls}} & \text{if } \dot{m}_{\text{ls}} \le 170 \, \frac{\text{t}}{\text{h}} \\ 1.464 \, c_{\text{coal}} + 1.573 \, c_{\text{CO2}} - 0.696 \, \dot{m}_{\text{ls}} & \text{if } \dot{m}_{\text{ls}} > 175 \, \frac{\text{t}}{\text{h}} \end{cases}$$
(40)

$$c_{\text{bio}}^{**} = 0.443 c_{\text{coke,ext}} + 1.362 c_{\text{CO2}} + 0.024 \dot{m}_{\text{ls}} \text{ if } \dot{m}_{\text{ls}} \ge 170 \frac{\text{t}}{\text{h}}$$
 (41)

In other words, if the emissions are free, at low steel production rates the biomass price should not exceed 60 % of the price of the coking coal for the biomass injection to be economical with respect to the use of own coke. At high production rates the biomass price should be somewhat lower. After the first threshold value is exceeded the biomass injection remains economically feasible as long as the biomass price is less than 44 % of the price of external coke. For all cases the threshold value is raised by around 1.4-1.6  $\notin$ /t for every 1  $\notin$ /t

increase in the CO<sub>2</sub> emission cost. Table 9 presents a summary of the critical biomass prices. For each case, for biomass prices lower than  $c_{bio}^*$  it is optimal to use the maximal biomass feed rate. For biomass prices higher than  $c_{bio}^*$  but lower than  $c_{bio}^{**}$  the biomass feed rate drops due to the maximal use of own coke. For biomass prices higher than  $c_{bio}^{**}$  the use of external coke becomes more economical compared to biomass injection.

$\dot{m}_{ m ls}$ (t/h)	170				180			
<i>c</i> <sub>CO2</sub> (€/t)	0	20	40	60	0	20	40	60
c <sup>*</sup> <sub>bio</sub> (€/t)	89	122	154	187	87	118	150	181
c*** bio (€/t)	137	164	191	219	137	164	192	219

Table 9 Threshold values for biomass,  $c_{\text{coal}}=145 \text{ }$ /t and  $c_{\text{coke,ext}}=300\text{ }$ /t.

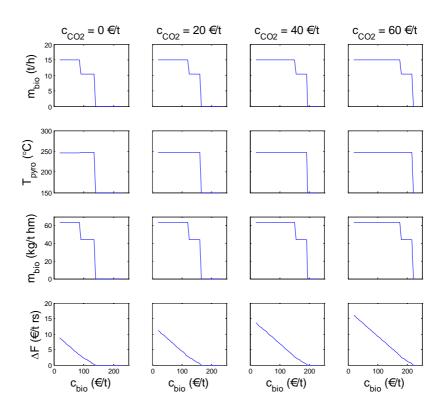


Fig. 14 Optimal biomass feed rate, pyrolysis temperature, specific biomass injection rate and price difference between operation with and without biomass for a liquid steel production of 180 t/h.

Table 10 Process variables for  $\dot{m}_{\rm ls}$ =180 t/h,  $c_{\rm CO2}$  = 20 €/t and  $c_{\rm bio}$  = 100 €/t, 150 €/t or 200 €/t. Sinter flow, blast temperature, bosh gas volume, and oil rate are at their upper limits, while slag basicity is at its lower limit (cf. Table 2 and Table 3). Values at constraints are given in bold.

Biomass price (€/t)	100	150	200
Production rate (t ls/h)	180	180	180
Blast volume (km <sup>3</sup> n/h)	126.9	128.2	133.3
Blast oxygen content (%)	32.0	31.3	29.8
Coal rate (t/h)	76.3	79.1	79.1
Coke from coke plant (t/h)	53.0	55.0	55.0
Internal coke rate (kg/t hm)	290.7	303.1	303.1
External coke rate (kg/t hm)	-	-	28.2
Limestone rate (kg/t hm)	14.8	16.1	19.2
Biomass feed rate (t/h)	15.0	10.5	-
Biomass injection rate (kg/t hm)	63.5	44.0	-
Pyrolysis temperature (°C)	247	247	-
Flame temperature (°C)	2119	2137	2179
Residence time (h)	7.2	7.0	6.6
Slag rate (kg/t hm)	205	207	211
Top gas CO (%)	26.6	26.2	25.3
Top gas CO <sub>2</sub> (%)	25.4	25.3	24.8
Top gas H <sub>2</sub> content (%)	9.5	9.0	8.0
Top gas temperature (°C)	147	141	125
Top gas heating value (MJ/m <sup>3</sup> n)	4.70	4.59	4.34
Share of BF gas to stoves (%)	35.2	35.9	38.1
Production costs (€/t ls)	240.6	244.1	245.1
Spec. fossil emissions (t $CO_2/t$ ls)	1.49	1.54	1.61

# 4.2 Blast furnace top gas recycling

Paper II investigates the concept of recycling the  $CO_2$ -stripped blast furnace top gas back into the furnace through (lower) tuyeres under a massive oxygen enrichment of the blast, and its effect on the process conditions and production economy of the steel plant. The system with top gas recycling was found to have

6 % higher maximum production than conventional operation; thus the system is optimized for steel production rates between 150 and 195 t/h, 196 t ls/h being the maximum production rate under the present process constraints.

Since the emission price  $c_{CO_2}$  and the price for CO<sub>2</sub> stripping and storage,  $c_{str}$ , were found to have a marked effect on the optimal top gas recycling degree, the cases shown in Table 11 were chosen to illustrate the optimal system.

Case	<i>c</i> <sub>CO2</sub> (€/t)	<i>c</i> <sub>str</sub> (€/t)	<i>c</i> <sub>heat</sub> (€/MWh)
А	0	0	10
В	20	0	10
С	0	20	10
D	20	20	10
Е	40	20	10
F	40	20	30

Table 11 Cases studied in Paper II.

In Case A the costs of CO<sub>2</sub> emissions and storage/stripping are zero. The optimal price of liquid steel together with the corresponding blast volume and oxygen content, the blast furnace top gas recycling degree ( $\beta$ ), the oil injection rate and the rate of reducing agents are shown in Fig. 15. It can be noticed that the steel price decreases consistently and the blast oxygen enrichment remains at its lower limit throughout the investigated production rate range. The optimal values for other parameters act in a more complex way. For the production rates between 150 t ls/h and 175 t ls/h the optimal top gas recycling degree decreases slowly from about 80 % to 75 %. The blast volume and the oil injection rate increase slowly and linearly from 30 km<sup>3</sup>n/h and 62 kg/t hm to 38 km<sup>3</sup>n/h and 125 kg/t hm while the rate of reducing agents remains relatively constant at around 345 kg/t hm. When the production rate increases to 180 t ls/h the parameters undergo more substantial transitions: the optimal top gas recycling degree drops from around 75 % to below 40 %, the blast volume increases by about 10 km<sup>3</sup>n/h and the oil injection rate and the rate of reducing agents both increase by 50 kg/t hm. The sudden change in the optimal values of the parameters taking place in the production rates 175-180 t ls/h is the result of a more economical solution, which is infeasible for the production rates under 180 t ls/h due to a too low top gas temperature, becoming feasible. Increasing the costs of stripping and storage to 20 €/t (Case C) yields optimal states relatively similar with Case A and for the production rates over 180 t ls/h identical values with Case A are obtained. The production cost of steel is about 10  $\epsilon$ /t higher at the lower production rates, compared to Case A, decreasing to  $5 \notin t$  higher at the maximum production rate. Also Case F ( $c_{CO_2}$ =40 €/t,  $c_{str}$ =20 €/t,  $c_{heat}$ =30 €/MWh) leads to optimal state of operation identical with the Case A.

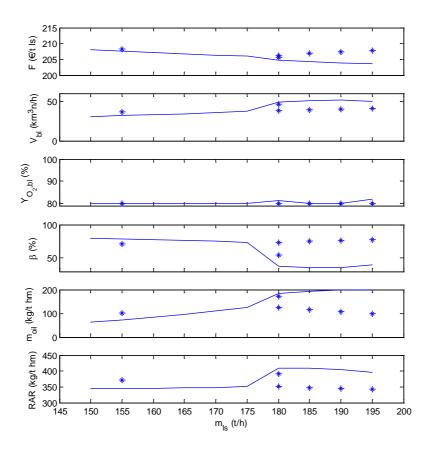


Fig. 15 Optimal price of liquid steel (*F*), blast volume  $(\dot{V}_{bl})$  and oxygen content( $Y_{O_2, bl}$ ), degree of BF top gas recycled ( $\beta$ ), specific oil injection rate ( $m_{oil}$ ) and specific rate of reductants (RAR) vs. steel production rates for  $c_{CO2} = c_{str}$ = 0  $\notin$ /t (Case A). Variables corresponding to local minima are depicted by asterisks.

In Case B the stripping and storage cost is at zero but the emissions cost is increased to 20  $\notin$ /t. This results in an increase of 12-15  $\notin$ /t in the price of liquid steel. For the production rates of 150-190 t ls/h the top gas recycling degree is at its theoretical maximum at 98.6 %, where the remaining 1.4 % is needed to purge the nitrogen from the system and the oxygen enrichment is maximum at 99 %. The blast volume stays at a low level increasing monotonically with the production rate and the oil rate and the RAR also remain at low levels: the oil rate varies between 40 and 60 kg/t hm while the RAR decreases slowly from 320 to 310 kg/t hm. In conclusion compared to Case A the cost combination investigated in Case B with  $c_{CO_2}=20 \notin/t$  but  $c_{str}=0 \notin/t$  leads to an optimal solution

with significantly lower emissions achieved at the maximum top gas recycling (Fig. 16). Case E ( $c_{CO_2}$ = 40 €/t,  $c_{str}$ =20 €/t) gives practically identical results with Case B, with the exception in the steel price which rises to 244-246 €/t ls.

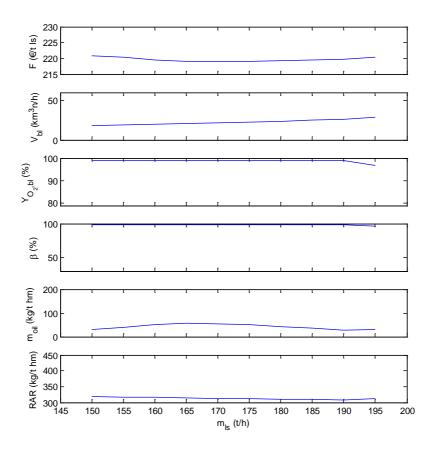


Fig. 16 Optimal price of liquid steel (*F*), blast volume  $(\dot{V}_{bl})$  and oxygen content( $Y_{O_2, bl}$ ), degree of BF top gas recycled ( $\beta$ ), specific oil injection rate ( $m_{oil}$ ) and specific rate of reductants (RAR) vs. steel production rates for  $c_{CO2} = 20$  $\notin/t$  and  $c_{str} = 0 \notin/t$  (Case B).

The most complex transitions in the parameters between the optimal states of the operation are achieved in Case D with equal costs of emissions and stripping/storage of  $20 \notin/t$ . Except the steel production costs, which slowly decrease from  $235 \notin/t$  to around  $230 \notin/t$  as the production rate increases, the variables have major transitions at production rates of 165-170 t ls/h and 185-190 t ls/h. At low production rates it is optimal to operate the system with maximum top gas recycling together with 99 % oxygen enrichment of the blast, but at 170 t

ls/h the optimal top gas recycling degree decreases to around 75 % and the oxygen enrichment drops to its minimum (80 %). At 190 t ls/h the optimal recycling degree drops further to about 55 %. The initially low blast volume increases with the production rate from 19 km<sup>3</sup>n/h to 50 km<sup>3</sup>n/h with larger steps at the above-mentioned points. The oil injection rate and the RAR follow similar patterns as the blast volume. The specific oil injection rate is only around 30 kg/t hm at the production rate of 150 t ls/h and it increases throughout the investigated range of production rate, with larger transitions at the named points ending close to its maximum value; the RAR follows a similar trend but at low level, increasing from 320 kg/t hm to 380 kg/t hm (Fig. 17).

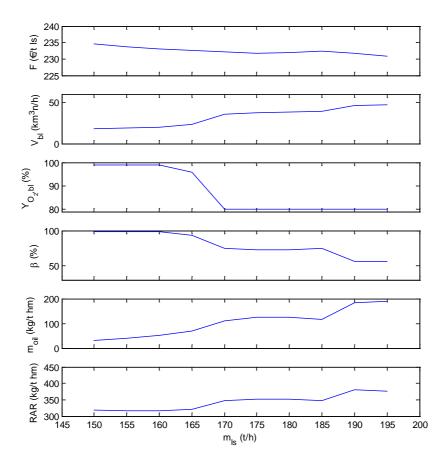


Fig. 17 Optimal price of liquid steel (*F*), blast volume  $(\dot{V}_{bl})$  and oxygen content( $Y_{O_2, bl}$ ), degree of BF top gas recycled ( $\beta$ ), specific oil injection rate ( $m_{oil}$ ) and specific rate of reductants (RAR) vs. steel production rates for  $c_{CO2} = c_{str}$ = 20 €/t (Case D).

# 4.3 Multi-objective optimization of top gas recycling

Paper III presents an alternative to the approach of Paper II to investigate the system: the problem with conflicting goals of reducing both the production costs and the emissions is formulated as a multi-objective optimization problem. The system is optimized for the steel production rates in the range 150-180 t ls/h, with steps of 10 t ls/h.

The case studied first can be seen as a limiting ideal case with zero costs of  $CO_2$  stripping and storage. The Pareto frontier derived from the optimization is presented in Fig. 18, which shows that the specific production costs vary in the range 206-228  $\notin$ /t ls (horizontal axis) while the emission rate is between 0.45 and 0.8 t CO<sub>2</sub>/t ls (vertical axis). The specific CO<sub>2</sub> emissions are about 1 t CO<sub>2</sub>/t ls lower than in the case of the traditional blast furnace operation (cf. Table 10) demonstrating that the top gas recycling with CCS is an efficient method for decreasing the CO<sub>2</sub> emissions from the steelmaking.

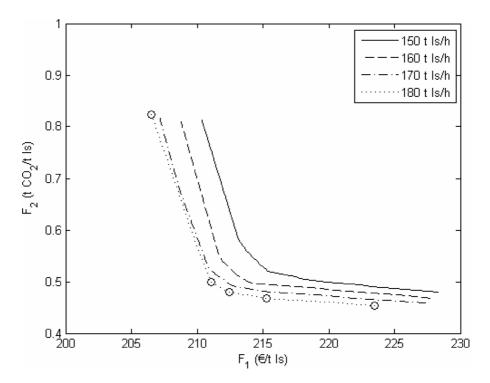


Fig. 18 Pareto frontiers for minimization of specific costs ( $F_1$ ) and specific emission rate ( $F_2$ ) at steel production rates of 150, 160, 170 and 180 t ls/h, for  $c_{str} =$ 0  $\in$ /t. Key process variables at the points indicated by circles are analyzed in Table 12.

Figure 18 also shows that the frontier moves left and downwards when the production rate is increased. This means that both objectives can be reduced by increasing the production rate. In addition, it can be noticed that for every production rate there is a clear conflict between the two objectives: at first there is a large drop in the specific emissions when the production costs are increased from their lowest value which then turns into a slower and more linear decrease. Figure 19 shows the behavior of some of the key variables of the system along the points on the Pareto frontiers. It can be noticed that at the minimum costs both the recycling degree,  $\beta$ , and the oxygen enrichment of the blast are at low levels but both increase rapidly to their maximum values. The increased injection of recycled gas is compensated for by reduced blast volume and oil rate. At the point where both the recycling degree,  $\beta$ , and the oxygen enrichment of the blast reach their maximum values, another method is needed to further decrease the specific CO<sub>2</sub> emissions. This is achieved by decreasing the ore rate while using more pellets, which means that part of the emissions are shifted outside the studied system to the external pellet producer. However, this transition further increases the production cost of steel. Finally, external coke is introduced, which again shifts part of the emissions outside the plant but also strongly affects the costs

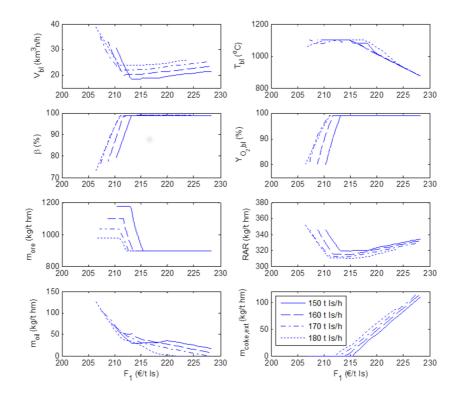


Fig. 19 Central variables for the states of the Pareto frontiers of Fig. 18: Blast volume, blast temperature, top gas recycling degree, blast oxygen content, specific ore rate, total rate of reductants, oil rate and external coke rate.

The Pareto frontiers presented in Fig. 18 experience some slope changes corresponding to changes in the state of operation of the system. For the highest steel production rate of 180 t ls/h these points have been marked by circles and the values of some of the key variables at these points are presented in Table 12. From the table the changes in the state of the operation can be noticed. At point 1, where the production costs are at their lowest level, the blast oxygen enrichment is close to its lower limit, the top gas recycling degree is about 73 % and the sinter plant is working at full capacity. No external coke is used and the oil rate is 125 kg/t hm. Moving right on the Pareto frontier to point 2, the top gas recycling degree and the blast oxygen content are increased to their maximum values. This increases the production costs only by 4.5  $\in$ /t ls while the rate of reductants, the oil rate and the specific emissions decrease considerably. At the next point a part of the emissions are "outsourced" to the pellet producer by raising the pellet rate to its maximum and also some external coke is needed. In consequence the specific emissions decrease by 20 kg/t ls while the production

costs increase only by 1.4  $\notin$ /t ls. Moving right on the Pareto frontier to the next point, more internal coke is replaced by external coke, raising the steelmaking costs by 2.9  $\notin$ /t ls and decreasing the emissions by 12 kg/t ls. At the last point studied the blast temperature has decreased from 1100 °C to 964 °C and oil is no longer used. The use of external coke has increased to 87 kg/t hm, which has reduced the specific emissions by another 14 kg/t ls but the steelmaking costs have increased by 8.2  $\notin$ /t ls.

Table 12 Key process variables for a steel production rate of  $\dot{m}_{\rm ls}$ =180 t/h and zero CO<sub>2</sub> stripping and storage costs at the five points indicated by circles in Fig. 18.

Variable	Point o	n Pareto frontier (Fig. 18)			
variable	1	2	3	4	5
Steel production costs (€/t ls)	206.5	211.0	212.4	215.3	223.5
Specific emissions (kg $CO_2/t$ ls)	824	500	480	468	454
Blast volume (km <sup>3</sup> n/h)	38.7	23.9	23.9	34.0	26.0
Specific blast volume $(m^3n/t hm)$	246	152	152	216	165
Blast oxygen content (%)	80.1	98.9	99.0	99.0	99.0
Top gas recycling degree (%)	73.3	98.6	<b>98.8</b>	<b>98.9</b>	<b>98.9</b>
Sinter feed rate (t/h)	160.0	160.0	146.7	146.7	146.7
Specific sinter rate (kg/t hm)	1018	1018	934	933	933
Specific pellet rate (kg/t hm)	522	522	600	600	600
RAR (kg/t hm)	352	311	310	310	321
Specific rate of hot reducing gas (m <sup>3</sup> n/t hm)	625	805	817	829	846
Specific coke rate, bought (kg/t hm)	0	0	2	21	87
Specific oil rate (kg/t hm)	125	44	35	22	0
Specific captured CO <sub>2</sub> (kg/t hm)	529	797	813	831	857
Blast temperature (°C)	1057	1100	1100	1100	964
Flame temperature (°C)	1900	1900	1900	1900	1905
Bosh gas volume (km <sup>3</sup> n/h)	193	190	191	192	195
Solid residence time (h)	8.79	8.09	7.93	7.70	7.06
Top gas volume (km <sup>3</sup> n/h)	192	193	196	199	204
Top gas temperature (°C)	159	130	105	101	100
Top gas CO (%)	37.5	45.8	46.0	46.4	48.2
Top gas $CO_2$ (%)	31.7	35.2	35.4	35.5	35.8
Top gas $H_2$ (%)	15.7	9.7	8.6	7.0	4.5
Top gas $N_2$ (%)	15.1	9.2	10.0	11.2	11.5
Top gas CO utilization, $\eta_{CO}$ (%)	45.8	43.4	43.5	43.4	42.6
Slag rate (kg/t hm)	196	202	204	216	214
Slag basicity, (CaO)/(SiO <sub>2</sub> ) (-)	1.00	1.00	1.00	1.11	1.04
Heat flow ratio in shaft (-)	0.87	0.90	0.93	0.93	0.93
Carbon input with coal (kg/t ls)	248.6	285.2	287.4	281.8	251.6
Carbon input with bought coke (kg/t ls)	0	0	1.8	16.1	66.4
Carbon input with oil (kg/t ls)	94.7	33.2	26.5	16.6	0
Carbon input with limestone (kg/t ls)	8.2	8.6	9.7	11.8	10.7
Carbon output with steel (kg/t ls)	1	1	1	1	1
Carbon capture (kg/t ls)	126.0	189.9	193.7	198.0	204.3
Carbon emissions (kg/t ls)	224.5	136.1	130.6	127.2	123.5

The effect of higher costs of  $CO_2$  stripping and storage can be seen in Fig. 20 and Fig. 21, where the Pareto frontiers are seen to move towards higher specific costs and emissions. For the higher  $c_{str}$  the initial decrease in the emissions from around 0.9 t  $CO_2/t$  ls to around 0.5 t  $CO_2/t$  ls is accompanied with a more considerable cost increase.

Figure 21 illustrates the states along the Pareto frontier for  $c_{str}=40 \text{ } \text{e/t}$  for the same key variables as in Fig. 19. The trends of the lines are generally similar to those already discussed for Fig. 19. The largest changes caused by the introduction of the stripping and storage cost are in the blast temperature. The lowest steelmaking costs are achieved with the lower blast temperature as the expensive recycling is minimized and when less gas is combusted in the hot stoves more income is received from the exported power and heat.

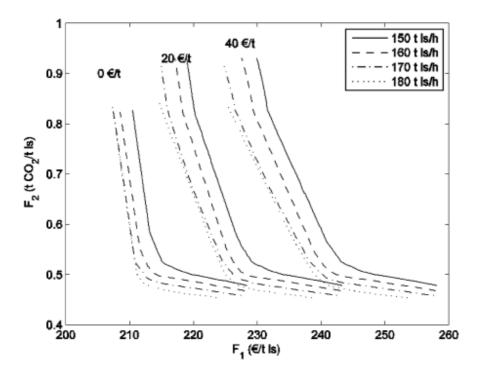


Fig. 20 Pareto frontiers for minimization of the specific costs  $(F_1)$  and the specific emission rate  $(F_2)$  at steel production rates of 150, 160, 170 and 180 t ls/h and  $c_{\text{str}} = 0$ , 20 and 40  $\notin/t$ .

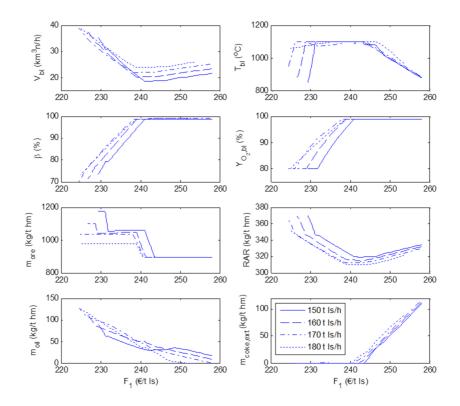


Fig. 21 Central variables for the states of the rightmost of the Pareto frontiers of Fig. 20 (c<sub>str</sub>=40 €/t ls): Blast volume, blast temperature, top gas recycling degree, blast oxygen content, specific ore rate, specific total rate of reductants, oil rate and external coke rate.

## 4.4 Nonlinear optimization of top gas recycling conditions

The linearization of the blast furnace model used in Papers II and III limits the blast oxygen to a content exceeding 80 volume-%, which makes a direct comparison between the traditional blast furnace operation and the operation under top gas recycling impossible. Furthermore, in Papers II and III also both the recycled top gas and the oxygen enriched blast were assumed to be heated, which would require a double set of hot stoves and increase the complexity and investments of the plant. Therefore, Paper IV studies a case where the blast furnace model was not linearized and the system shown in Fig. 9 is optimized with nonlinear programming which makes a direct comparison between the traditional operation and the operation with top gas recycling possible. Special

attention was paid to the optimal heating of the injected gases; either hot top gas with cold oxygen or hot oxygen enriched blast with cold top gas is injected at the tuyeres, which enables the whole integrated plant to be operated with just one set of hot stoves.

The nonlinear optimization problem is solved numerically with respect to blast furnace input variables by sequential quadratic programming subject to constraints of the blast furnace model (cf. Table 7). To reach the solution, multiple starting points are needed and scaling of the constraints is necessary due to high nonlinearity of the problem for the cases with considerable top gas recycling.

Applying the same constraints and cost factors as in Paper II, the results from the linearized model can be compared with those from the nonlinear model. Figure 22 shows that all the depicted variables behave in a similar way in both models. However, the transition between the different states of operation takes place smoother in the nonlinear model. The main difference between the results is found in the blast oxygen content, where the nonlinear model suggests to operate the plant with a blast oxygen content close to its upper limit throughout the studied production range. It should be noted that the solutions produced by the approach, where the blast furnace model is linear will always be located at the vertices, while the nonlinear model can produce optimal points where even no constraints are active.

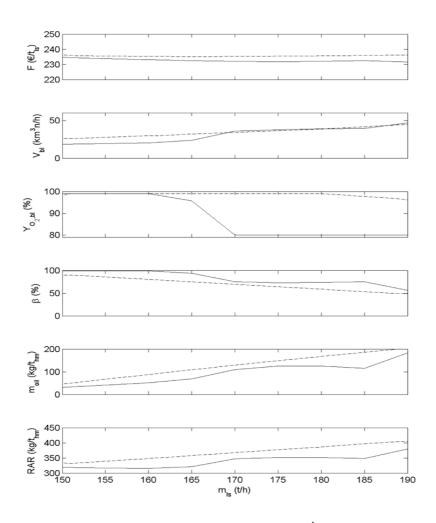


Fig. 22 Minimum cost of liquid steel (*F*), blast volume  $(\dot{V}_{bl})$  and oxygen content( $Y_{O_2, bl}$ ), degree of BF top gas recycled ( $\beta$ ), specific oil injection rate ( $m_{oil}$ ) and specific rate of reductants (RAR) vs. steel production rates for  $c_{CO2} = c_{CCS}$ = 20 €/t. Solid lines represent the linearized model of Paper II and dashed lines the nonlinear model.

In addition to the nonlinearity, the other major difference in the system studied in Paper IV compared to that of Papers II-III lies in the gas preheating system. In the earlier studies both the oxygen enriched blast and the recycled top gas were simulated to be heated to the blast temperature, which in reality would require two sets of hot stoves. In Paper IV two different gas preheating methods were evaluated:

• Method 1: Hot blast and cold recycled top gas

• Method 2: Hot recycled top gas and cold oxygen

The minimum cost of liquid steel for both methods as a function of the production rate and  $CO_2$  emission cost under constant  $CO_2$  stripping and storage cost of 40  $\epsilon/t$  is illustrated in Fig. 23. The results show that at a low  $CO_2$  emission cost, Method 1 is better, while at a higher emission cost Method 2 is more economical. The threshold value of the emission cost is found to be around 25-28  $\epsilon/t$ , depending on the production rate.

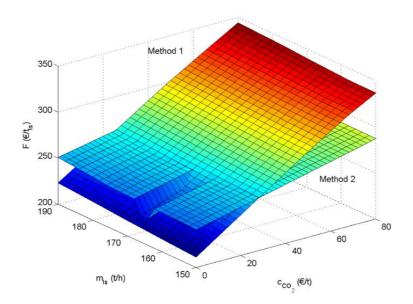


Fig. 23 Minimum cost of liquid steel at  $c_{CCS} = 40 \text{ €/t}$  as function of the production rate and CO<sub>2</sub> cost for hot blast and cold recycled gas (Method 1) and hot recycled gas and cold oxygen (Method 2).

Figure 24 and Fig. 25 show the corresponding top gas recycling degrees for both gas heating methods in the optimal states. It can be noticed that with Method 1 the top gas recycling only becomes economic when the emission cost exceeds 50  $\in$ /t. With Method 2 the recycling degree is close to maximal throughout the studied region, except for a valley at a production rate of 165 t ls/h. The valley, which can also be noticed in Fig. 23, is caused by the possibility to inject considerably more oil at this production rate. Also the other minor variations in the optimal recycling degree for Method 2 in Fig. 25 can be explained by changes in the oil injection rate and it would be economical to increase the oil injection rate and lower the recycling degree but the constraints do not allow for this.

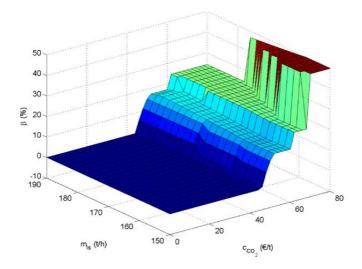


Fig. 24 Top gas recycling degree at  $c_{\text{CCS}} = 40 \text{ €/t}$  as function of the production rate and CO<sub>2</sub> emission cost for Method 1.

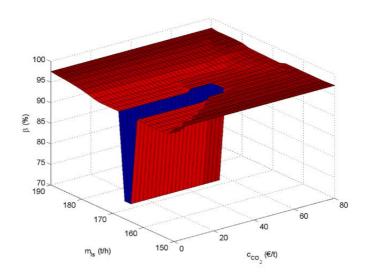


Fig. 25 Top gas recycling degree at  $c_{\text{CCS}} = 40 \text{ } \text{€/t}$  as function of the production rate and CO<sub>2</sub> emission cost for Method 2.

The behavior of some of the key variables as a function of the production rate at constant  $c_{CO_2}=20 \text{ €/t}$  and  $c_{ccs}=40 \text{ €/t}$  is illustrated in Fig. 26. At these conditions Method 1 is more economical at all production rates except at 165 t ls/h, where

the mentioned valley occurs, lowering the steelmaking cost for Method 2 to almost that of Method 1. For Method 2, at this point, the oil rate jumps to its maximum causing a large decrease in the coke rate and smaller drop in the top gas recycling degree. Comparing the methods throughout the investigated interval, Fig. 26 clearly shows how the top gas recycling lowers the coke rate and generates about 1 ton less  $CO_2$  emissions per ton of liquid steel due to storing of the stripped carbon dioxide.

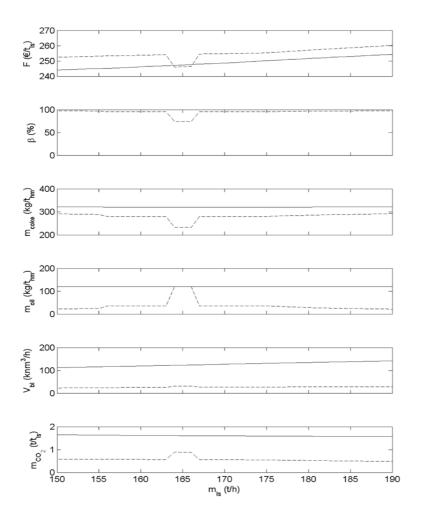


Fig. 26 Minimum cost of liquid steel, top gas recycling degree, coke rate, oil rate, "blast" rate and specific CO<sub>2</sub> emissions at  $c_{CO2} = 20 \text{ €/t}$  and  $c_{CCS} = 40 \text{ €/t}$ versus production rate for Method 1 (solid lines) and Method 2 (dashed lines).

## 4.5 Effect of using Fastmet DRI in blast furnace

In Paper V, the effect and economy of using direct reduced iron (DRI) from the Fastmet process in the blast furnace is studied. By partial substitution of pellets or sinter in the blast furnace burden, the amount of energy used for the iron ore reduction in the blast furnace can be decreased, which in turn leads to lower coke consumption and higher productivity. In this study the system, including the rotary hearth furnace (RHF) for the Fastmet process, is optimized by nonlinear programming with respect to the inputs of the blast furnace model. Prices of the raw materials for the Fastmet process together with the  $CO_2$  emission allowances and DRI rates are varied to analyze the effect on the blast furnace operation parameters and on the economics of the studied system.

The system studied in Paper V is optimized for a production range of 130 to 170 tons of rolled steel per hour. Three cases were studied:

- Case 1: Normal blast furnace operation, 100 % pellet burden, 100 kg/thm briquettes in BF charge produced from fines
- Case 2: Coal used as reductant in RHF, fines used in RHF, DRI used in BF together with pellets
- Case 3: Brazilian or European charcoal used as reductant in RHF, fines used in RHF, DRI used in BF together with pellets

The reductant used in the rotary hearth furnace is either coal or charcoal. Using charcoal in the RHF has two benefits: it is a renewable carbon source and it increases the DRI production in the rotary hearth furnace. On the other hand charcoal is more expensive than coal. In this study the coal used in the RHF costs 90  $\notin$ /t while two different prices were used for the charcoal: 200  $\notin$ /t for Brazilian charcoal and 343  $\notin$ /t for European charcoal. In addition, it is good to notice that the price for coking coal used in this study is 145  $\notin$ /t, meaning that lower quality coal can be used in the RHF. For Cases 2 and 3 a minimum DRI rate of 60 kg/thm was applied to ensure the treatment of the fines of the steel plant.

Figure 27 shows the minimum cost of rolled steel as a function of steel production rate for all the studied cases. The effect of the price of the DRI feed material on the optimal rolled steel production cost is shown in the panel columns while the panel rows depict the effect of the emission cost. First, the figure clearly shows that using DRI in the blast furnace charge can be economical (any of the thin lines is below the thick line) only when the price of DRI feed material is less than 70 % of the price of blast furnace pellets. The use of Fastmet DRI in the blast furnace increases the total emissions of the integrated system. This can be seen as the increasing costs for Case 2 (thin solid line) for any of the columns compared to costs for Case 1. On the other hand, using charcoal as a reductant in the Fastmet process becomes more attractive when the emission cost increases as the fossil  $CO_2$  emissions from the system decrease. In

general using DRI becomes more economical as the steel production rate increases.

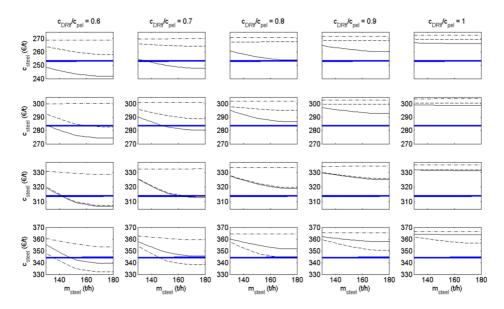


Fig. 27 Minimum cost of rolled steel. Thick line depicts Case 1, thin lines (solid: Case 2, dashed: Case 3, charcoal 200 €/t in RHF, dash-dotted: Case 3, charcoal 343 €/t in RHF) represent cost with different prices for the DRI feed. Emission costs for the panel rows in descending order: c<sub>CO2</sub> = 0 €/t, 20 €/t, 40 €/t and 60 €/t.

The fact that the total carbon consumption of the studied system increases when DRI is produced can also be seen in Fig. 28, where the optimum DRI rate as a function of the relative cost of the DRI feed is presented for different steel production rates and emission costs. For Case 2, where coal is used as a reductant in the RHF, an increase in the emission cost induces the optimal DRI rate to drop. The opposite happens when charcoal is used in the Fastmet process: the optimal DRI rate increases as the emission costs increase. It is also good to notice that in the case of the more expensive European charcoal the maximum DRI rate is only reached at extreme conditions when the steel production rate and the emission cost are high and the DRI feed is cheap.

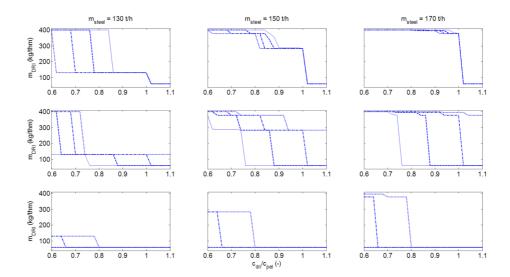


Fig. 28 Optimum DRI rate versus costs of DRI feed for different steel production rates and emission cost. Upper panels: Case 2, Middle panels: Case 3, charcoal  $200 \notin/t$ , Lower panels: Case 3, charcoal  $343 \notin/t$ . Solid lines  $c_{CO2} = 0 \notin/t$ , dashed lines  $c_{CO2} = 20 \notin/t$ , dash-dotted lines  $c_{CO2} = 40 \notin/t$  and dotted lines  $c_{CO2} = 60 \notin/t$ .

## 4.6 Comparison of the models

The studied system of an integrated steel works with rather small modifications between the presented papers is very complex and the optimal conditions of the system vary considerably between the cases studied. Even within a single case the optimal state of the system can exhibit complex transitions as the production rate increases revealing new optimal solutions, which have not been feasible at the lower production rates.

In the following figures the optimal conditions of the systems in Papers I, II, IV and V are presented for a production of 1000 kg hot metal at the production rate of 180 t steel/h. With a raw biomass price of  $150 \notin$ t combined with an emission cost of 20  $\notin$ t the system of Paper I utilizes 44 kg/thm charcoal which leads to a coke consumption of 303 kg/thm.

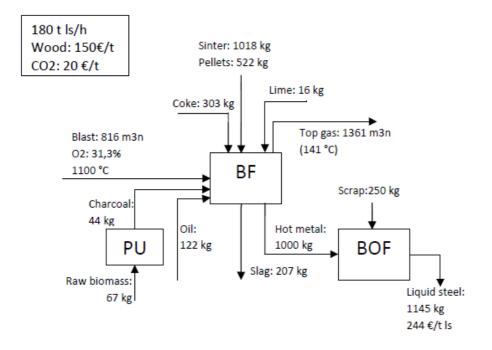


Fig. 29 Optimal operation of the system in Paper I at 180 t ls/h.

The system presented in Papers II and III included the possibility to operate the blast furnace with top gas recycling. The blast furnace model was linearized to simplify the optimization problem. With the cost of CO<sub>2</sub> stripping and storage at 40  $\notin$ /t and the emissions price of 20  $\notin$ /t the system operates with a low coke consumption (226 kg/thm) combined with a high oil injection (126 kg/thm). Due to the relatively high price of the CO<sub>2</sub> stripping the optimal top gas recycling ratio stays relatively low at 73 % and the blast oxygen content is at the lower limit. It is good to remember that in the systems of Papers II and III the oxygen enriched blast was "mixed" with the recycled and stripped top gas before heating the gas mixture to the blast temperature.

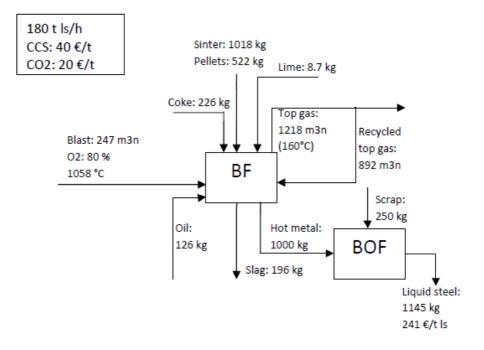


Fig. 30 Optimal operation of the system in Papers II and III at 180 t ls/h.

In Paper IV, the system is nonlinearly optimized without the linearization of the blast furnace model. The blast heating system in the modeling is also made more realistic and either the blast or the recycled top gas is separately heated. With prices of  $c_{CO2} = 20 \text{ €/t}$  and  $c_{CCS} = 40 \text{ €/t}$  the preheating Method 1 (cf. section 4.4) makes a traditional blast furnace operation without the top gas recycling (Fig. 31) the best choice. Method 2 (Fig. 32) in turn chooses to recycle 62 % of the top gas, which decreases the reductant (coke+oil) consumption by around 130 kg/thm. However, at these levels of emissions and stripping costs Method 1 is more economical.

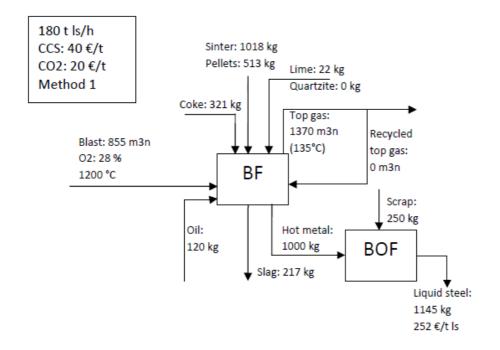


Fig. 31 Optimal operation of the system in Papers IV at 180 t ls/h for Method 1.

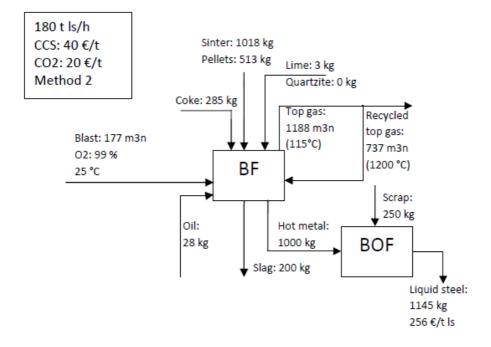


Fig. 32 Optimal operation of the system in Papers IV at 180 t ls/h for Method 2.

In Paper V, a rotary hearth furnace is added to the system to enable a DRI charge to the blast furnace. Charging of directly reduced iron produced by Fastmet 68

technology in the rotary hearth furnace into the blast furnace reduced the coke rate of the blast furnace. However coal or charcoal is needed to reduce the iron bearing material in the rotary hearth furnace.

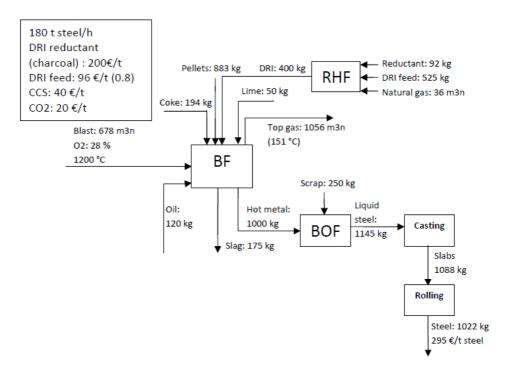


Fig. 33 Optimal operation of the system in Paper V at 180 t steel/h.

The fossil  $CO_2$  emissions per one ton of hot metal from the systems presented in Fig. 29-Fig. 33 are reported in Table 13. It can be seen that the specific emission is highest for operation according to Fig. 31, which was the system studied in Paper IV with Method 1 (hot blast and cold recycled top gas). As the system in this case chooses to operate without the top gas recycling, this case can be seen as the normal blast furnace operation. The fossil emissions from the operation with the biomass injection (Paper I, Fig. 29) are somewhat lower while a more substantial drop is achieved when the rotary heart furnace is added to the studied system (Paper V, Fig. 33). It should be noted that the maximum use of biomass in the study of Paper I was restricted, and this upper bound naturally influences the rate of emissions from the system. The lowest emission rate of 0.59 t  $CO_2$ /thm is achieved for Fig. 32. This is the system studied in Paper IV with Method 2 (cold oxygen blast and hot recycled top gas).

Table 13 Specific  $CO_2$  emissions for systems presented in Fig. 29-Fig. 33.

	Fig. 29	Fig. 30	Fig. 31	Fig. 32	Fig. 33
t CO <sub>2</sub> /thm	1.76	0.95	1.81	0.59	1.26

## 5 CONCLUSIONS AND FUTURE WORK

The modeling of the integrated steel works presented in this thesis, with a blast furnace operated in some modern and innovative ways, gives useful information about the relations of the variables of this complex system. The systems presented in this thesis are not only simulated but also optimized with respect to the production economics, which offers important information about the requirements for cost-efficient production.

Paper I studies the potential of using biomass in the blast furnace. The objective of the study is to illustrate the effect of the raw material prices, emission costs as well as the pyrolysis temperature on the process economy.

Papers II, III and IV investigate the blast furnace top gas recycling, combined with the  $CO_2$  stripping and storage. Paper II investigates the effect of recycling the  $CO_2$ -stripped top gas under a massive oxygen enrichment on the production economy and the emissions of the steel plant. Paper III, in turn, examines the conflicting goals of decreasing both the  $CO_2$  emissions and the liquid steel production costs by multi-objective optimization.

The blast furnace model is linearized in Papers II and III, limiting the study in terms of the blast oxygen content. However, an advantage of the linear formulation is the robustness of the solution and the efficiency of the optimization. In Paper IV, the studied system is optimized by nonlinear programming offering a better comparison to traditional blast furnace operation, but at higher computational cost. Special attention is paid to the optimal heating of the tuyere gases.

Paper V illustrates the potential of using direct reduced iron (DRI) as a partial substitute for pellets in the blast furnace. The DRI is produced with a Fastmet process utilizing a rotary hearth technology. The optimization of the system reveals the influence of the DRI feed, raw material prices and emission costs on the optimal operation conditions.

The optimization presented in Paper I revealed that the injection of the preprocessed biomass can be economically feasible. Even with zero emission costs and at the high steel production rates it is economical to use biomass in the system, as long as the price of the biomass falls below a certain share of the coal price. The largest benefits occur in situations where cheap biomass is available. However, in this study there were no costs from the pyrolysis process, as it was assumed that the by-products from the process could be utilized in the pyrolysis unit. Biomass injection also lowers the specific fossil emissions from the system.

The blast furnace operation with the carbon capture and storage and the recycling of the stripped top gas, investigated in Papers II-IV, showed a great potential in decreasing the  $CO_2$  emissions of an integrated steel plant. However, with the current price structure, where the cost for the CCS is considerably higher than the

emissions price, the blast furnace operation with the CCS and top gas recycling is not feasible from an economical point of view.

The benefits of using the DRI, produced with the Fastmet process, in the blast furnace depend on the price ratio between the DRI pellets and the blast furnace pellets and on the reductant used in the rotary hearth furnace. If the DRI feed material is low-priced, there is an economical advantage in using DRI in the blast furnace. With zero emissions cost the mentioned advantage exists only if coal is used as a reductant in the RHF. At higher emission costs DRI becomes economical also in cases where charcoal is used as a reductant. The use of charcoal decreases the amount of fossil  $CO_2$  emissions from the system compared to operation without DRI and if coal is used in the RHF the  $CO_2$  emission is even higher due to the high energy demand of the Fastmet process.

In the future the unit processes of the studied system, which are now described with simple linear relationships, can be further developed into more detailed models. Considering Paper I, the model of the pyrolysis unit could be improved by including an energy balance for the pyrolysis step. The effect of the pyrolysis time and utilization of the valuable pyrolysis byproducts should also be taken into consideration. In addition, the  $CO_2$  stripping unit used in Papers II-IV could be updated with a more detailed model that would consider the influence of the quantity and the composition of the gas. Considering the use of DRI in the blast furnace, the effect of other processes on the DRI production should be evaluated.

## 6 **REFERENCES**

- Arasto A, Tsupari E., Kärki J., Pisilä E. and Sorsamäki L., "Post-combustion capture of CO<sub>2</sub> at an integrated steel mill – Part I: Technical concept analysis", International Journal of Greenhouse Gas Control, Vol. 16 (2013), pp. 271-277.
- Austin P.R., Nogami H. and Yagi J., "Prediction of blast furnace performance with top gas recycling", ISIJ International, Vol. 38 (1998), pp. 239-245.
- Babich A. I., Gudenau H. W., Mavromatis K. T., Froehling C., Coves A., García L., "Choice of technological regimes of a blast furnace operation with injection of hot reducing gases", Revista de Metallurgia (Madrid), Vol. 38 (2002), pp. 288 - 305.
- Babich A., Senk D. and Fernandez M., "Charcoal Behaviour by Its Injection into the Modern Blast Furnace", ISIJ International, Vol. 50 (2010), pp. 81-88.
- BIR (Bureau of International recycling), "World steel recycling in figures 2008-2012", available at: http://www.bir.org/assets/Documents/publications/brochures/7587Ferrous Report2013.pdf [Cited 05/09/13], accessed October 2013.
- Biswas A. K., "Principles of Blast Furnace Ironmaking", Cootha Publishing House, Brisbane, (1987).
- Burdyny T. and Struchtrup H., "Hybrid membrane cryogenic separation of oxygen from air for use in the oxy-fuel process", Energy, Vol. 35 (2010), pp. 1884-1897.
- CC&ST (Carbon capture and sequestration technologies), Non-power plant carbon dioxide capture and storage projects, available at: http://sequestration.mit.edu/tools/projects/storage\_only.html [Cited 12/11/12], accessed October 2013.
- Chatterjee A., "Hot metal production by smelting reduction of iron oxide", PHI Learning Private Limited, New Delhi, (2010).
- Chu M. and Yagi, J.-I., "Numerical evaluation of blast furnace performance under top gas recycling and lower temperature operation", Steel Research International, Vol. 81 (2010), pp. 1043-1050.
- Cordero T., Marquez F., Rodriquez-Mirasol J. and Rodriqiez J. J., "Predicting heating values of lignocellulosics and carbonaceous material from proximate analysis", Fuel, Vol. 80 (2001), pp. 1567-1571.
- Danloy G., Berthelemot A., Grant M., Borlee J., Sert D., van der Stel J., Jak H., Dimastromatteo V., Hallin M., Eklund N., Sundqvist L., Sköld B.-E., Lin R., Feiterna A., Korthas B., Müller F., Feilmayr C. and Habermann A., "ULCOS Pilot testing of the low-CO<sub>2</sub> blast furnace process at the experimental BF in Luleå", Revue de Métallurgie, Vol. 106 (2009), pp. 1-8.

- Desai B., Ramna R.V. and Sathaye J.M., "Effect of hot reducing gas (HRG) injection on blast furnace operational parameters: theoretical investigation", Ironmaking and Steelmaking, Vol. 35 (2008), pp. 43-50.
- Floudas, C. A., "Nonlinear and Mixed-Integer Optimization: Fundamentals and Applications", Oxford University Press, Oxford (1995).
- Ghosh A. and Chatterjee A., "Ironmaking and Steelmaking: Theory and Practise", Prentice-Hall of India Private Limited, New Delhi, (2008).
- Gibbins J. and Chalmers H., "Carbon capture and storage", Energy Policy, Vol. 36 (2008), pp. 4317-4322.
- Gielen D., "CO<sub>2</sub> Removal in the iron and steel industry", Energy Conversion and Management, Vol. 44 (2003), pp. 1027-1037.
- Haugen H. A., Eldrup N., Bernstone C., Liljemark S., Pettersson H., Noer M., Holland J., Nilsson P.A., Hegerland G. and Pande J.O., "Options for transporting CO<sub>2</sub> from coal fired power plants Case Denmark", Energy Procedia, Vol. 1 (2009), pp. 1665-1672.
- Helle H., Helle M. and Saxén H., "Evaluation of partial replacement of oil by biomass as injectant in the blast furnace", 3rd Nordic Symposium for Young Scientists in Metallurgy, ed. by L. Holappa and M. Kekkonen, Helsinki University of Technology, Espoo, Finland, TKK, (2008), pp. 4-7.
- Huitu K., Kekkonen M. and Holappa L., "Novel steelmaking processes –Literature study and critical review", Helsinki University of Technology Publications in Materials Science and Engineering, TKK-MT-207, Espoo, Finland, (2009)
- Intergovernmental Panel on Climate Change (2005) Special report on carbon dioxide capture and storage. Cambridge University Press, New York.
- Johnson E., "Goodbye to carbon neutral: Getting biomass footprints right", Environmental Impact Assessment Review, Vol. 29 (2009), pp. 165-168.
- Khoo H. H. and Tan R. B. H., "Life cycle investigation of CO<sub>2</sub> recovery and sequestration", Environmental Science & Technology, Vol. 40 (2006), pp. 4016-4024.
- Kim Y. and Worrel E., "International comparison of CO<sub>2</sub> emission trends in the iron and steel industry", Energy Policy, Vol. 30 (2002), pp. 827-838.
- Knudsen J. N., Jensen J. N., Vilhelmsen P.-J. and Biede O., "Experience with CO<sub>2</sub> capture from coal flue gas in pilot-scale: Testing of different amine solvents", Energy Procedia, Vol. 1 (2009), pp. 783-790.
- Kujanpää L., Rauramo J. and Arasto A., "Cross-border CO<sub>2</sub> infrastructure options for a CCS demonstration in Finland", Energy Procedia, Vol. 4 (2011), pp. 2425-243.

- Lüngen H. B. and Schmöle P., "Blast furnace operation without coke and carbon?", Stahl und Eisen, Vol. 124 (2004), pp. 63-72.
- Madlener R. and Schmid C., "Combined heat and power generation in liberalised markets and a carbon-constrained world", GAIA Ecological Perspectives for Science and Society, Vol. 12 (2003), pp. 114–120.
- Mathieson J. G., Rogers H., Somerville M. A., Jahanshahi S. and Ridgeway P., "Potential for the use of biomass in the iron and steel industry" Chemeca 2011, Paper 252 (CD), Sydney, Australia.
- Mathieson J. G., Norgate T., Jahanshahi S., Somerville M. A., Haque N., Deev A., Ridgeway P. and Zulli P., "The potential for charcoal to reduce net greenhouse gas emissions from the Australian steel industry", 6<sup>th</sup> International Congress on the Science and Technology of Ironmaking (2012), Rio de Janeiro, Brazil, pp. 1602-1613.
- Meijer K., Denys M., Lasar J., Birat J.-P., Still G. and Overmaat, B., "ULCOS: ultralow CO<sub>2</sub> steelmaking", Ironmaking & Steelmaking, Vol. 36 (2009), pp. 249-251.
- Midrex, World direct reduction statistics 2012, available at: http://www.midrex.com/uploads/documents/MidrexStats2011-6.7.12.pdf [Cited 05/09/13], accessed October 2013.
- Murai R., Sato M. and Ariyama T., "Design of innovative blast furnace for minimizing CO<sub>2</sub> emission based on optimization of solid fuel injection and top gas recycling", ISIJ International, Vol. 44 (2004), pp. 2168-2177.
- Nill J., "Technological Competition, Time, and Windows of Opportunity the Case of Iron and Steel Production Technologies" IÖW Discussion Paper 58/03, Institute for Ecological Economy Research, Berlin, Germany (2003)
- Nogami H., Chu M. and Yagi J., "Numerical analysis of blast furnace operations with top gas recycling", Revue de Métallurgie, Vol. 102 (2005), pp. 189-197.
- Nogami H., Yagi J., Kitamura S. and Austin P. R., "Analysis of material and energy balances of ironmaking systems on blast furnace operations with metallic charging, top gas recycling and natural gas injection", ISIJ International, Vol. 46 (2006), pp. 1759-1766.
- OECD/IEA, "Energy Technology Perspectives 2010, Scenarios and Strategies to 2050", available at: http://www.iea.org/publications/freepublications/publication/etp2010.pdf [Cited 07/05/12], accessed October 2013.
- Ogden J. and Johnson N., "Techno-economic analysis and modelling of carbon dioxide (CO<sub>2</sub>) capture and storage (CCS) technologies", Developments and innovations in CCS technology, Vol. 1, Maroto-Valer M. M. (ed.), Woodhead Publishing Limited, Cambridge (2010)
- Olajire A. A., "CO<sub>2</sub> capture and separation technologies for end-of-pipe applications A review", Energy, Vol. 35 (2010), pp. 2610-2628.

- Parfomak P. W. and Folger P., "Carbon Dioxide (CO<sub>2</sub>) Pipelines for Carbon Sequestration: Emerging Policy Issues", (2007). Available at: http://www.policyarchive.org/handle/10207/bitstreams/3227\_Previous\_Ve rsion\_2007-04-19.pdf [Cited 12/11/12], accessed October 2013.
- Pires J. C. M., Martins F. G., Alvim-Ferraz M. C. M. and Simões M., "Recent developments on carbon capture and storage: An overview", Chemical Engineering Research and Design, Vol. 89 (2011), pp. 1446-1460.
- Rist A. and Meysson N., "A dual graphic representation of blast-furnace mass and heat balances", Journal of Metals, Vol. 19 (1967), pp. 50-59.
- Ranta J., "Production of charcoal", (in Finnish), Master Thesis, Helsinki University of Technology, Helsinki, Finland, (1994).
- Saxén H., Brämming M., Wikström J.-O. and Wiklund P., "Theoretical limits on operation under high oxygen enrichment in the blast furnace", 60th Ironmaking Conference, Vol. 60 (2001), pp. 721-733.
- Suopajärvi H. and Fabritius T., "Effects of Biomass Use in Integrated Steel Plant Gate-to-gate Life Cycle Inventory Method", ISIJ International, Vol. 52 (2012), pp. 779–787.
- Suopajärvi H. and Fabritius T., "Evaluation of the possibility to utilize biomass in Finnish blast furnace ironmaking", Proc. 4<sup>th</sup> International Conference on Process Development in Iron and Steelmaking, Vol. 1 (2012), Luleå, Sweden, pp. 629-638.
- Thorn T., "Simulering av masugnsdrift vid höga olje- och syremängder (Simulation of blast furnace operation under high oil and oxygen enrichment)", Master Thesis, Åbo Akademi University, Åbo, Finland, (2002).
- Tobiesen F. A., Svendsen H. F. and Mejdell T., "Modeling of blast furnace CO<sub>2</sub> capture using amine absorbents", Industrial & Engineering Chemistry Research, Vol. 46 (2007), pp. 7811-7819.
- Tseitlin M. A., Lazutkin S.E. and Styopin G.M., "A flow-chart for iron making on the basis of 100% usage of process oxygen and hot reducing gases injection", ISIJ International, Vol. 34 (1994), pp. 570-573.
- Teir S., Pikkarainen T., Kujanpää L., Tsupari E., Kärki J., Arasto A. And Aatos S., "Carbon capture and storage (CCS) Technology review", (in Finnish), VTT Working Papers 161, VTT, Finland (2011).
- Tsupari E., Kärki J., Arasto A and Pisilä E., "Post-combustion capture of CO2 at an integrated steel mill Part II: Economic feasibility", International Journal of Greenhouse Gas Control, Vol. 16 (2013), pp. 278-286.

- Ueda S. and Ariyama T., "Evaluation of Biomass Injection into Blast Furnace for Reducing CO<sub>2</sub> Emission", Proc. 3<sup>rd</sup> International Conference on Process Development in Iron and Steelmaking, Vol. 2 (2008), Luleå, Sweden, pp. 217-226.
- Ueda S., Watanabe K., Yanagiya K., Inoue R. and Ariyama T., "Improvement of Reactivity of Carbon Iron Ore Composite with Biomass Char for Blast Furnace", ISIJ International, Vol. 49 (2009), pp. 1505-1512.
- ULCOS, Ultra-Low CO<sub>2</sub> Steelmaking, available at: http://www.ulcos.org/en/index.php [Cited 05/09/13], accessed October 2013.
- World Steel Association, Data from World Steel Association, available at: http://www.worldsteel.org/ [Cited 03/02/14], accessed February 2014.
- Zuo G. Q. and Hirsch A., "The trial of the top gas recycling blast furnace at LKAB's EBF and scale-up", Revue de Métallurgie, Vol. 106 (2009), pp. 387-392.

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