

Hannu Suopajärvi

BIOREDUCER USE IN BLAST FURNACE IRONMAKING IN FINLAND

TECHNO-ECONOMIC ASSESSMENT AND CO₂
EMISSION REDUCTION POTENTIAL

UNIVERSITY OF OULU GRADUATE SCHOOL;
UNIVERSITY OF OULU,
FACULTY OF TECHNOLOGY



ACTA UNIVERSITATIS OULUENSIS
C Technica 513

HANNU SUOPAJÄRVI

**BIOREDUCER USE IN BLAST
FURNACE IRONMAKING IN
FINLAND**

Techno-economic assessment and CO₂ emission
reduction potential

Academic dissertation to be presented with the assent of
the Doctoral Training Committee of Technology and
Natural Sciences of the University of Oulu for public
defence in the OP auditorium (L10), Linnanmaa, on 23
January 2015, at 12 noon

UNIVERSITY OF OULU, OULU 2014

Copyright © 2014
Acta Univ. Oul. C 513, 2014

Supervised by
Professor Timo Fabritius
Docent Eva Pongarácz

Reviewed by
Professor Risto Soukka
Doctor Chuan Wang

Opponent
Professor Margareta Wihersaari

ISBN 978-952-62-0705-6 (Paperback)
ISBN 978-952-62-0706-3 (PDF)

ISSN 0355-3213 (Printed)
ISSN 1796-2226 (Online)

Cover Design
Raimo Ahonen

JUVENES PRINT
TAMPERE 2014

Suopajärvi, Hannu, Bioreducer use in blast furnace ironmaking in Finland. Techno-economic assessment and CO₂ emission reduction potential

University of Oulu Graduate School; University of Oulu, Faculty of Technology

Acta Univ. Oul. C 513, 2014

University of Oulu, P.O. Box 8000, FI-90014 University of Oulu, Finland

Abstract

Most of the steel produced in the world is based on the integrated blast furnace-converter route, which is based on the use of virgin raw materials. Large amounts of fossil-based, carbon containing reductants are used in blast furnaces, which results in carbon dioxide emissions into the atmosphere. Fossil carbon dioxide emissions from steel production can be reduced by new technologies or moving from non-renewable to renewable energy sources. Biomass-based reductants could be one way to reduce the specific emissions from blast furnace-based steel production.

The aim of this thesis was to examine the techno-economic and CO₂ mitigation potentials of using bioreducers in blast furnace ironmaking. Bioreducer feasibility was analyzed in the Finnish context, but the research methods used can be applied more widely. The metallurgical properties of bioreducers were evaluated and compared to fossil-based reductants. The impact of bioreducers on blast furnace behavior and on other steel plant processes was evaluated, with an emphasis on the reductions achieved in CO₂ emissions at the plant scale. The CO₂ emissions, energy consumption and production costs of bioreducers were evaluated, as was the availability of energy wood for bioreducer production.

The results show that solid, liquid and gaseous bioreducers can be produced with thermochemical conversion technologies. However, their suitability for blast furnace use varies greatly. The highest substitution of fossil-based reductants in a blast furnace is achieved with charcoal injection. The carbon footprint of torrefied wood, charcoal and Bio-SNG is moderate compared to fossil-based reducing agents and their production is energetically feasible. The economic feasibility of bioreducers is currently weak in comparison to fossil-based reducing agents, but competitive when compared to other CO₂ emission reduction measures such as carbon capture and storage. The biomass availability assessment revealed that sufficient amount of energy wood could be available for bioreducer production in the areas where Finnish steel plants are situated. The feasibility of bioreducer production could be improved by producing a number of products from the biomass and taking advantage of the process of integration possibilities.

Keywords: biomass, bioreducer, blast furnace, carbon footprint, CO₂ mitigation, CO₂ mitigation cost, energy return on investment, industrial ecology, ironmaking, modeling, production cost, reducing agent

Suopajärvi, Hannu, Biopelkistimien käyttö masuunipohjaisessa raudanvalmistuksessa Suomessa. Teknistaloudellinen arviointi ja CO₂ päästöjen vähennyspotentiaali

Oulun yliopiston tutkijakoulu; Oulun yliopisto, Teknillinen tiedekunta

Acta Univ. Oul. C 513, 2014

Oulun yliopisto, PL 8000, 90014 Oulun yliopisto

Tiivistelmä

Suurin osa maailmassa tuotetusta teräksestä valmistetaan integroidulla masuuni-konvertteri reitillä, joka perustuu neitseellisten raaka-aineiden käyttöön. Masuuniprosessissa käytetään suuri määrä fossiilisia, lähinnä hiilipohjaisia pelkistimiä, jotka aiheuttavat hiilidioksidipäästöjä ilmakehään. Fossiilisia hiilidioksidipäästöjä voidaan teräksenvalmistuksessa vähentää uusilla teknologioilla tai siirtymällä uusiutumattomista energialähteistä uusiutuviin. Biomassasta valmistetut pelkistimet voisivat olla yksi mahdollinen keino alentaa masuunipohjaisen teräksenvalmistuksen ominaispäästöjä.

Tämän työn tavoitteena oli tarkastella biopelkistimien käytön teknistaloudellista potentiaalia masuunikäytössä ja aikaansaattavia hiilidioksidipäästövähennyksiä eri systeemirajauksilla. Työssä keskityttiin tarkastelemaan biopelkistimien hyödynnettävyyttä lähinnä Suomen tasolla, vaikka käytetyt tutkimusmenetelmät ovat sovellettavissa myös laajemmin. Työssä arvioitiin biopelkistimien metallurgisia ominaisuuksia, niiden vaikutusta masuuniprosessiin ja laajemmin muihin terästehtaan prosesseihin, pääpainon ollessa saavutettavan CO₂ päästövähennyksen tarkastelussa. Työssä tarkasteltiin biopelkistimien valmistuksen CO₂ päästöjä, energiankulutusta ja tuotantokustannuksia sekä energiapuun saatavuutta biopelkistimien tuotantoon.

Tulokset osoittavat, että biomassasta voidaan valmistaa kiinteitä, nestemäisiä ja kaasumaisia pelkistimiä termokemiallisilla konversioteknologioilla, joiden soveltuvuus masuunikäyttöön vaihtelee suuresti. Masuuniprosessissa suurin fossiilisten pelkistimien korvaavuus saavutetaan käyttämällä puuhiili-injektiota. Torrefioitun puun, puuhiilen ja Bio-SNG:n hiilijalanjälki on varsin maltillinen verrattuna fossiilisiin pelkistimiin ja niiden tuotanto on energietehtäimesti järkevää. Biopelkistimien taloudellinen kannattavuus verrattuna fossiilisiin pelkistimiin on tällä hetkellä heikko, mutta kilpailukykyinen verrattuna muihin CO₂ päästöjen vähennyskeinoihin, kuten hiilidioksidin talteenottoon ja -varastointiin. Energiapuun saatavuus biopelkistimien valmistukseen on suurin alueilla, jotka sijaitsevat lähellä Suomen terästehtaita. Biopelkistimien tuotannon kannattavuutta voitaisiin parantaa tuottamalla useita tuotteita ja hyödyntämällä prosessi-integraatioita.

Asiasanat: biomass, biopelkistin, CO₂ vähennys, CO₂ vähennyskustannus, hiilijalanjälki, mallinnus, masuuni, pelkistin, raudanvalmistus, sijoitetun fossiilienergian tuottosuhde, teollinen ekologia, tuotantokustannus

Acknowledgements

This thesis contains work mainly financed by the Graduate School in Chemical Engineering (GSCE). Funding has also been received from the Finnish Funding Agency for Technology and Innovation (Tekes) under the Showcase 4 project of the System Integrated Metal Production program of the Finnish Metals and Engineering Competence Cluster (FIMECC Ltd). This thesis has also been supported by scholarships awarded by the Emil Aaltonen Foundation and Finnish Foundation for Technology (TES). The topic of this thesis is strongly linked to the Bioreducer project (decision number 70022/10) funded by the Finnish Funding Agency for Technology and Innovation (Tekes).

The opportunity to pursue a doctoral degree came to me quite accidentally. I would like to thank Professor Emeritus Jouko Härkki and Mr. Juho Välikangas, who informed me about the graduate school openings in 2009 which led to this thesis project. I also would like give credit to Mr. Mikko Angerman, who helped me a lot at the beginning of this thesis project. My sincere gratitude goes to my supervisors Professor Timo Fabritius and Docent Eva Pongrácz, who let me design the structure of the work quite freely, but gave guidance and support when needed. I also would like to thank my pre-examiners Professor Risto Soukka from Lappeenranta University of Technology and Doctor Chuan Wang from Swerea MEFOS for their valuable comments. Furthermore, I would like to thank my colleagues at the Process Metallurgy Group. Special thanks go to Mr. Antti Kemppainen and Mr. Petri Sulasalmi for our fruitful discussions, which fortunately concentrated on topics other than work.

Finally, I would like to thank my family and friends. My parents have always encouraged me to study further, which has been very important for me. I would like to express my gratitude to my good friend Anna-Leena, with whom I shared my life for a long time. My biggest role model guiding my research is my son Julius. Once, we were wondering about issues related to space or something, and he just said: “We cannot understand everything—we are not researchers.” Researchers or not, we cannot understand everything, but it is the curiosity of a child that every researcher should bear in mind. Julius, you are the fuel and light for my everyday life. I love you now and forever.

Oulu, October 2014

Hannu Suopajarvi

Abbreviations

BF	Blast furnace
BFG	Blast furnace gas
Bio-SNG	Biomass-based synthetic natural gas
BOF	Basic oxygen furnace
CCS	Carbon capture and storage
CFP	Carbon footprint
COG	Coke oven gas
DRI	Direct reduced iron
DTF	Drop tube furnace
EAF	Electric arc furnace
EROI	Energy return on investment
EU ETS	European Union Emissions Trading System
HHV	Higher heating value
HM	Hot metal
ILUC	Indirect land use change
LR	Logging residue
NG	Natural gas
PC	Pulverized coal
SDW	Small-diameter wood
SEC	Specific energy consumption
ST	Stump
TGR-BF	Top gas recycling blast furnace
ULCOS	Ultra-Low Carbon Dioxide Steelmaking

List of original publications

This thesis is based on the following publications, which are referred to throughout the text by their Roman numerals

- I Suopajarvi H, Fabritius T (2012) Effects of biomass use in integrated steel plant – gate-to-gate life cycle inventory method. *ISIJ Int* 52(5):779–787.
- II Suopajarvi H, Pongrácz E, Fabritius T (2013) The potential of using biomass-based reducing agents in the blast furnace: A review of thermochemical conversion technologies and assessments related to sustainability. *Renew Sustain Energy Rev* 25:511–528.
- III Suopajarvi H, Fabritius T (2013) Towards More Sustainable Ironmaking – An Analysis of Energy Wood Availability in Finland and the Economics of Charcoal Production. *Sustainability* 5(3): 1188–1207.
- IV Suopajarvi H, Pongrácz E, Fabritius T (2014) Bioreducer use in Finnish blast furnace ironmaking – Analysis of CO₂ emission reduction potential and mitigation cost. *Appl Energy* 124: 82–93.

All the above publications were written by the author of this thesis, whose main responsibilities were the research design, data analysis and reporting of the results.

Contents

Abstract	
Tiivistelmä	
Acknowledgements	7
Abbreviations	9
List of original publications	11
Contents	13
1 Introduction	15
1.1 Aim of this study	16
1.2 Research context and outline	17
2 State of the art in CO₂ mitigation in steelmaking	21
2.1 Steel production routes	21
2.2 Blast furnace process and reducing agents	23
2.3 CO ₂ breakthrough programs	25
2.3.1 Ultra-Low Carbon Dioxide Steelmaking	25
2.3.2 Other CO ₂ breakthrough programs	28
3 Bioreducers	31
3.1 Top-charged bioreducers	32
3.2 Injected bioreducers	34
3.3 Commercial use of bioreducers in BF	36
3.4 Biomass raw materials for reducing agent production	37
3.5 Thermochemical conversion technologies for biomass upgrading	38
3.6 Environmental and economic considerations of bioreducers	40
3.6.1 CO ₂ emissions of bioreducers	40
3.6.2 Energy return on investment of bioreducers	42
3.6.3 Production cost of bioreducers	42
4 Materials and methods	45
4.1 System boundaries	45
4.2 Data sources	48
4.3 Comparison of the metallurgical properties of fossil-based and renewable reductants	49
4.4 Blast furnace and integrated steel plant modeling	52
4.5 Carbon footprint assessment	54
4.6 Energy return on investment	55
4.7 Economic analyses	56
5 Results and discussion	57
	13

5.1	Assessment of the metallurgical properties of bioreducers	57
5.2	The impact of bio reducer use on BF behavior	58
5.3	The impact of bio reducer use on integrated steel plant CO ₂ emissions	59
5.4	Carbon footprint of bioreducers	62
5.5	Primary energy consumption of bio reducer production	65
5.6	Production costs of bioreducers	66
5.7	CO ₂ reduction potential and mitigation costs of bioreducers in Finnish BF ironmaking	69
5.8	Energy wood availability in Finland for bio reducer production	72
5.9	Evaluation of the reliability of the results	74
5.9.1	Blast furnace model	74
5.9.2	Unit processes in gate-to-gate model	75
5.9.3	Carbon footprint model	76
5.9.4	Forest chip availability assessment	77
6	Summary and conclusions	79
6.1	Summary	79
6.2	Concluding remarks	81
6.3	Recommendations for further research	83
	References	85
	Original publications	95

1 Introduction

Steel is one of the most important construction materials in the world today. Crude steel production in 2013 was 1,607 million tonnes (World Steel Association 2014a). The major proportion (about 70%) of the steel is produced from virgin raw materials with the integrated blast furnace–basic oxygen furnace (BF–BOF) route (World Steel Association 2014b). Production of steel from virgin raw materials is energy intensive, requires the use of fossil-based reducing agents (e.g. coke, coal, oil, natural gas) and results in high CO₂ emissions. Production of one tonne of steel with the BF–BOF route emits around 2.0–2.5 tonnes of fossil CO₂ emissions, depending on the technology efficiency, geographical location of the plant and emission allocation methodology (Burchart-Korol 2013, Norgate & Langberg 2009, Norgate *et al.* 2012). The cumulative energy demand of steel production is 21.0–35.4 GJ/t steel (Norgate & Langberg 2009, Burchart-Korol 2013).

To promote the environmental compliance of steel production, the European steel industry has been involved in developing several breakthrough technologies to significantly decrease CO₂ emissions in iron and steelmaking. The Ultra-Low Carbon Dioxide Steelmaking (ULCOS) program was initialized in 2004 to find technologies to cut CO₂ emissions by 50% compared to the present best practice technologies. Over 80 process routes were initially screened, and four process routes were selected for further investigation. Some of them proceeded to the pilot or demonstration phases, but they are still far from commercialization (Birat *et al.* 2012).

CO₂ emission reduction in the iron and steel industry could also be approached from the fuel switch point of view. Fossil-based reducing agents such as coke, coal and natural gas are used in blast furnaces to remove oxygen from iron ore to produce hot metal. Renewable biomass could be used to replace some of the fossil-based reducing agents in the BF and contribute to lower overall fossil CO₂ emissions.

The technological feasibility of using biomass-based reducing agents (bioreducers) in BF has been studied by several authors and with several research methods. According to these studies, there is a true potential to utilize renewable reducing agents in the BF in major quantities (e.g. Babich *et al.* 2010, Mathieson *et al.* 2012). Besides the technological feasibility, adoption of biomass as an alternative raw material in iron and steelmaking should be viewed from different perspectives. The studies conducted earlier have mainly concentrated on the use

of charcoal in BF. Despite the few studies (e.g. Helle *et al.* 2009, Wiklund *et al.* 2012) where bioreducers have been introduced at integrated steelworks, there is a need for more detailed plant site considerations of material and energy balances, CO₂ reduction potential and environmental burden.

There are a couple of studies in which the availability, life cycle impacts and economics of using charcoal as a reducing agent have been studied (e.g. Norgate & Langberg 2009, Norgate *et al.* 2012). The raw material in these studies was short-rotation eucalyptus, which is not available in Northern European climate conditions such as in Finland. There is a need to produce country-specific data about the environmental and economic feasibility of alternative bioreducers. This kind of approach makes it possible to conduct CO₂ mitigation cost analyses. At the same time, the availability of biomass should be considered. Because of the shift toward renewable energy sources, there is growing demand on biomass resources, which might diminish the availability of biomass for steel industry use.

1.1 Aim of this study

The general aim of this thesis was to gain knowledge of the feasibility of using bioreducers in Finnish blast furnace-based steelmaking. The research approach can be defined as a systems approach, which means that no particular phenomenon has been the focus of the research. Several research methods from the industrial ecology toolbox, ranging from process modeling based on mass and energy balances and thermodynamics to carbon footprint calculations and production cost analyses, have been used to assess the feasibility of utilizing bioreducers in blast furnace ironmaking. To achieve the aim of the thesis, the following objectives were set:

- Review the metallurgical properties of different bioreducers.
- Estimate the availability of energy wood (logging residues, small-diameter wood and stumps) in Finland, with an emphasis on competitive use.
- Assess the impact of bioreducer use on blast furnace behavior.
- Evaluate the gate-to-gate (plant-wide) system impacts of biomass introduction on an integrated steel plant.
- Calculate and compare the production costs, carbon footprint and primary energy use of bioreducers produced from energy wood.
- Ascertain the CO₂ reduction potential and mitigation costs when using bioreducers in Finnish blast furnace ironmaking.

1.2 Research context and outline

Any time that new raw material is introduced to the blast furnace, there will be changes in the material and energy balances of the furnace that also reflect to the surrounding processes. Bioreducers have different chemical properties compared to fossil-based reducing agents, which also implies changes in the material and gas balances. Besides metallurgical unit process and steel plant system considerations, the role of thermochemical conversion technologies to produce bioreducers for blast furnace ironmaking are also of importance.

Thermochemical conversion technologies differ greatly from each other in terms of processing temperature, heating rate, energy demand, product distribution, and cost. All these factors influence the product properties and feasibility from the technological and economic points of view. In order to choose the best conversion technology and bioreducer for BF ironmaking, the maturity of the technology, conversion efficiency, etc. must be thoroughly evaluated.

Assessment of life cycle-based CO₂ emissions and energy consumption and the production cost of bioreducer production from wood available in Finland is conducted. Forest-based biomass (termed “energy wood”) comprising logging residues, first thinnings and stumps are considered as suitable feedstock for bioreducer production.

Biomass availability is one of the factors to facilitate the use of renewable bioreducers in blast furnace ironmaking. Biomass resources are not evenly distributed around the world, thus making utilization in metallurgical industry possible for a limited number of nations. Wood can be considered the best alternative for bioreducer production due to its fairly constant chemical composition and well-developed harvesting technologies. In Finland, where the annual growth of wood is rather low, the best solution could be the utilization of energy wood. However, this residue stream will be under serious competition during the next decades. Evaluation of whether there is enough raw material for the steel industry is needed.

The principles of industrial ecology and industrial symbiosis can be seen as an umbrella for this research. Industrial ecology, according to Seager & Theis (2002), is a relatively new science that has no consistent definition or uniform analytical framework. Industrial ecology is a multidisciplinary concept that borrows the idea of closed-loop systems from biological ecosystems (Jelinski *et al.* 1992). In industrial ecology, processes and industries are seen as interacting systems, where systems with linear material and energy flows are replaced with

closed-loop systems (Gibbs & Deutz 2007). According to Chertow (2000), industrial ecology operates at three levels: (1) the facility or firm level; (2) the inter-firm level; and (3) the regional/global level. A similar approach can be identified in this research.

Industrial symbiosis is part of industrial ecology, defined as “engaging traditionally separate industries in a collective approach to competitive advantage involving physical exchange of materials, energy, water, and by-products. The keys to industrial symbiosis are collaboration and the synergistic possibilities offered by geographic proximity” (Chertow 2000). Kalundborg in Denmark has been used as an example of self-organized industrial symbiosis that has developed over time.

Today, industrial symbioses are more carefully planned by utilizing various tools (Brent *et al.* 2012, Sokka *et al.* 2010, Zhou *et al.* 2012). Seager & Theis (2002) state that industrial ecology can be made analytical with two perspectives: life cycle assessment and systems analysis. Life cycle assessment (Sokka *et al.* 2010), substance flow analysis (Zhang *et al.* 2013), process modeling and simulation (Zhang *et al.* 2008) and optimization (Karlsson & Wolf 2008) are tools that have been used to evaluate the economic, environmental and technological feasibility of industrial symbioses.

The research context of the thesis is presented in Fig. 1 with reference to published research Papers I–IV. Only the major process gas streams are shown in the plant site diagram. For example blast furnace gas is also utilized in heating the blast air in hot stoves and to fire up coking batteries with coke oven gas. More detailed description of the contribution of the original publications to thesis objectives is presented in Table 1.

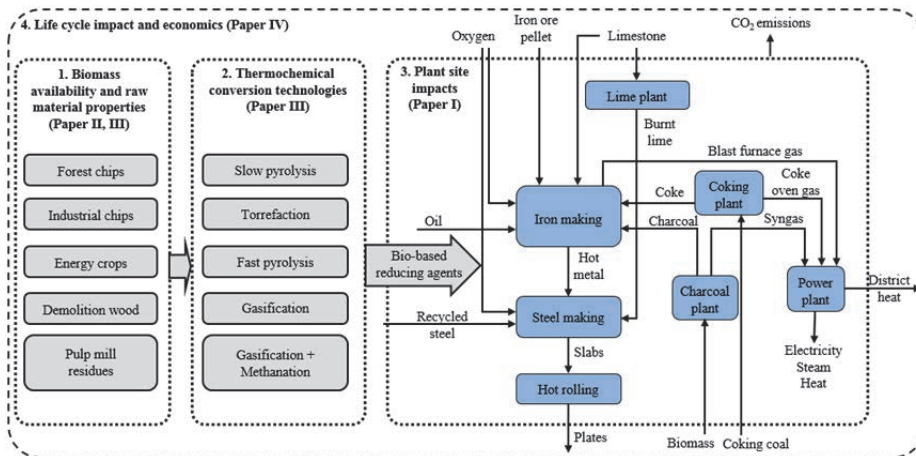


Fig. 1. Research context of the current study.

Table 1. List of original papers and their contribution to the research aim.

Original papers	Contribution to the research aim
Paper I Effects of biomass use in integrated steel plant – gate-to-gate life cycle inventory method	The impact of charcoal as a tuyère injectant on the blast furnace process was evaluated. Additionally, the impact of biomass use on energy balances at plant site scale and CO ₂ reduction potential was evaluated.
Paper II The potential of using biomass-based reducing agents in the blast furnace: A review of thermochemical conversion technologies and assessments related to sustainability	An in-depth review was conducted of the possibility to replace some fossil-based reducing agents with bioreducers in the blast furnace ironmaking route. The paper included thermochemical biomass conversion technology and a review of the resulting product properties. Implications related to bioreducer use were evaluated at separate system levels, including unit process assessment (blast furnace), process integration opportunities, biomass availability, life cycle impact and economic evaluations. Discussion of the future of bioreducers in ironmaking applications was included in the paper.
Paper III Towards More Sustainable Ironmaking – An Analysis of Energy Wood Availability in Finland and the Economics of Charcoal Production	Availability of energy wood (i.e. logging residues, small-diameter wood and stumps) in Finland was evaluated with an emphasis on regional wood availability and possible competing uses. The economics of wood-based charcoal production was compared with fossil-based reducing agents by taking into account the effect of the European Union Emissions Trading System (EU ETS).
Paper IV Bioreducer use in Finnish blast furnace ironmaking – Analysis of CO ₂ emission reduction potential and mitigation cost	An environmental and economic evaluation was conducted of three bioreducers: charcoal, torrefied wood, and wood-based synthetic natural gas (Bio-SNG), produced from logging residues, small-diameter wood and stumps. Additionally, the CO ₂ emission reduction potential of these bioreducers is calculated, using Ruukki (a Finnish steel producer) as a reference. Finally, the CO ₂ emission mitigation cost for each bioreducer was calculated when compared to metallurgical coke.

2 State of the art in CO₂ mitigation in steelmaking

2.1 Steel production routes

Steel can be produced from virgin raw materials or from recycled steel. The majority of steel is produced through primary steel production routes utilizing virgin raw materials. The blast furnace–basic oxygen furnace (BF–BOF) route is the dominant technology route today (Fig. 2 a). The blast furnace is the key technology in which hot metal is produced. Raw materials for a blast furnace must be agglomerated before their charging into the furnace. Alternative technologies for blast furnaces to produce hot metal have been developed in order to avoid the agglomeration processes (coal coking, iron ore sintering or pelletizing). These technologies include e.g. COREX smelting reduction technology (Fig. 2 b). Alternatively, direct reduced iron (DRI) can be produced instead of hot metal, which is further refined into steel in an electric arc furnace (EAF) (Fig. 2 c). Numerous DRI technologies are available, based on shaft furnace, rotary kiln and fluidized bed technologies. In 2012, around 74.0 million tonnes of DRI was produced globally. The majority of DRI (71%) is produced with gas-based technologies such as Midrex and HYL/Energiron (Midrex 2013). Coal-based technologies constitute the rest of DRI production. The production of steel from recycled scrap (Fig. 2 d) is limited by the availability of raw material (Yellishetty *et al.* 2011).

Production of steel requires the use of an extensive amount of energy in the form of reducing agents and electricity. The best practice specific energy consumption (SEC) (primary energy) of the blast furnace–basic oxygen furnace route is 20.4 GJ/t steel (Worrell *et al.* 2008). Other estimates of the BF–BOF route SECs range from 18 to 23 GJ/t steel (World Steel Association 2011, IEAGHG 2013). The smelt reduction route best practice primary SEC is 23.6 GJ/t steel (Worrell *et al.* 2008). Other estimates range from 18.9 to 28.6 GJ/t crude steel (Larsson 2004). The best practice SEC of producing steel from direct reduced iron is 20.6 GJ/t steel (Worrell *et al.* 2008). According to the World Steel Association (2011), energy intensity could be as high as 30.9 GJ/t crude steel. Scrap-based steel production requires much less energy than primary steel production. The best practice primary SEC is 8.0 GJ/t (Worrell *et al.* 2008).

According to the World Steel Association (2011), the energy intensity of EAF steel production is 9.1–12.5 GJ/t crude steel.

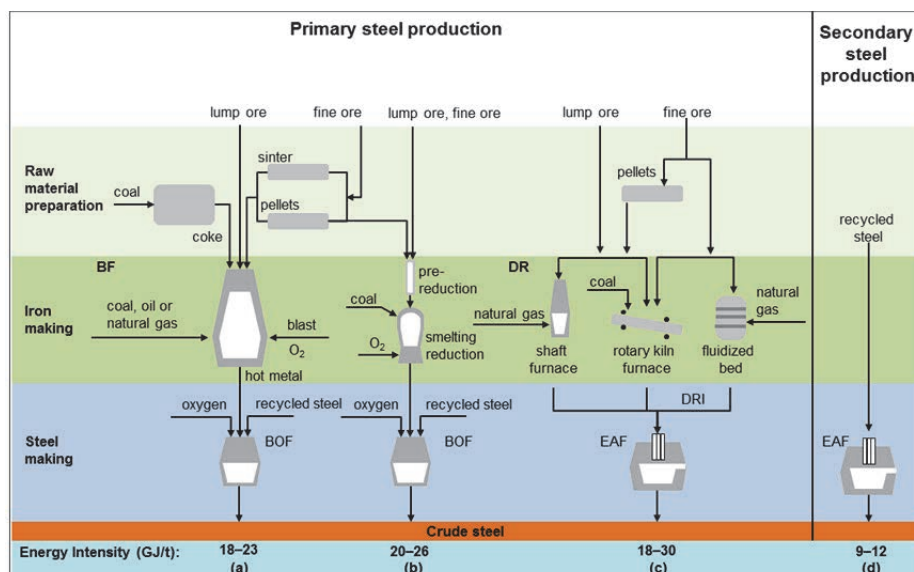


Fig. 2. Steel production routes (according to World Steel Association 2011, IEAGHG 2013).

Reducing agents used today in the iron and steelmaking industry are mainly fossil-based carbon-bearing materials, namely coal, oil and natural gas. According to Birat (2009), direct CO₂ emissions from an integrated steel mill with BF–BOF technology are approximately 1.8 tCO₂/t hot-rolled coil. Majority of the CO₂ emissions in an integrated steel plant result from the use of coke and injected reducing agents in the blast furnace. The total carbon consumption of the blast furnace depends on the input materials used and the efficiency of the process. The typical amount of reducing agent used in European blast furnaces is 300–350 kg of coke and some 150–200 kg of coal per tonne of hot metal produced (Luengen *et al.* 2011). Smelting-reduction technology (e.g. COREX), which has been developed to avoid or at least decrease the amount of coke in the hot metal production, uses some 900 kg of coal and 100 kg of coke per tonne of hot metal produced (Hu *et al.* 2009). According to these values, smelting-reduction technologies do not provide a solution for decreasing the CO₂ emissions of hot metal production. Export gases from the COREX process have a higher volume

and energy content than blast furnace export gases, and with by-product credits COREX might be comparable to BF technology in a CO₂ emission comparison (Hu *et al.* 2009).

According to the International Energy Agency (2007), the CO₂ emissions of the BF–BOF route are 1.7 tCO₂/t crude steel, and the DRI–EAF route based on the use of natural gas produces 1.1 tCO₂/t crude steel. DRI–EAF steel production based on coal use emits 2.5 tCO₂/t crude steel. CO₂ emissions from scrap-based steel production in EAF are approximately 0.4 t/t crude steel.

2.2 Blast furnace process and reducing agents

According to Collins English Dictionary (2013), a reducing agent is defined as: “a substance that reduces another substance in a chemical reaction, being itself oxidized in the process”. In blast furnace ironmaking, iron oxides are reduced and melted into hot metal with fossil-based carbon from coke, pulverized coal (PC), oil, or natural gas (NG), resulting in fossil CO₂ emissions. A blast furnace is large counter-current reactor with a steel stack lined with refractory bricks. The iron burden, coke and limestone are charged from the top of the furnace. Reducing agents (PC, oil or NG) are injected from the lower part of the furnace (the tuyère zone) with oxygen-enriched air (Geerdes *et al.* 2009) (Fig. 3).

Removal of the oxygen from the iron oxides in the upper part of the blast furnace occurs in stages and at different temperatures. Firstly, hematite (Fe₂O₃) is reduced into magnetite (Fe₃O₄), which is further reduced into wüstite (FeO) with carbon monoxide. The reduction of higher iron oxides is based on indirect reduction with reducing gases. The wüstite reduction occurring in the lower part of the furnace happens with two mechanisms: either by indirect reduction with CO gas, or by direct reduction with solid carbon.

Reducing gases needed in the iron oxide reduction are generated by burning coke and injected carbonaceous reducing agents with oxygen-enriched air. Metallurgical coke, produced from coking coal, is the primary fuel and reducing agent in the blast furnace process. Coke is a mechanically strong, porous material that performs three functions in the blast furnace: (1) it acts as a reducing agent; (2) it provides energy to the process; and (3) it serves as a support medium for the burden material (Xu & Cang 2010). The amount of coke that is used in modern blast furnaces is around 300–350 kg/t hot metal, depending on the amount of auxiliary reducing agents used (Luengen *et al.* 2011). By injecting auxiliary

reducing agents into the blast furnace, the amount of coke could be decreased to a level of 200 kg/t hot metal (Danloy *et al.* 2008, Hooey *et al.* 2010).

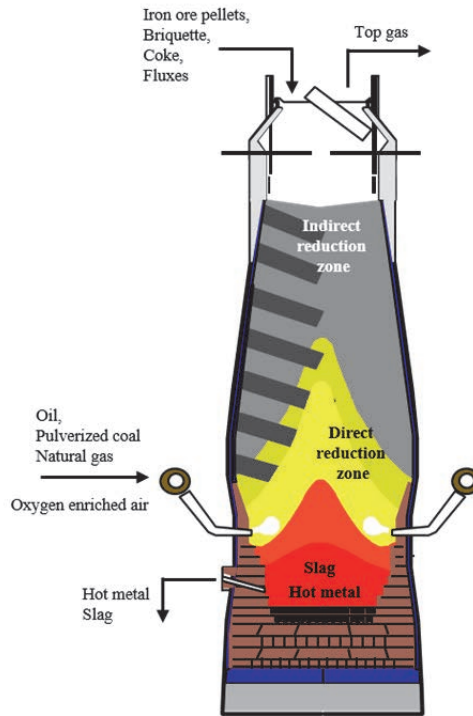


Fig. 3. Schematic presentation of the blast furnace process.

Injected reducing agents are used in the blast furnace because they are cheaper than coke and make the controllability of the furnace more efficient. Pulverized coal is the most used auxiliary fuel in the iron blast furnace. High injection rates can be achieved with pulverized coal, which means that the amount of coke can be decreased substantially. The chemical properties of the coal have a significant impact on the behavior of coal in the blast furnace. Good-quality coal has a high carbon content and a suitable volatile proportion. The amount of ash decreases the heating value of the coal. (Geerdes *et al.* 2009)

Extra-heavy oil is used in the blast furnace as an auxiliary fuel. The injection of oil into the blast furnace became a widely practiced method in the 1960s to increase the productivity and control of the furnace. The oil crisis of the 1970s

changed the paradigm towards coal injection (Mathieson *et al.* 2005). The best values in total reducing agent consumption in European blast furnaces with oil injection (458.5 kg/t hot metal) have been achieved by the Finnish steel mill Ruukki, with 358.0 kg coke and 100.5 kg oil/t hot metal (Luengen *et al.* 2011).

Natural gas is used as a reducing agent in countries where it is inexpensive. The largest volume of injected natural gas has been achieved in the USA, with 155 m³/t hot metal (Babich *et al.* 2002). Higher injection rates of natural gas lead to local supercooling of the furnace hearth. Plastics and coke oven gas are also reducing agents used in blast furnaces (Babich *et al.* 2002).

2.3 CO₂ breakthrough programs

Carbon dioxide emissions are the main greenhouse gas emissions from the steel industry. Reductions in CO₂ emissions have been achieved by enhanced material and energy efficiency, process integration and increased utilization of by-products such as slags (Johansson & Söderström 2011, Larsson 2004, Makkonen *et al.* 2002). Major reductions in energy and reducing agent consumption have been achieved with these measures, and further reduction is difficult. In recent years, large programs have been initiated to further cut CO₂ emissions with new technologies that would replace the blast furnace.

2.3.1 Ultra-Low Carbon Dioxide Steelmaking

The Ultra-Low Carbon Dioxide Steelmaking (ULCOS) program was initiated in 2004, with the aim of reducing CO₂ emissions by at least 50% per tonne of steel. Almost 50 companies and organizations have been involved with the ULCOS program. During the first years of the ULCOS program, several existing and emerging technologies were reviewed. Alternative technologies were evaluated from CO₂ emission, technical, economic and social viewpoints. Four innovative technologies were selected for further development: (1) the top gas recycling blast furnace; (2) HIsarna, a new smelting reduction process; (3) ULCORED, advanced direct reduction; and (4) ULCOWIN, electrolysis of iron ore (Meijer *et al.* 2009).

In the ULCOS **top gas recycling blast furnace** (TGR-BF or ULCOS-BF), four different versions with alternative top gas recycling schemes were developed. All the versions include the injection of recycled gas into the BF hearth via tuyères. In versions 1 and 4, recycled gas is also injected through the shaft tuyères. The temperature of the injected gas varies, but in all cases pure

oxygen and coal injection are applied. Concerning only the blast furnace system, the highest carbon savings (about 25%) compared to the base case blast furnace are expected to be achieved with version 4 (Fig. 4). In this version, heated, recycled gas is injected from the tuyères at hearth and shaft level. Nitrogen-free top gas makes the recovery of CO₂ from the top gas and storage possible. Carbon capture and storage (CCS) is needed to reach the 50% reduction target in CO₂ emissions (van der Stel *et al.* 2013, Danloy *et al.* 2008).

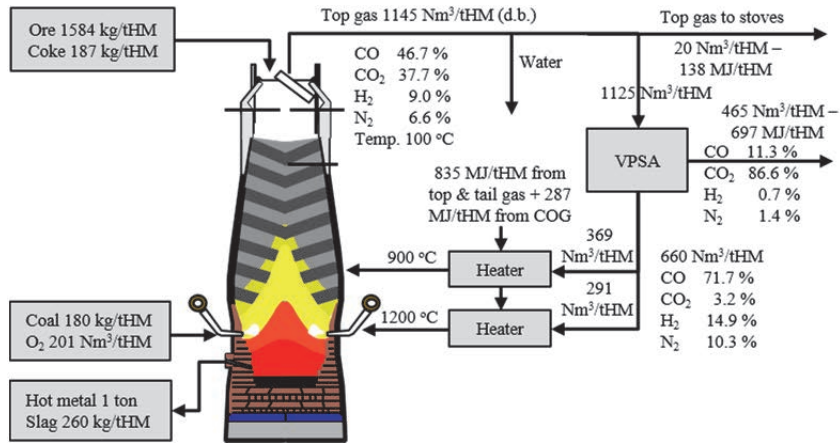


Fig. 4. Top gas recycling blast furnace (version 4) (according to Danloy *et al.* 2008).

Blast furnaces are usually part of the steel production route in an integrated steelmaking plant. For conventional blast furnace operations, blast furnace gas is used to produce hot blast air, and extra BFG is often used in power plants to produce electricity to the system. In the case of the TGR-BF, there will be a deficit of BFG flow to the power plant because most of the top gas is recycled back to the BF. The realizable carbon saving should therefore be evaluated with extended system boundaries.

The fact is that top gas recycling does not reduce the specific energy consumption of iron oxide reduction in the blast furnace. However, it changes the way that carbon from coke and injected reducing agents is utilized. In the TGR-BF process, carbon is more efficiently utilized in reduction application (Hooey *et al.* 2013). Carbon savings with extended system boundaries have been calculated in the ULCOS program (van der Stel *et al.* 2013). According to the calculations

with integrated steel plant system boundaries, ULCOS-BF showed a maximum of 15% CO₂ emission reduction potential per tonne of hot rolled coil. When CCS is applied, a 60% decrease in the direct emissions of an integrated steel plant can be achieved. In the study, the BFG deficit in the steel plant was assumed to be fulfilled with natural gas. No reliable life cycle assessment study for a steel product (hot rolled coil) produced via TGR-BF route could be found in the literature.

The second technology studied in the ULCOS project to radically reduce CO₂ emissions is **Hisarna**. This technology is based on the combination of two technologies: Cyclone Converter Furnace and HIs meltTM Smelt Reduction Vessel. These two technologies are incorporated into a single vessel in which iron ore fines are reduced into hot metal, comparable to blast furnace hot metal (Meijer *et al.* 2011). According to model calculations from Link (2008), the coal consumption of Hisarna would be some 480 kg/tHM, which can be considered low compared to other smelting reduction technologies such as COREX. Hisarna technology is still in the development stage. The first pilot plant has been constructed and trials have been conducted. According to Meijer *et al.* (2011), the feasibility of Hisarna originates from: (1) the possibility to use thermal coals instead of metallurgical coals; (2) the possibility to use low-quality iron ore feed; and (3) 20% primary energy and CO₂ savings without geological storage.

ULCORED is an enhanced direct reduction technology concept with several advantages compared to state-of-art DR processes. Direct reduced iron is produced in a shaft furnace by removing the oxygen from the iron ore without melting it. In the ULCORED process, modifications have been made to enhance the efficiency and CO₂ intensity of steel production. The concept includes e.g. the use of 100% oxygen and partial oxidation instead of reformers. The ULCORED process has been designed to operate either with natural gas or syngas produced e.g. from coal, waste plastic, or biomass. It is estimated that a 20% energy demand reduction could be achieved compared to existing DR processes. (Knop *et al.* 2008)

Iron can also be produced by electrolysis of iron ore. In ULCOS, this technology presents the most “breakthrough” route to decrease the CO₂ emissions of steel production. In the **ULCOWIN** process, iron is produced in aqueous alkaline electrolysis. In electrolysis, no coal-based reducing agents are needed, but a vast amount of electricity is required. CO₂ emissions can be radically decreased if CO₂-lean electricity is available. Electrolysis yields oxygen as a by-product that can be utilized in other industrial applications. (Meijer *et al.* 2009)

The breakthrough technologies being considered in the ULCOS program are in different commercialization stages. According to estimates by Birat (2011), ULCOS-BF and ULCORED could be commercialized in 2020, HIsarna in 2030, and ULCOWIN in 2040.

2.3.2 Other CO₂ breakthrough programs

The American Iron and Steel Institute (AISI) has run several programs since 2004 dedicated to create a breakthrough in technology development. Two technologies have been selected for further development: ironmaking by molten electrolysis and ironmaking by hydrogen flash smelting. Molten electrolysis is based on iron production with electrolysis in which an electric current passes between two electrodes in a molten salt solution containing dissolved iron oxide. The operating temperature of the process is almost 1,600 °C and no greenhouse gases are generated besides those from the electricity production. CO₂ emissions for steel produced could be as low as 345 kg CO₂/tonne liquid steel. Hydrogen flash smelting is an ironmaking process similar to the flash smelting furnace used in copper production. Reducing agents that are suitable for this furnace are hydrogen, natural gas and synthetic gas produced from coal or waste plastics. According to the calculations, hydrogen flash smelting requires 38% less energy than the blast furnace, and its CO₂ emissions are only 4% of those generated in the blast furnace when hydrogen is used as a reducing agent (Vehec 2011).

In Japan, the COURSE 50 program started in 2008 with the aim of decreasing steel production CO₂ emissions by approximately 30%. This will be achieved by developing technologies to reduce CO₂ emissions from blast furnaces and by developing technologies to capture, separate, and recover CO₂ from blast furnace gas. Blast furnace technology developments include the use of hydrogen as reducing agent, reforming of coke oven gas by utilizing unused waste heat, and the development of high-strength and high-reactivity coke for reduction with hydrogen. Carbon capture and storage technology developments include the further development of chemical and physical absorption and more efficient utilization of unused waste heat from steel plants in CO₂ separation and recovery (Miwa *et al.* 2011).

POSCO, a Korean steel producer, has its own CO₂ Breakthrough Framework to develop technologies to decrease CO₂ emissions (Birat 2011). Several breakthrough technologies, such as CO₂ capture, slag heat recovery, power generation by recovering waste heat from stacks, and hydrogen steelmaking, are

being developed (POSCO 2011). The Australian CO₂ Breakthrough Program has been running since 2006, with a special focus on the utilization of biomass as a reducing agent and the utilization of slag waste heat (Jahanshahi *et al.* 2011, Mathieson *et al.* 2011).

3 Bioreducers

Biomass-based reducing agents – *bioreducers* – are defined as solid, liquid or gaseous materials produced from biomass that are suitable for iron oxide reduction in the blast furnace. Bioreducers can be used in several forms in the blast furnace. Biomass-based components could be introduced to the process from the top of the furnace, or from the bottom of the furnace via tuyère injection (Fig. 5). Identified top-charged inputs enabling the introduction of bio-carbon into the blast furnace include: (a) biocoke, which is defined here as the coke produced from the mixture of the coking coal and biomass-derived, heat-treated char; (b) lump charcoal; (c) carbon composite agglomerates; and (d) ferrocoke, in which part of the fossil-based carbon would be replaced with biomass-based carbon. Injected bioreducers include: (e) torrefied biomass; (f) charcoal; (g) bio-oil; (h) syngas; and (i) Bio-SNG.

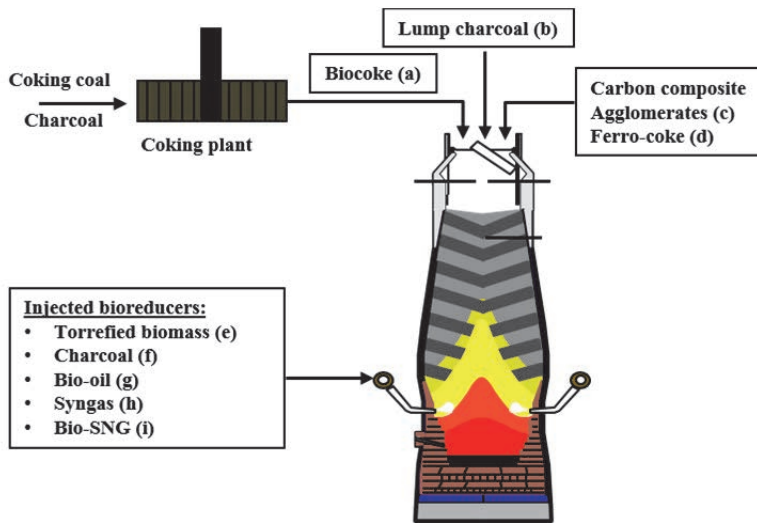


Fig. 5. Bioreducers in the blast furnace.

Top-charged and injected reducing agents have different requirements. Top-charged reducing agents should be mechanically strong and contain as little volatile matter as possible. Low mechanical strength results in dust generation, which causes problems in blast furnace top gas cleaning systems. Generation of the fines in the upper part of the furnace, inside the iron ore and coke bed, can

decrease the reducing gas permeability, which in turn results in insufficient reduction of the iron oxides. Injection could provide more flexible possibilities to replace part of the fossil-based reducing agents with bioreducers. Mechanical strength is not necessary; the most important factors are good burning efficiency and the chemical properties of the injected bioreducer.

As blast furnaces have been operated for decades, raw material properties and process parameters have been optimized so that smooth running and productivity are achieved. Resulting from this, new alternative reducing agents applied in BF ironmaking should be comparable to those reducing agents used in normal operation. In the following sub-sections, the state of the art concerning bioreducer use in the blast furnace is presented.

3.1 Top-charged bioreducers

Biocoke (a) is a product, which is produced from a blend of coal and biomass (Ng *et al.* 2011). Biocokes have been produced from different raw materials and tested with procedures familiar from coke research. Pine and chestnut sawdust (Montiano *et al.* 2014), eucalyptus and olive wood (Diez & Borrego 2013) torrefied wood (Thomas *et al.* 2011), charcoal from different wood species (Diez & Borrego 2013, MacPhee *et al.* 2009) and tar (Diez *et al.* 2012) have been used as additives in biocoke production. During the coking of bituminous coal, the coal goes through different physical changes; softening, melting, liquefaction, devolatilization, vesiculation, hardening into coke, and further devolatilization (Crelling 1989). Because the biomass component acts as an inert material in the coal blend in regard to softening and melting (Diez *et al.* 2012), the amount of biomass addition to the coal blend is modest. It has been proposed that some 5% of the coal could be replaced with biomass-derived charcoal without worsening the quality of the coke too much (MacPhee *et al.* 2009). With larger amounts, the mechanical strength of the bio-coke produced decreases significantly.

The main reason for adding a biomass-based component to the coal blend is to avoid the fossil CO₂ emissions resulting from blast furnace reduction reactions. However, there could also be other benefits associated with biocoke use. It has been reported that biocokes are significantly more reactive than cokes produced from coal (Ng *et al.* 2011, MacPhee *et al.* 2009, Diez & Borrego 2013). It has been shown that increased reactivity of the coke with CO₂ (the solution loss reaction $C + CO_2 \rightarrow 2CO$) in the blast furnace improves the reaction efficiency and shifts the thermal reserve zone to lower temperatures (Nomura *et al.* 2005).

With a lower thermal reserve zone temperature, the equilibrium concentration of the FeO-Fe reduction reaction is shifted to higher CO gas utilization efficiency. Overall, this results in more efficient utilization of the input carbon in the reduction process (CO gas utilization efficiency at the furnace top) and a further decrease in the reducing agent rate (Nomura *et al.* 2005, Kasai & Matsui 2004).

Lump charcoal (b) charging could be used to replace some 20% of the top-charged coke in modern, large-scale blast furnaces (Norgate & Langberg 2009). Top-charging of lump charcoal was investigated in the European Commission-funded SHOCOM project as a possible short-term CO₂ mitigation option in steelmaking (Hanrot *et al.* 2011). The amount of top-charged lump charcoal in the study was only 20 kg/tHM, but it resulted in calculated coke savings of about 30 kg/tHM (Hanrot *et al.* 2009). The coke saving is a result of a lower reserve zone temperature, which increases the efficiency of the furnace. It was also argued that top-charged charcoal would be consumed in the upper part of the furnace and the possible permeability problem is avoided.

Top-charging of the charcoal is a common practice in mini blast furnaces in Brazil (Noldin 2011). Charcoal can be used as a single reductant in mini blast furnaces, which are considerably smaller than modern, large blast furnaces. The typical working volume of a mini blast furnace is 50–350 m³ (Noldin 2011). The largest blast furnaces in Europe have working volume of 4,800 m³ (Luengen *et al.* 2011) and even larger blast furnaces have been constructed in Asian countries. Two blast furnaces in Finland have working volumes of 1,200 m³ (Kinnunen *et al.* 2011). The largest charcoal-operated blast furnace in Brazil has a working volume of 570 m³ (Poveromo 2013). According to Goncalves *et al.* (2012), quite large blast furnaces (570 m³) can be operated without major problems.

Carbon composite agglomerates (c) can be used as an umbrella term for materials that are produced by agglomerating carbon- and iron-bearing materials. According to Ahmed *et al.* (2013), the iron source is fine iron ore and the carbonaceous material can be fine coke, fine coal or charcoal. Pellets can be cold-bonded with or without a binder; briquettes can be hot- or cold-pressed. In Japan, the development of ferrocoke has attracted interest in recent years (Takeda *et al.* 2011). **Ferro-coke (d)** is a composite consisting of metallic iron, iron oxide and carbon. The rationale for using coal-iron ore agglomerates in the blast furnace lies in the lowering of the reduction equilibrium temperature, as in the case of using reactive coke. With carbon composite agglomerates, a faster reduction and gasification reaction is achieved by arranging the iron ore and carbonaceous materials closely together (Yokoyama *et al.* 2012). Charcoal has been used as a

carbon source in composite pellet production at the laboratory scale (Konishi *et al.* 2010), while carbon composites made from coke and iron oxides, bonded with cement, have been trialed in a commercial blast furnace in Japan with excellent results concerning the consumption of reducing agents in the blast furnace (Yokoyama *et al.* 2012).

3.2 Injected bioreducers

Green biomass was studied as a possible injected reducing agent by Chen & Wu (2009). The research method was to utilize a drop tube furnace (DTF) to simulate the extremely rapid heating rates (10^4 – 10^5 K/min) experienced by the injected char particles in the blast furnace raceway area. Pulverized coal was partially replaced with rice husk, which is a by-product of rice production. According to the results, co-injection of PC and biomass should be possible from the viewpoint of char burnout. However, full mass and energy balance calculation of the various effects of changing raw material should be conducted. Green biomass differs greatly from the pulverized coals used in the blast furnace. The proportion of carbon is low and the oxygen content high, which results in a low higher heating value (HHV).

Torrefied biomass (e) injection into the blast furnace has been studied with laboratory investigations, modeling and simulation. Chen *et al.* (2012) have evaluated the combustibility of torrefied biomass with DTF experiments. Biomasses torrefied at temperatures of 300 °C showed comparable burning behavior to high volatile bituminous coal. According to recently conducted optimization studies (Wiklund *et al.* 2012, Wiklund *et al.* 2013), optimal heat-treatment temperature with oil-biomass co-injection is in the range of torrefaction temperatures. However, the pyrolysis unit has been modeled not taking into account possible by-products, which might have a considerable impact on the optimization results. Mathieson *et al.* (2011), conducted a modeling study with different injected reducing agents. According to this study, injection of torrefied wood as the sole injected reducing agent results in a considerable coke rate increase compared to injection of pulverized coal. The high level of volatile matter of torrefied wood leads to cooling of the raceway and a requirement for increased oxygen enrichment of the blast. It seems that torrefied biomass could be used co-injected with coals and in situations when BF productivity could be lower.

Charcoal (f) is the most studied bioreducer for blast furnace injection. This is because charcoal has chemical properties comparable to pulverized coal (Machado *et al.* 2010a). Laboratory investigations, such as thermogravimetric, DTF and injection rig studies, have been used to examine the burning properties of different charcoals (Babich *et al.* 2010, Machado *et al.* 2010a, Mathieson *et al.* 2012). The combustibility of the charcoal in the raceway region has been examined also by simulation study (Wijayanta *et al.* 2014). Several papers concerning the numerical modeling and simulation of charcoal injection have been published in recent years (Babich *et al.* 2010, Mathieson *et al.* 2011, de Castro *et al.* 2011, de Castro *et al.* 2013)

Laboratory investigations have indicated that charcoal injection could be feasible from the technology point of view. The combustion behavior of the charcoals is comparable to that of pulverized coals and, according to pilot tests conducted by Babich *et al.* (2010), combustion efficiency is even higher. The thermochemical behavior of the charcoals has been found to be superior to pulverized coals. However there are some aspects that might be problematic in industrial-scale use of charcoal in the blast furnace (Mathieson *et al.* 2012). Grinding the charcoal to a specified particle size might be difficult to achieve. Another difficulty might arise from the low density of the charcoals. High injection rates of charcoal might be difficult to realize without modifications to conveying systems.

Modeling results over the whole blast furnace have shown that charcoal has a higher coke replacement ratio than pulverized coal. The coke replacement ratio is defined as the amount of metallurgical coke replaced by the injected reducing agent. Babich *et al.* (2010) used mathematical modeling to investigate the impact of pulverized coal and charcoal injection. In the PC injection case (200 kg/tHM), the coke consumption was 283.0 kg/tHM. The minimum coke consumption with 200 kg/tHM charcoal injection was 257.2 kg/tHM. Similar results have been reported by Mathieson *et al.* (2011) and de Castro *et al.* (2011). In all of the papers, good-quality charcoal outperforms pulverized coal injection. There are several reasons for this: The low amount of ash and high basicity (CaO content) of the charcoal result in a decrease in slag volume and blast volume, which results in a reduced coke rate and an increase in blast furnace productivity (Babich *et al.* 2010). The low amount of ash, low sulfur content, and high basicity of the charcoal can also result in a lower demand for limestone in the blast furnace. Decomposition of limestone requires energy from the coke, and with the use of charcoal, coke consumption could be decreased. From the laboratory and

modeling work conducted by various researchers, it can be concluded that charcoal could be used as a single injected reducing agent in the blast furnace.

The suitability of **bio-oil (g)** as a blast furnace injectant has been evaluated only in one publication. Ng *et al.* (2010) examined bio-oil injection in the blast furnace with mathematical modeling. Injecting 140 kg/tHM bio-oil into the BF resulted in coke consumption of 455 kg/tHM, whereas in the base case with pulverized coal injection of 140 kg/tHM, the coke consumption was 370 kg/tHM. From this calculation, a coke replacement ratio of 0.25 can be concluded.

Injection of **syngas (h)** produced from biomass into the blast furnace could be one alternative. Hot reducing gas injection has been considered as one possibility to develop BF technology further. Coke oven gas (COG), gas from coal pyrolysis, COREX process gas and BF top gas are examples of gas sources found suitable for BF injection (Babich *et al.* 2002, Hooey *et al.* 2010, Ziebig *et al.* 2008). Analysis of biomass-derived syngas behavior in the BF as a reducing agent has not been conducted to date. The possibility of using syngas from biomass gasification has been investigated in the work of Bürgler and Di Donato (2008).

Biomass-based synthetic natural gas (Bio-SNG) (i) could also be suitable gaseous reducing agent for BF injection (Suopajärvi *et al.* 2013). Bio-SNG has similar properties to NG, and there should be no problems in deploying Bio-SNG in the BF. The amounts of NG used in blast furnaces vary, but the highest rate achieved is 155 kg/tHM (Babich *et al.* 2002). This figure is exceptionally high; a more realistic figure would be nearer 90 kg/tHM. With higher NG injection amounts, the adiabatic flame temperature starts to become low. The coke replacement ratio of natural gas reported in the literature is 0.9–1.15 (Worrell *et al.* 2010).

3.3 Commercial use of bioreducers in BF

Bioreducer use today at the industrial scale is limited to the mini blast furnaces operated in Brazil. Mini blast furnaces in Brazil are considerably smaller than coke-based blast furnaces. Charcoal is used as the reducing agent and the iron burden is almost exclusively small fractions of lump ore (Noldin 2011). The fuel rate is somewhat higher in charcoal BF than with coke BF. The amount of slag in charcoal BF is considerably lower than in coke BF. The amount of slag in charcoal BF is around 150–160 kg/tHM (Nogami *et al.* 2004, Goncalves *et al.* 2012), while the amount of slag in Brazilian blast furnaces is around 250 kg/tHM, and around 200 kg/tHM in European blast furnaces (Paananen *et al.* 2011).

Industrial use of biomass in European blast furnaces has not been practiced since fossil reducing agents replaced charcoal. A pilot trial with biomass pellet injection was run at the Voestalpine Linz plant in 2010. Some 1,000 tonnes of wood pellets at rates of 20 and 30 kg/tHM were injected without technological problems with a coke replacement ratio of 0.35 (Bürgler *et al.* 2011).

3.4 Biomass raw materials for reducing agent production

Biomass has a strong role e.g. in Finnish renewable energy targets, and there are intentions to increase the use of bioenergy further. Increasing the use of bioenergy raises questions about the availability of biomass and efficient upgrading technologies. The availability of biomass for iron- and steelmaking was evaluated by Piketty *et al.* (2009) in the Brazilian context. They evaluated the availability of the land for short rotation wood (eucalyptus) plantations. In the Nordic countries, the annual growth of wood is much lower than e.g. in Brazil because of the shorter growing season. The average annual growth of trees in Finland is below $5 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$ (Alam *et al.* 2010), whereas in Brazil the annual growth of eucalyptus can be as high as around $40 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$ (Piketty *et al.* 2009). Despite the slow growth rate in Finland, the amount of wood in Finnish forests is continually increasing. This wood is mainly utilized as a raw material in the wood-processing, in the pulp and paper and in the energy industries.

Biomass-based raw material resources are vast. The majority of wood-based energy is recovered from liquid black liquor (Ylitalo 2013), which is a by-product from pulp production. In addition to black liquor, various solid residues such as forest chips, bark, sawdust and industrial wood residues are produced during different stages of wood processing. These wood fractions are used today in energy production, but could also be raw material for bioreducer production.

Other forms of biomass, such as straw, do not contribute significantly to Finnish energy production. Forest chips produced from logging residues, stumps and small-diameter wood is the source of wood that has the most significant increase potential in Finland. In 2012, the use of forest chips was 7.6 million m^3 (15.2 TWh) of which 3.6 million m^3 came from small-diameter wood, 2.2 million m^3 from logging residues, 1.1 million m^3 from stumps and roots, and the rest from large-sized timber (Metla 2013). A considerable increase in the utilization of forest chips in energy production is planned in the future. The target set by the Ministry of Employment and the Economy (2010) is to increase the use of forest chips in energy production from the current level to 25 TWh in 2020.

3.5 Thermochemical conversion technologies for biomass upgrading

Several thermochemical biomass conversion technologies have been developed to enhance the properties of biomass. The main thermochemical conversion technologies for biomass upgrading are presented in Fig. 6. With thermochemical conversion technologies, solid, liquid, and gaseous products can be produced from biomass. Alongside the main product, which depends on the conversion technology, usually all the other forms of the matter are produced. For example, in slow pyrolysis where the main product is charcoal, pyrolysis gas and various tars are also produced.

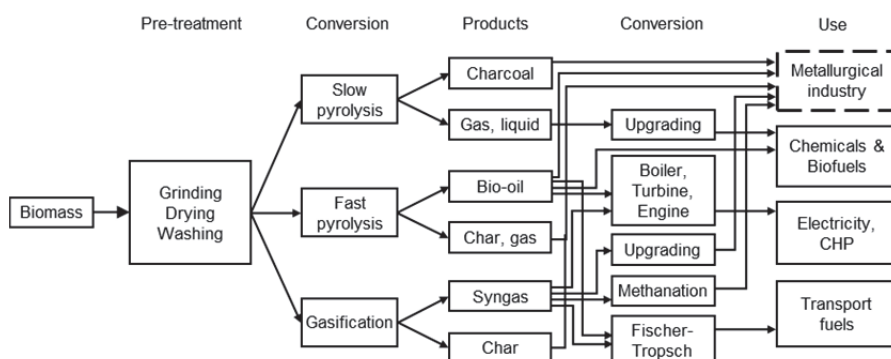


Fig. 6. Thermochemical conversion technologies to produce biofuels from biomass (Paper II, reprinted by permission of Elsevier).

Typical product weight yields for pyrolysis of wood are presented in Table 2.

Table 2. Typical product weight yields (db) of pyrolysis of wood (Bridgewater 2012).

Mode	Conditions	Liquid	Solid	Gas
Fast	~ 500 °C, short hot vapor residence time ~ 1 s	75%	12% char	13%
Intermediate	~ 500 °C, hot vapor residence time ~ 10–30 s	50% in 2 phases	25% char	25%
Carbonization	~ 400 °C, long vapor residence hours to days	30%	35% char	35%
Gasification	~ 750–900 °C	5%	10% char	85%
Torrefaction	~ 290 °C, solids residence time ~ 10–60 min	0% unless condensed, then up to 5%	80% solid	20%

Torrefaction and slow pyrolysis are heat-treatment technologies for biomass in the absence of oxygen with modest heating rates and long vapor residence times. In torrefaction, the final temperature is 230–330 °C, and in slow pyrolysis usually 300–900 °C (Antal & Gronli 2003, Prins *et al.* 2006). The main product from both processes is solid char. Torrefaction enhances the calorific value and grindability of the biomass (Phanphanich & Mani 2011). The higher heating value (HHV) of the torrefied biomass depends on the temperature and becomes quite close to that of fossil coal when the temperature is 300 °C (Chen *et al.* 2012). Good-quality charcoal has a higher carbon content and a lower proportion of volatiles than pulverized fossil coals (Mathieson *et al.* 2011).

In torrefaction, mass and energy yields are high, because torrefaction temperatures are kept at lower levels. The mass yield of logging residue in torrefaction at temperature of 225 °C is around 88%, and 52% at 300 °C from dry wood. The energy yields are 92% and 72%, respectively (Phanphanich & Mani 2011). When the temperature is further increased to 450 °C, the mass yield drops to a level of 30% and the energy yield to around 50% (Fagernäs *et al.* 2012). It has been suggested that the thermal efficiency of torrefaction could be as high as 96% (Uslu *et al.* 2008); on the other hand, commercial torrefaction could be realized at 90% efficiency or lower (van der Stelt *et al.* 2011). According to values presented by Roberts *et al.* (2010), the energy efficiency of biomass to charcoal is 55%, and the energy efficiency of biomass to biochar and heat is almost 84%. This number does not include power demand, but does include utility fuel consumption. Several concepts to enhance the efficiency of the slow pyrolysis system have been proposed, including production of Bio-SNG or methanol from pyrolysis gases (Larsson *et al.* 2013, Shabangu 2014).

In fast pyrolysis of biomass, higher heating rates (10^3 – 10^4 K/s) are used, aiming for the maximum yield of the liquid fraction (Zhang *et al.* 2007). The temperature in fast pyrolysis is also modest, as in slow pyrolysis, around 500 °C, but the vapor residence time is short (Bridgewater 2003). The chemical properties of the bio-oil resemble that of biomass. Bio-oil has high oxygen and water content. The HHV of bio-oil produced from wood-based feedstock is around 16–19 MJ/kg (Mohan *et al.* 2006). The main benefits of producing liquids from biomass are increasing the energy intensity, storability and transportability of the product. The thermal efficiency of fast pyrolysis is around 60–70% (Uslu *et al.* 2008, Scott *et al.* 1999). Fast pyrolysis technologies are gradually being commercialized. In Finland, a 50,000-tonne bio-oil production plant integrated with a combined heat and power plant was commissioned in 2013 (Fortum 2013).

Gasification refers to the thermochemical conversion of biomass to gaseous form by using a gasification medium consisting of air, oxygen, steam, nitrogen, carbon dioxide, or a combination of these (Kumar *et al.* 2009). Gas of medium calorific value (12–18 MJ/Nm³) can be produced with oxygen or steam gasification (McKendry 2002). The composition of the gas depends highly on the technology applied and the gasification agent. Syngas appropriate for blast furnace use should contain a minor amount of nitrogen. Additionally, the proportion of CO + H₂ in the gas should be as high as possible, in the range of 90% (Suopajarvi *et al.* 2013). This requires the use of oxygen/steam as the gasification agent and removal of water and carbon dioxide from the syngas. Gasification as such is a considerably efficient processing method. The hot gas efficiency (chemical and sensible heat in raw product gas divided by the energy in the feed) can be as high as 95–97%, and up to 85% for cold gas efficiencies (Bridgewater 2003). The overall efficiency is lower due to the need for energy to dry the biomass and power demands.

Bio-SNG can be produced from biomass feedstock by utilizing gasifier technology as the first step in the chain. After the biomass has been converted to gas, it is cooled and tars are removed. The gas is then cleaned of pollutants. CO and H₂ in producer gas are then converted to CH₄, CO₂ and H₂O in methanation units. Further upgrading includes water and CO₂ removal and compression of the gas. The net overall efficiency from biomass to SNG can be up to almost 70% (van der Meijden *et al.* 2010).

3.6 Environmental and economic considerations of bioreducers

3.6.1 CO₂ emissions of bioreducers

Few life cycle assessments were found where the CO₂ emission profile of the possible bioreducers was evaluated. CO₂ emissions from charcoal production were analyzed by Roberts *et al.* (2010), Norgate *et al.* (2012) and Sjolie (2012), from bio-oil production by Steele *et al.* (2012) and Fan *et al.* (2011), and from Bio-SNG production by Hacetoglu *et al.* (2010) and Steubing *et al.* (2011). The results of these studies were calculated on a gCO₂e/MJ basis to provide comparability of the results and are presented in Fig. 7.

The CO₂ emission figures presented in Fig. 7 can be taken as indicative when comparing the environmental performance of different bioreducers, since the

underlying assumptions, raw materials, technologies and geographical locations are different in the LCA studies. One of the main factors influencing heavily the magnitude of the CO₂ emissions of bioenergy systems is indirect land use change (ILUC). ILUC emissions occur when expansion of biofuel demand leads to deforestation or conversion of grazing land to crop cultivation (Mathews & Tan 2009). According to Plevin *et al.* (2010), ILUC emissions may even negate the climate benefits of some biofuels, especially if feedstock used to produce the biofuel competes with food production land area. The impact of ILUC is taken into account in the case of slow pyrolysis of switchgrass A and B by Roberts *et al.* (2010). CO₂ emissions are considerably higher compared to two other raw materials (late stover and early stover) in the same study, which do not include ILUC emissions. CO₂ emissions vary significantly between 3.4 and 45 gCO₂e/MJ of product if ILUC is not taken into account. In switchgrass scenario B, CO₂ emissions are 119 gCO₂e/MJ biochar. The CO₂ emissions of bioreducers are moderate compared to fossil-based reducing agents, e.g. coal (115 gCO₂/MJ) and natural gas (75 gCO₂/MJ) (Burnham *et al.* 2012), if ILUC is not considered.

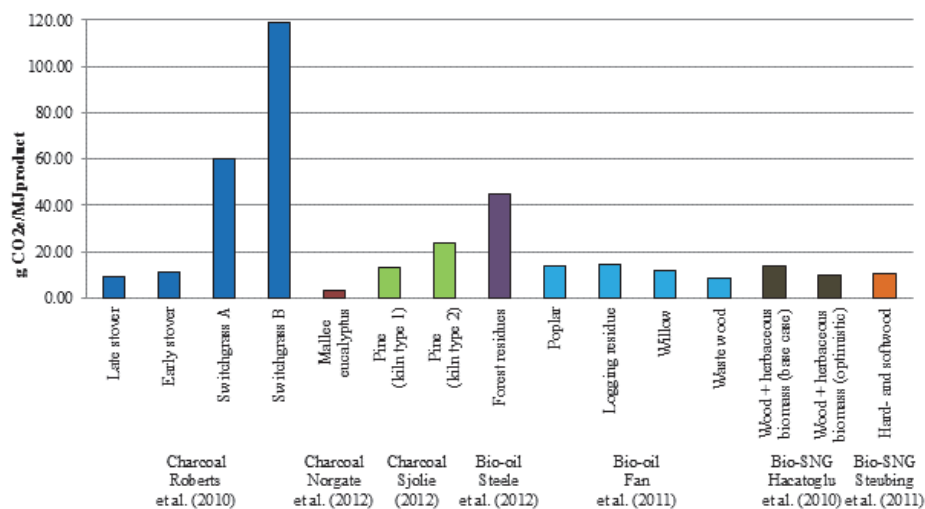


Fig. 7. Carbon footprint of bioreducers (Paper II, reprinted by permission of Elsevier).

3.6.2 Energy return on investment of bioreducers

Energy consumption of bioreducer production revealed that there is true potential in the utilization of bioreducers in steel industry. In contrast to biofuels such as corn ethanol, which may have very low EROI of around 1.0 and in some cases below 1.0 (Murphy & Hall 2010), the EROI of analyzed bioreducers is well above 1.0 (Gaunt & Lehmann 2008, Steele *et al.* 2012, Bailis *et al.* 2013, Hacatoglu *et al.* 2010, Roberts *et al.* 2010), which means that production is energetically feasible. The highest EROI found in the literature was reported for charcoal, at 15–35, depending on the technology (Bailis *et al.* 2013). The net energy ratio (a concept similar to EROI) of biochar was 2.8–3.1 in the work of Roberts *et al.* (2010), and 5.3–6.9 in the work of Gaunt and Lehmann (2008). Hacatoglu *et al.* (2010) evaluated the EROI of Bio-SNG with different production technologies and plant capacities. The EROI was lower with larger capacities and higher with advanced technology, ranging from 5.1 to 8.0. Direct EROI estimates for bio-oil could not be found in the literature. However, Steele *et al.* (2012) estimated that production of 1 MJ of bio-oil requires 0.52 MJ of energy input, taking into account energy from renewable sources as well. According to the literature review by Raugei *et al.* (2012), the EROI of oil and natural gas have decreased from above 100 to approximately 20. The EROI values for coal are in the range of 40 to 80. It can be seen that the production of fossil-based fuels is energetically more feasible than bioreducer production, which is mainly because of more efficient production technologies. Biomass harvesting, transportation and pre-processing require a substantial amount of energy per unit of energy produced.

3.6.3 Production cost of bioreducers

The charcoal production cost in Brazil, where short rotation forests are used as raw material, is around €193/t charcoal. In Australia, the production cost has been approximated at US\$386/t charcoal (Noldin 2011, Norgate & Langberg 2009). If it is assumed that the HHV of charcoal is 31 GJ/t, the cost of charcoal would be €6.2/GJ and US\$12.45/GJ. The bio-oil production cost without feedstock cost in a 40 MW capacity fast pyrolysis plant has been calculated at €75–150/t and €6–12/GJ bio-oil, depending on the cost data of capital investment (Uslu *et al.* 2008). Other estimates range from €8 to €19.6/GJ (€130–318/t), with feedstock

costs ranging from €0 to €100/t. The capacity of the plant was 2t/h (Peacocke *et al.* 2006).

Hacatoglu *et al.* (2010) have estimated the production cost of Bio-SNG for different scales of operation. The highest production cost was US\$25/GJ₂₀₀₅ with a small unit size (dry biomass input of 500 t/day) and with current technology. The lowest production cost was US\$16/GJ₂₀₀₅ with a large unit size (dry biomass at 5,000 t/day) and with more advanced technology. The production cost of syngas has not been reported in the literature. However, it can be approximated to be lower than Bio-SNG production cost.

4 Materials and methods

The methods used in this thesis are, for the most part, based on the toolbox of industrial ecology described in section 1.2. Several methods were used to fulfill the objectives of the thesis. Since the blast furnace is the most important unit process influenced by possible reductant change, the evaluation of the chemical and physical properties of bioreducers is of great importance. The metallurgical properties of possible bioreducers were evaluated based on the established body of scientific literature and compared to fossil-based reductants. Industrial trials with alternative reductants are costly and difficult to arrange without interruptions to normal operation. Therefore, a process modeling and simulation approach was used to evaluate the impact of charcoal injection on blast furnace behavior. Change in the reductant material and amount injected to the blast furnace also impact on the other process units in an integrated steel plant. The integrated steel plant model (gate-to-gate model), which combines the most important unit processes with material and process gas flows, was used to evaluate changes in CO₂ emissions and energy flows.

The environmental and economic performance of bioreducers was analyzed with carbon footprint, energy return on investment and production cost analyses. Carbon footprint and energy return on investment assessments were conducted with the developed life cycle model (cradle-to-gate model) for three bioreducers: charcoal, torrefied wood and Bio-SNG, which were considered the most feasible injectants. CO₂ mitigation potential and mitigation cost were calculated with a reference case of a Finnish carbon steel producer. The influence of EU ETS on the fuel switch from fossil-based to renewable reductants was evaluated. The availability of energy wood for bioreducer production in Finland was investigated by reviewing recent publications and the author's own calculations, with an emphasis on competing use. In the following sections, the system boundaries, data sources and methods applied are more thoroughly described.

4.1 System boundaries

The implications of bioreducer use in the blast furnace were evaluated at separate system levels. The impact of bioreducer use on material and energy balances and CO₂ emissions was conducted at the unit process (blast furnace) and plant site levels. The simplified gate-to-gate system boundary presented in Fig. 8 was used in this study (Paper I). In this assessment, the oxygen plant was not taken into

account. Besides that, all the major process units and gas streams between unit processes were modeled. For example, blast furnace gas was assumed to be used in hot stoves to heat the blast air, in coking plant to fire up the coking batteries with coke oven gas, and in power plant to produce energy for the plant site. The gate-to-gate model does not include sintering plant, which resembles the situation in the reference plant. The main iron-bearing material is iron ore pellets, and the majority of the internal iron-bearing residues are recycled back to the blast furnace via a briquetting plant. CO₂ emissions resulting from the raw material supply or from external electricity consumption were not included in the study. Carbon released from the charcoal was considered to be carbon-neutral. The unit process models and their connections are described in detail in section 4.4.

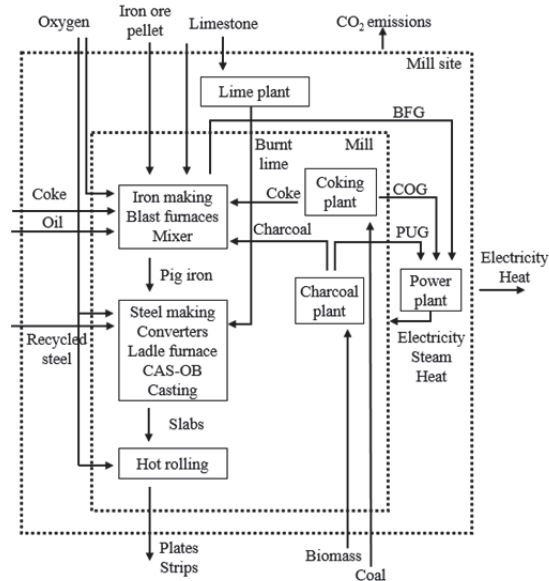


Fig. 8. Gate-to-gate system boundary (Paper I, reprinted by permission of ISIJ International).

System boundaries were further expanded in Paper IV, in which the carbon footprint, energy return on investment, and production costs of three different bioreducers produced from forest-based raw materials, torrefied wood, charcoal and Bio-SNG, was evaluated. The system boundary of the study is illustrated in Fig. 9, which represents a cradle-to-gate system definition. In addition, the CO₂

4.2 Data sources

The Ruukki steel plant, located on the northern shore of the Gulf of Bothnia, was used as a reference when evaluating the possible biomass consumption amounts, CO₂ reduction potential and economic feasibility of bioreducer use. According to Kinnunen *et al.* (2011), the annual production capacity of the two blast furnaces at the Ruukki steel plant is 2.6 million tonnes hot metal. In recent years, steel production has been 2.3 million tonnes, and hot metal production some 2.0 million tonnes (Environmental Data Monitor 2014). Recycled steel is used as a raw material in basic oxygen furnaces, which increases the steel production rate compared to the amount of hot metal.

Input data concerning the chemical properties of the raw materials in blast furnace and other unit processes in gate-to-gate system modeling (Paper I) were collected from the available literature sources and from plant data. In integrated steel plants, various raw materials are used, which has an impact on the process behavior and outputs of the processes. For example, several coal grades with varying carbon and ash contents, volatile amount and ash chemistry were used as coke plant model inputs.

In carbon footprint (CFP) and energy return on investment (EROI) calculations (Paper IV), the forest chip production chain data was collected from Finnish literature sources. Thermochemical conversion technology data for slow pyrolysis, torrefaction and gasification plus methanation was collected from the literature and created with modeling. Cost data needed in the production cost analyses of bioreducers were derived from literature sources. Finnish data was used whenever available.

Forest chip availability for steel industry in Finland was examined via analysis of recent energy wood availability assessments (Hakkila 2004, Maidell *et al.* 2008, Pöyry Energy 2007, Kärhä *et al.* 2010) and the author's own calculations based on the available distribution coefficients for logging residues and stumps (Paper III). The availability estimates were divided into theoretical, techno-ecological and techno-economic potentials. Theoretical estimates assume that the yield of logging residues and stumps is 100%. Ecological and economic restrictions are also ignored. The theoretical potential of small-diameter wood depends e.g. on the defined lower limit of the wood diameter and suitable stands. In the techno-ecological potential calculations, the yield of logging residues, stumps and thinnings is below 100% because of the ecological constraints. Also, the willingness of forest owners to offer energy wood to the market is taken into

account. Techno-economic potential estimates also take into account subsidies and energy producers' willingness to pay. Besides the forest chip production potential estimations, emphasis was placed on the assessment of possible other future users of energy wood fractions, which would weaken the availability of energy wood for bioreducer production.

4.3 Comparison of the metallurgical properties of fossil-based and renewable reductants

In this thesis, the suitability of bioreducers for blast furnace reductants were evaluated. The properties of torrefied biomass, charcoal, bio-oil, syngas and Bio-SNG were evaluated and compared to fossil-based reductants that are currently used as BF reductants. These bioreducers were selected because they can all be produced with promising thermochemical conversion technologies. The main emphasis was on the evaluation of the bioreducer properties from the viewpoint of their injection to the blast furnace.

The chemical properties of solid, liquid and gaseous bioreducers are presented in Tables 3–5 and compared to fossil-based reductants. The chemical properties and heating value of pulverized coals set the basis for comparison of the solid bioreducers. Pulverized coals (PC) have varying chemical properties depending on the coal source. Typically, the carbon content of fossil coal is 80–90%, with an ash content of around 10%. The heating value of the pulverized coals is typically around 30 MJ/kg. The chemical properties of solid bioreducers produced from wood-based feedstock depend on the final temperature of the heat treatment. Torrefied wood (TW) has a high volatile content and lower heating value. The heating value of the torrefied wood approaches the heating value of pulverized coal when the torrefaction temperature is 300 °C (TW 2). The ash content of the torrefied wood is low, but depends on the properties of the raw material. The carbon content of charcoal (CC) can be high, even higher than in pulverized coals. The fixed carbon and heating value depend on the pyrolysis temperature. (Table 3)

Table 3. Chemical properties of fossil coal, torrefied wood and charcoal.

	Proximate analysis			Ultimate analysis					Heating value (HHV) (MJ/kg)
	wt. (%)			wt. (%), daf					
	Volatile matter	Fixed C daf	Ash db	C	H	O	N	S	
PC 1 ¹	27.7	72.3	9.5	86.4	4.9	6.0	2.1	0.6	31.6
PC 2 ¹	18.5	81.5	10.3	89.3	4.3	3.5	2.0	0.5	32.0
PC 3 ²	32.5	54.5	9.8	83.5	5.3	8.6	2.0	0.6	30.1
TW 1 ³ (at 225 °C)	82.2	17.9	1.4	50.2	6.1	42.7	0.3	ND ⁶	19.8
TW 2 ³ (at 300 °C)	52.9	44.8	2.3	66.1	4.9	27.3	0.5	ND ⁶	26.4
CC 1 ⁴	32.7	58.2	4.5	82.2	4.5	3.6	0.6	0.02	33.5 ⁷
CC 2 ¹	21.8	78.2	4.6	83.7	3.1	12.0	1.1	0.1	30.0
CC 3 ⁵	9.5	88.6	1.9	91.7	2.4	3.6	0.3	<0.05	34.4 ⁷

¹ Machado *et al.* (2010b)² Shen *et al.* (2009)³ Phanpanich & Mani (2011)⁴ Ooi *et al.* (2011)⁵ MacPhee (2009)⁶ ND: Not determined⁷ Calculated from the ultimate analysis with equation provided by Channiwala & Parikh (2002)

The chemical properties and other important properties from the viewpoint of blast furnace injection of heavy fuel oil and bio-oils produced from wood and microalgae are presented in Table 4. Fossil-based oil has high carbon and hydrogen content, which results in a high heating value. Bio-oils have high oxygen content and they contain water, which is difficult to remove with current separation technologies. The heating value of bio-oil is low. Bio-oils produced from biomass-based feedstock contain a substantial amount of carboxylic acids, resulting in a low pH value of 2–3. The strong acidity results in corrosive behavior to part of the metals used as construction materials (Zhang *et al.* 2007). The sulfur content of fossil-based heavy oil can be as high as 2.0% (Slaby *et al.* 2006) whereas the sulfur content of bio-oil is very low.

Table 4. Chemical properties of heavy fuel oil and bio-oils.

	C	H	O	N	Ash	Moisture	HHV	Viscosity	Solids	pH	Specific
			(wt%)				(MJ/kg)	(cP)	(wt%)		gravity
Heavy fuel oil ¹	85	11	1	0.3	0.1	0.1	40	180	1	-	0.94
								(at 50 °C)			
Bio-oil	54-	5.5-	35-	0-0.2	0-0.2	15-30	16-19 (as	40-100	0.2-1	2.5	1.2
(wood) ¹	58	7.0	40				produced)	(at 50 °C)			
Bio-oil	62.1	8.8	19.4	9.7	ND ³	-	30	100	ND ³	ND ³	1.06
(Micro-algae 1) ²							(moisture free)	(at 40 °C)			
Bio-oil	76.2	11.6	11.2	0.9	ND ³	-	41	20	ND ³	ND ³	0.92
(Micro-algae 2) ²							(moisture free)	(at 40 °C)			

¹ Czernik & Bridgewater (2004)² Miao & Wu (2004)³ ND: Not determined

Gaseous blast furnace injectants (natural gas, hot reducing gases and coke oven gas) differ considerably from each in their chemical composition. Similar gas compositions can be produced from biomass-based feedstock with alternative gasification technologies (Table 5). Natural gas, which is mainly methane, is used as a reductant in some blast furnaces. Bio-SNG produced from biomass with gasification and methanation can have similar chemical properties to natural gas.

Table 5. Chemical properties of hot reducing gas, coke oven gas and syngas produced from biomass with different gasification technologies.

	H ₂	CO	CO ₂	CH ₄	C ₂ H ₄₋₆	N ₂	LHV
			(vol% dry basis)				(MJ/m ³)
Recycled BF top gas ¹	14.9	71.7	3.2	-	-	10.3	10.7
Coke oven gas ²	66.0	6.0	2.0	21.0	3.0	3.0	17.5
Allothermal gasification ³	40.0	25.0	21.0	10.0	2.5	1.5	14.0
Autothermal gasification ³	26.0	20.0	35.0	13.0	3.0	3.0	12.0
Entrained flow gasification ³	39.0	38.0	20.0	0.1	0	3.0	10.0

¹ Danloy *et al.* (2008)² Lundgren *et al.* (2013)³ Zhang (2010)

The ash content of liquid and gaseous reductants is low, but solid fossil-based reductants can contain significant amounts of ash (>10%). Solid bioreducers produced from wood have a low amount of ash, but bioreducers produced from e.g. agricultural residues have a high ash content. The ash chemistry of fossil-based coals and biomass-derived bioreducers differs considerably (Table 6). The pulverized coals have a high content of SiO₂ and Al₂O₃, whereas the charcoals contain elements that increase basicity (e.g. Ca and Mg). In general, fossil-based coals have lower proportion of alkalis (Na and K) than wood-based coals. The proportion of phosphorus (P) is somewhat similar.

Table 6. Chemical analyses of the ashes of pulverized coals and charcoals.

Ash analysis (db)	PC 1 ¹	PC 2 ¹	CC 1 ²	CC 2 ²	CC 3 ²
SiO ₂	46.00	50.14	28.46	3.82	21.47
Al ₂ O ₃	25.19	26.73	3.96	1.17	4.84
Fe ₂ O ₃	14.61	9.03	1.95	1.14	4.40
TiO ₂	1.24	1.33	0.23	0.15	0.28
P ₂ O ₅	1.64	1.55	1.29	4.04	2.58
CaO	4.12	3.91	39.46	53.04	38.45
MnO	0.14	0.09	0.62	ND ³	ND ³
MgO	1.69	1.53	4.32	11.45	4.05
SO _x	1.20	0.78	1.91	4.67	2.62
Na ₂ O	0.21	0.41	0.12	0.47	1.62
K ₂ O	1.22	1.04	2.40	4.32	9.28

¹ Machado *et al.* (2010b)

² MacPhee (2009)

³ ND: Not determined

4.4 Blast furnace and integrated steel plant modeling

The plant-wide impact of introducing biomass in solid form (i.e. as charcoal to blast furnace as a tuyère injectant) was investigated with mathematical modeling and simulation (Paper I). Special emphasis was placed in the study on the development of a blast furnace model that can be used to evaluate the effect of different reducing agents on furnace behavior. The blast furnace model is based on a conceptual division of the furnace into two active zones that interact via a thermal reserve zone. The model is based on mass and energy balances calculated separately for both segments of the furnace. This kind of blast furnace modeling

has been reported by Peacey & Davenport (1979), Rasul *et al.* (2007), and Hooley *et al.* (2010). The model assumes that all the higher oxides in the upper zone are reduced to wüstite when entering the thermal reserve zone. The conceptual division and thermal reserve zone are based on the assumption that, above the thermal reserve zone in temperatures below 1,200 K, the CO₂ produced by the wüstite reduction reaction is no longer reconverted to CO by the Boudouard reaction.

In the calculation procedure, the input parameters of the blast furnace model were kept as unchanged as possible. The adiabatic flame temperature was kept as a control parameter and was maintained in the feasible region with increased blast temperature and oxygen enrichment. The blast air volume was calculated after this. The amount of fluxes (limestone and quartzite) was tuned to reach a slag basicity of 1.08. Other major unit processes modeled in the work include coke ovens, limestone calcination, biomass pyrolysis, basic oxygen furnace, CAS-OB, continuous casting, slab furnace, hot rolling and power plant.

In coke production, coal is dry distilled at high temperatures (900–1,100 °C). The heat required for the coking process is taken from the burning of the coke oven and blast furnace gases. The yield of the coke in the model is assumed to depend on the amount of volatiles in the coal mix. It is assumed that all the ash goes into the coke and sulfur is taken into account with a yield factor. The proportion of carbon in coke is assumed to depend on the ash and other elements in the coke. The heating value of COG is assumed to be 16.8 MJ/m³. The amount of gases; COG and BFG needed to provide sufficient heat for the coking process is calculated with the procedure described by Ertem & Özdabak (2005).

Limestone calcination is conducted in shaft kilns where heat is used to decompose limestone (mainly CaCO₃) into burnt lime (CaO) and CO₂. Limestone is used in the blast furnace and burnt lime in the basic oxygen furnace. Thermal decomposition is endothermic, needing heat to proceed. This heat is obtained by burning coke oven gas in the kiln. Emissions into the air are released from the decomposition of the limestone and from the burning of the fuels.

The biomass pyrolysis process, which is hypothetical in nature, is modeled based on mass and heat balances. The heat requirement of the pyrolysis process is satisfied with the burning of the pyrolysis unit by-products. Some external energy is needed to start the pyrolysis process, but it is considered negligible.

The basic oxygen furnace is a process where hot metal and scrap are refined into steel by oxygen blowing with argon stirring. The BOF model is based on the mass and energy balance calculated for the whole furnace. The oxygen demand is

calculated from the carbon balance resulting from the predefined carbon content in crude steel. The need for additives (burnt lime and dolomite) is calculated based on the relations developed by Turkdogan (1996) for targeted slag basicity and silicon content. BOF gases (BOFG) are used for district heat production, thus no BOFG goes to the power plant. Secondary steelmaking processes are considered by giving them certain yield factors. Steel losses come from splashes, dust, etc. The amount of energy needed in the physical steelmaking processes is taken from industrial practice.

The blast furnace is the key process in this assessment because it determines the inputs and outputs of the majority of the other process units. The unit processes described above were linked together into a plant site model (Fig. 8), which was used in the calculation of gate-to-gate life cycle inventory. The selected functional unit in the plant-wide simulation was 1 tonne of hot-rolled plate. Factory simulation tool software was used in the modeling and simulation tasks. The thermodynamic data used in the calculations was taken from HSC Chemistry® (Roine *et al.* 2007), which is linked to the Factory simulation tool.

Three scenarios for blast furnace simulations were selected. The base case scenario, with oil injection, describes the normal practice in the reference plant. The second scenario (CC case 1) under investigation was the full replacement of oil injection with charcoal injection. In the third scenario (CC case 2) the charcoal injection was increased to 150 kg/t hot metal, which is the normal injection amount for pulverized coal (Larsson 2004). The blast furnace model was used to calculate e.g. coke rate, slag rate, top gas composition and volume for these scenarios. The same injection amounts were used in the gate-to-gate simulation; however, in the results section they were calculated per tonne of hot-rolled plate produced. In the CC case 1 scenario, charcoal was assumed to be produced outside the system boundaries, and in the CC case 2 scenario, charcoal was produced inside the system boundaries and by-product gases from biomass pyrolysis were utilized in the power plant to produce electricity.

4.5 Carbon footprint assessment

Carbon footprint methodology was used to evaluate the CO₂ emissions resulting from bioreducer production. Carbon footprint has gained a lot of attention over the last few years. It is a surprisingly new concept, even though greenhouse gas emission studies have been conducted for numerous years. The newly provided definition for CFP is (Wiedmann & Minx 2008): “The carbon footprint is a

measure of the exclusive total amount of carbon dioxide emissions that is directly and indirectly caused by an activity or is accumulated over the life stages of a product.”

In the CFP, only direct and indirect CO₂ emissions are included in the analysis, whereas in life cycle assessment other emissions and environmental burden are also covered. Carbon footprint is calculated for the product (either goods or services), and the whole life cycle is included (Wiedmann & Minx 2008).

In recent years, several carbon footprint methodologies have been developed. The differences between ISO/TS 14067, the GHG Protocol Product Standard, PAS 2050 and Climate Declaration methodologies are discussed in the publication of Garcia & Freire (2014). The major differences in methodologies are the treatment of biogenic CO₂, multifunctionality and unit process exclusions. In this thesis, a bottom-up (process analysis) approach was used to calculate the CFP for bioreducers. The methodology applied in this thesis was, for the most part, based on the ISO 14040 and ISO 14044 standards, which were available at the time of the study. For the most part, ISO/TS 14067 follows the guidelines of ISO 14040 and ISO 14044. Indirect emissions in this work follow the definition of carbon stock change provided by Repo *et al.* (2011). The CFP is defined as CO₂-equivalent, which means that CH₄ and N₂O emissions are transformed into CO₂ equivalents by using defined coefficients. The definition of CFP used in this study differs slightly from the definition of Wiedmann & Minx (2008). The system boundary of the CFP calculation is illustrated in Fig. 9.

4.6 Energy return on investment

The energy return on investment indicator was used to evaluate the energetic performance of different bioreducers on a life cycle basis (Raugei *et al.* 2012). The higher the EROI, the more efficiently the energy carrier is produced (Murphy & Hall 2010). The EROI was defined as the ratio of energy embodied in the products (bioreducer and energetic by-products) to the energy embodied in the fossil-based fuels utilized to produce it (equation 1).

$$EROI = \frac{E_{bioreducer \text{ and } by-products}}{E_{in, non-renewable}} \quad (1)$$

The definition of the EROI does not consider the energy inputs of machinery and production plant construction, which are to be considered in full EROI calculation (Murphy & Hall 2010). In this work, the primary energy consumption of each process stage was calculated according to fuel consumption and was aggregated as a single performance indicator.

4.7 Economic analyses

Economic evaluation of the bioreducer feasibility for blast furnace reductant was conducted in Papers III–IV. In Paper III, production cost analysis was performed for charcoal production in Finland. The raw material for charcoal production was logging residues, which were collected, chipped and transported to the pyrolysis facility. Cost data was taken from recent Finnish literature sources. The cost of the carbonization stage was taken from the literature and two estimates were used, resulting in two production cost estimates.

More in-depth production cost analyses were made in Paper IV for three bioreducers: torrefied wood, charcoal and Bio-SNG. The raw materials in this assessment were logging residues, small-diameter wood, and stumps, and the cost data was based on Finnish sources. The capacity of the bioreducer production plant was assumed to be 50 MW dry biomass input. Annualized investment and operating costs were derived from the developed process models. The cost data used in the plant investments was taken from the literature. The impact of bioreducer production plant size on production costs was also evaluated. Additionally, the production cost of metallurgical coke was calculated based on the cost data presented in the literature to allow comparison between bioreducers and coke.

After calculating the CO₂ emission and production costs of the bioreducers, the CO₂ reduction potential achieved with each of the bioreducers was evaluated, using a Finnish steel producer as a reference. In Paper III, the economics of wood-based charcoal production was compared to fossil-based reducing agents; metallurgical coke, pulverized coal and oil by taking into account the effect of the European Union Emissions Trading System (EU ETS). Economic analysis in Paper IV included a feasibility assessment of bioreducers compared to metallurgical coke. Finally, CO₂ mitigation costs when replacing part of the metallurgical coke in the blast furnace with bioreducers were evaluated.

5 Results and discussion

5.1 Assessment of the metallurgical properties of bioreducers

Wood-based biomass has more suitable chemical properties for producing solid bioreducers with torrefaction and slow pyrolysis than herbaceous biomass and agricultural residue. The carbon content and heating value of torrefied and pyrolyzed biomass depend mainly on the final temperature of the heat treatment. The major differences between biomasses are in ash chemistry. The proportion of ash in wood-based biomasses is usually low, which is beneficial for the BF process. Because of the low ash amount, the proportion of unwanted elements such as phosphorus (P), potassium (K), and sodium (Na) also remains at acceptable level. In the case of herbaceous biomasses, the amount of ash is considerably higher (Paper II).

Ash chemistry also has an impact on slag formation in the blast furnace. There must be a balance between acid and basic slag components to ensure e.g. suitable slag viscosity and the ability of slag to pick up unwanted elements from hot metal. The introduction of basic solid bioreducers into the BF might result in a lower need for basic additives (limestone). The amount of slag in the BF would decrease, which means a productivity increase in the amount of hot metal.

Charcoal produced at high temperatures has better properties for BF injection than torrefied wood, which still has a high proportion of oxygen and volatile matter and a lower heating value. However, torrefied wood could be part of the reductant portfolio and used together with pulverized coal injection.

The properties of bio-oil are not feasible from the BF perspective. The heating value of bio-oil is too low—half of the heating value of heavy distillation residue oil used in some blast furnaces. The high proportion of oxygen and water in bio-oil lowers the heating value and results in a low coke replacement ratio and possible problems with flame temperature. Bio-oil is corrosive, and its viscosity differs from the oil used as an injectant.

From the viewpoint of chemical composition, gaseous bioreducers would be suitable for BF injection. The properties of syngas depend on the gasification technology and gasification agent applied. However, the proportion of CO and H₂ in syngas suitable for injection should be considerably high, which necessitates extensive gas conditioning. The chemical properties of Bio-SNG are comparable to natural gas, which is used as an injectant in blast furnaces.

5.2 The impact of bioreducer use on BF behavior

Bioreducers have different effects on blast furnace behavior. The coke replacement ratio is one indicator that can be used to evaluate the efficiency of alternative reductants. The lowest replacement ratio is with bio-oil, around 0.25. The coke replacement ratio of torrefied wood is better, around 0.4, but still significantly lower than that of fossil-based reductants. Good quality charcoal has a high coke replacement ratio (0.8–1.1), which is even higher than fossil-based pulverized coals. The coke replacement ratio of Bio-SNG is similar to natural gas, which ranges from 0.9 to 1.15. The coke replacement ratio of syngas is more difficult to evaluate, since gas could be injected at different temperatures. (Papers II and IV) Even though Bio-SNG has the highest coke replacement ratio, injection of charcoal would reduce coke consumption the most (Paper II). The effect of charcoal injection on blast furnace behavior was more thoroughly examined with the developed blast furnace model.

The blast furnace process performance was examined with three distinct scenarios where the amount of tuyère injection was changed. In the base case scenario, oil was used as a tuyère-injected reducing agent. In the second scenario (CC case 1), charcoal, produced outside the system boundaries, was used to replace oil injection completely. In the third scenario (CC case 2), the amount of charcoal injection was further increased, and it was also assumed that charcoal is produced inside the system boundaries and that by-product gases from biomass pyrolysis are utilized in the power plant to produce electricity.

Injected reducing agents have varying coke replacement ratios in the BF. The coke replacement ratio of oil and charcoal were calculated using the assumptions made in the development of the blast furnace process model. The simulation results suggested that the replacement ratio of charcoal against coke is 0.97. Correspondingly, the replacement ratio of oil against coke is around 1.12. This means that to replace specific heavy distillation residue oil injection, the amount of charcoal must be 1.15 times higher. The amount of specific heavy distillation residue oil used in the base case scenario to produce one tonne hot metal was 90 kg/t. This means that to replace this amount, using the replacement ratio, 103.5 kg charcoal is needed to fully replace oil injection (CC case 1). The third case (CC case 2) describes a case where the amount of charcoal injection has been further increased to 150 kg/t hot metal.

The results of blast furnace process simulation revealed that the biggest influence of charcoal between the base case and CC case 1 relate to the change of

blast volume, which decreases from 944 to 922 Nm³/t hot metal. The blast volume further decreased in CC case 2, but this change was also affected by the elevated blast temperature and permitted drop in the flame temperature. The amount of slag decreased slightly from 190 kg/t hot metal in the base case to 188 kg/t hot metal and 186 kg/t hot metal in CC case 2. The amount of gases produced in the process also decreased quite a lot. The combined effect of decreasing the volume of slag and gases in the furnace mean that the productivity of the furnace increases. The heating value of the BFG decreased slightly when changing from oil injection to charcoal injection. However, according to the calculations, the heating value of the BFG is enough to raise the blast air temperature to the needed regime.

The amount of reducing agents in the base case scenario was 469.0 kg/tHM, 481.5 kg/tHM in CC case 1, and 475 kg/tHM in CC case 2. It can be deduced that the amount of reducing agents needed to produce one tonne of hot metal slightly increased when oil was replaced with charcoal. However, the amount of fossil carbon released into the atmosphere decreased significantly.

5.3 The impact of bio reducer use on integrated steel plant CO₂ emissions

In Paper I, gate-to-gate CO₂ emissions were calculated with the simulation model and compared to available data in the literature (Birat 2009, Iosif *et al.* 2010). In the work of Iosif *et al.* (2010), CO₂ emissions from an actual integrated steel plant, from the modeling scenario and data from the World Steel Association (previously IISI) were provided. The functional unit in these assessments was 1 tonne of hot rolled coil (Table 7). Further evaluation of CO₂ emissions by process compared to the literature is presented in Fig. 10.

Table 7. CO₂ emissions (kg/functional unit) calculated with the model compared to the literature (Paper I, reprinted by permission of ISIJ International).

	Total CO ₂ emissions [kg/FU]
Present work	1780
Plant data (Iosif <i>et al.</i> 2010)	1949
Model data (Iosif <i>et al.</i> 2010)	1740
IISI (Iosif <i>et al.</i> 2010)	2007
Plant data (Birat 2009)	1815

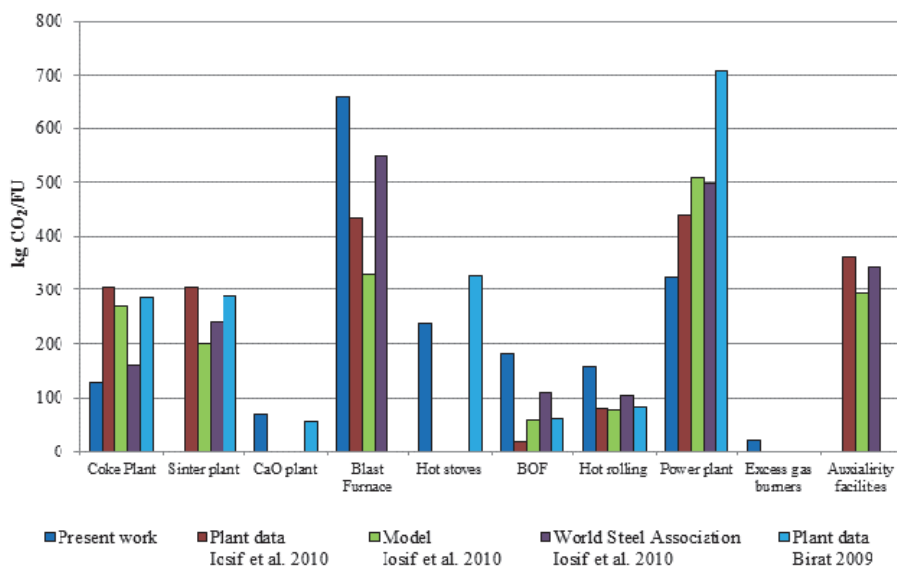


Fig. 10. CO₂ emissions of the simulated gate-to-gate system processes compared to literature (Paper I, reprinted by permission of ISIJ International).

The allocation of CO₂ emissions between processes was resolved by considering the final conversion to CO₂ as a process-specific emission. For example, only the amount of CO₂ generated in the BF from carbon-based reductants or CO conversion to CO₂ in the power plant resulting from BFG burning was reported in the specific process unit CO₂ emissions. The largest amount of CO₂ emissions, 660 kgCO₂/FU in the base case, was emitted from the BF. The heating of blast air in the hot stoves with BFG produced 237 kgCO₂/FU. The power plant, which uses BFG and some COG, produced 324 kgCO₂/FU. The total amount of CO₂ emissions were quite similar (Table 7), but the process-specific CO₂ emissions differed considerably. Differences in the process-specific CO₂ emissions are probably due to different fuel mixes, differences in the process chain and the allocation procedure.

In CC case 1, charcoal was produced outside the system boundaries and the only changing parameter was the full oil replacement with charcoal. In this scenario the plant-wide changes were modest. The decreased amount and calorific value of the BFG would change the energy balance of the integrated steel plant. This could result in a reduction of electricity output from the power plant from

177.6 to 152.6 kWh/t FU if all the other processes used the same amount of BFG as in the base case.

In CC case 2, a pyrolysis plant was assumed to be located inside the system boundaries. The simulation results of the pyrolysis plant suggested that the utilizable energy content of by-product syngas was 24.9 MJ/kg charcoal, of which 20% was used to fulfill the pyrolysis process energy requirement. The amount of utilizable energy used in the power plant was therefore 19.9 MJ/kg charcoal. CO₂ emissions produced in the burning of biomass-based by-products were considered to be CO₂-neutral and were not included in the carbon dioxide balance.

The total CO₂ emissions of the integrated steel plant for the three scenarios studied are presented in Fig. 11. “CO₂-neutral” in the chart means that the charcoal-based emissions are considered carbon-neutral. The CO₂ emissions resulting from the pyrolysis process and from by-product burning in the power plant were also considered carbon-neutral.

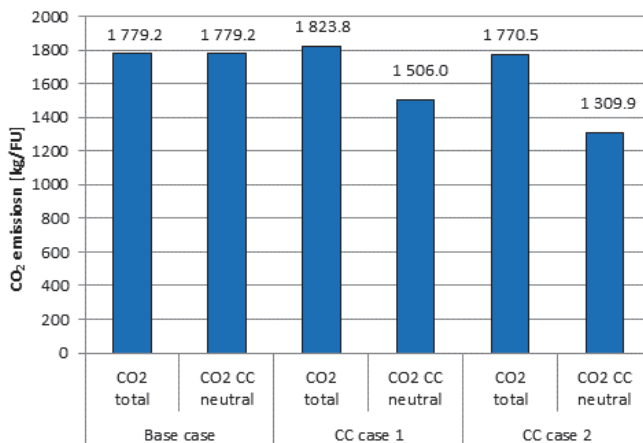


Fig. 11. Total and fossil CO₂ emissions of examined scenarios (Paper I, reprinted by permission of ISIJ International).

In CC case 2, fossil CO₂ emissions decreased basically in two ways. The fossil carbon input to the blast furnace decreased when the oil and some of the charged coke were replaced. The reduction in the amount of coke also decreased the emissions from the coking process. The total reduction in fossil CO₂ emissions between base case and CC case 1 was 273 kgCO₂/FU, a 15.4% decrease. The

reduction between base case and CC case 2 was 469 kgCO₂/FU, equal to a 26.4% decrease in fossil CO₂ emissions.

If the calculated values are compared to the production volumes of an existing steel plant in Finland, with 2.6 million tonnes hot rolled plate output, the fossil CO₂ emissions with gate-to-gate system boundaries would decrease from 4.63 million tonnes to 3.92 million tonnes between the base case and CC case 1. This drop would be achieved with minor changes to the infrastructure of the steel plant. Between the base case and CC case 2 the reduction would be from 4.63 million tonnes to 3.41 million tonnes. However, this scenario would entail major investments in existing plant structure. Difficulties might also rise from the fact that a huge amount of biomass would be needed in a single location to produce the charcoal.

5.4 Carbon footprint of bioreducers

The carbon dioxide emission reduction potential was investigated at two system levels. The CO₂ reduction potential of replacing oil injection with charcoal injection was investigated with the developed steel plant model. In this assessment, only direct emissions were taken into account (Paper I). The Finnish integrated steel plant was used as an example to illustrate the annual CO₂ reduction potential. System boundaries were further expanded to take into account the CO₂ emissions from the production of bioreducers (Paper IV). The carbon footprint was calculated for torrefied wood, charcoal and Bio-SNG produced from Finnish energy wood.

The carbon footprint of torrefied wood, charcoal and Bio-SNG were calculated with the developed life cycle model. Charcoal and Bio-SNG production yield by-products (pyrolysis gas and excess heat) that can be utilized in different applications. It was assumed that by-products are utilized to produce electricity with 30% efficiency.

An overview of the CO₂ emissions (gCO₂/MJ per product) is presented in Fig. 12, along with the life cycle CO₂ emissions of natural gas (75 gCO₂/MJ) and coal (115 gCO₂/MJ) (Burnham *et al.* 2012). Bioreducers were produced from different raw materials: “SDW 50%” refers to small-diameter wood with 50% moisture, “SDW 30%” to small-diameter wood with 30% moisture, “LR” stands for logging residues and “ST” for stumps. The moisture of LR and ST were assumed to be 50% and 30%, respectively, upon arrival at the bioreducer production plant. The carbon footprint of bioreducers was evaluated with different

assumptions concerning the life cycle stages taken into account. The purpose of Fig. 12 is to show how the CFP would change with different assumptions. In the base case results (the blue bars), direct emissions from the forest chip supply chain and life cycle emissions from fossil fuel use were considered. Emissions from fertilizer production increase the CFP (the red bars) to some extent. Incorporation of by-product credits lowered the CFP of charcoal and Bio-SNG (the green and violet bars). The CFP becomes considerably high if indirect carbon stock changes are taken into account, even with by-product credits (the light blue and orange bars).

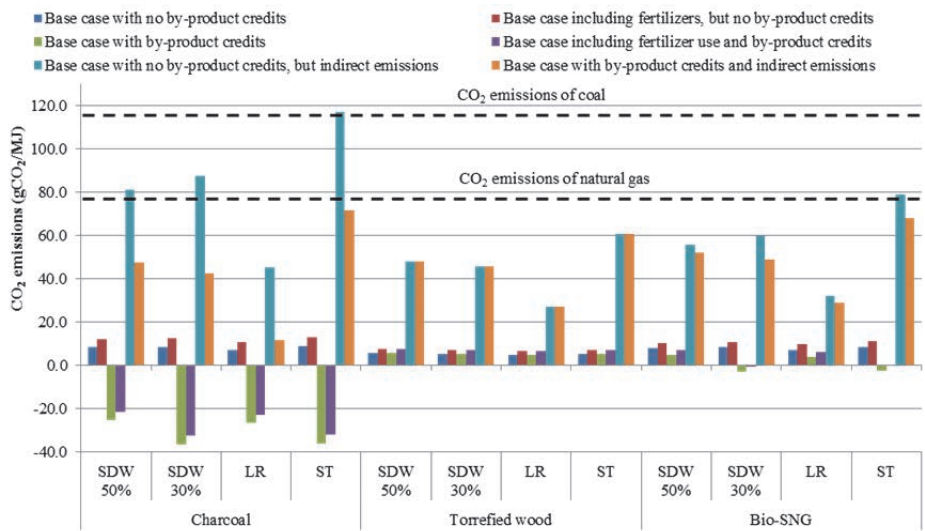


Fig. 12. CO₂ emissions of bioreducer production depending on system boundaries (Paper IV, reprinted by permission of Elsevier).

The CFPs of bioreducers in the base case without co-product credits, fertilizer use or indirect carbon stock change were: 7.2–9.0 gCO₂/MJ, 4.9–5.6 gCO₂/MJ and 7.4–8.5 gCO₂/MJ for charcoal, torrefied wood and Bio-SNG, respectively. The results indicate that CO₂ emissions from pre-treatment and transportation, on average, contributed the major proportion in charcoal and torrefied wood production, about 25–49% of total CO₂ emissions. In Bio-SNG production, CO₂ emissions from electricity use were dominant, at 62% of the total CO₂ emissions. With fertilization use incorporated in the CFP figures, the emissions were

10.8–12.9 gCO₂/MJ for charcoal, 6.7–7.5 gCO₂/MJ for torrefied wood, and 9.7–11.1 gCO₂/MJ for Bio-SNG.

Indirect emissions from carbon stock change increased emissions significantly. If the indirect carbon stock change was taken into account, but no by-product credits, the CO₂ emissions increased to 45–117 gCO₂/MJ, 27–61.0 gCO₂/MJ and 32–79 gCO₂/MJ for charcoal, torrefied wood and Bio-SNG, respectively. If the by-product credits were taken into account with the assumption that electricity is produced from by-products (fertilizer use and carbon stock change ignored), a net credit will be gained with charcoal and Bio-SNG. The net impact of charcoal production would be from –36.3 to –25.0 gCO₂/MJ, and the net impact of Bio-SNG production from –2.7 to 4.7 gCO₂/MJ.

The carbon footprint of bioreducers differed to some extent from each other when evaluated against produced units of energy. The difference was larger when the CFP was calculated per tonne of bioreducer produced. In the base case, without co-product credits, fertilizer use or indirect carbon stock change, the carbon footprint was 105.9–122.3 kgCO₂/t, 214.1–266.7 kgCO₂/t and 368.4–425.7 kg/t for torrefied wood, charcoal and Bio-SNG, respectively. Major factors contributing to differences in CFP between bioreducers were the differing raw material needs, product yield and energy consumption of bioreducer production. Moisture of the feedstock material (here energy wood) also plays a role if by-product credits from bioreducer production are considered. If the moisture content of the raw material is 30% instead of 50%, less energy is needed in the drying process, which can be used to produce electricity. However, drying the wood at the roadside results in dry-matter losses, which takes away some of the advantage of lower-moisture energy wood.

System boundary definitions and allocation of CO₂ emissions play an important role when assessing the carbon footprint of biomass-based energy carriers. In this work, the anticipated carbon stock change with a 100-year time horizon was also evaluated. The carbon footprint of bioreducers would become almost comparable to natural gas or coal if carbon stock change is considered. However, in current practice, carbon stock change is not taken into account when considering wood-based raw materials. In the future, more emphasis might be placed on the sustainability criteria of bioenergy, which would also have an impact on the carbon footprint of bioreducers.

5.5 Primary energy consumption of bioreducer production

The production of bioreducers from energy wood was found to be energetically feasible. The EROI indicator was well above one for all bioreducers produced from different wood-based raw materials. The EROI values calculated for the primary product, i.e. for the bioreducer, were 3.2–3.9, 5.4–6.2 and 3.1–3.5 for charcoal, torrefied wood and Bio-SNG, respectively (Fig. 13). The EROI can also be calculated for all the products that can be further utilized, e.g. in energy production. The torrefaction process does not yield any by-products as they are utilized inside the system. The pyrolysis process yields pyrolysis gas, which can be utilized in several ways. In the EROI calculations, the energetic value is considered. The EROI of charcoal would be 5.7–6.5 if by-products are taken into account. This is higher than the EROI of torrefied wood. Bio-SNG production yields exothermic heat, which can be used for power production, hence contributing to a higher EROI of 3.4–3.9.

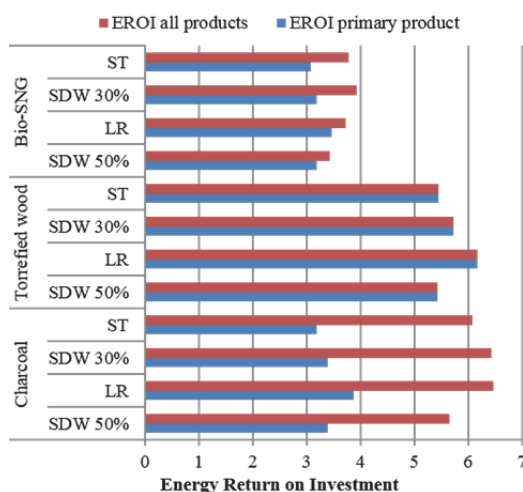


Fig. 13. Energy return on investment of charcoal, torrefied wood and Bio-SNG (Paper IV, reprinted by permission of Elsevier).

The differences in EROI resulted from varying energy consumptions during the energy wood handling and transportation. Bioreducers produced from the logging residues had the highest EROI, because there are fewer processing steps in the production chain. Torrefied wood had the best performance measured by EROI.

This is because the energy yield of torrefaction is high and the energy needed in the production stage is low.

5.6 Production costs of bioreducers

Simplified production cost analysis was conducted in Paper III for charcoal. The result of that analysis is presented in Fig. 14 for two charcoal production stage cost estimates derived from the literature. The columns represent the charcoal production costs of each stage of the production and solid lines (blue and green) represent the accumulated costs per tonne of charcoal produced. Charcoal production costs ranged from €268/t to €478/t, produced from logging residues. According to this analysis, the cost of the raw material to produce one tonne of charcoal would be €188/t, which is 70% of the lower charcoal production cost estimate and 39% of the higher production cost estimate.

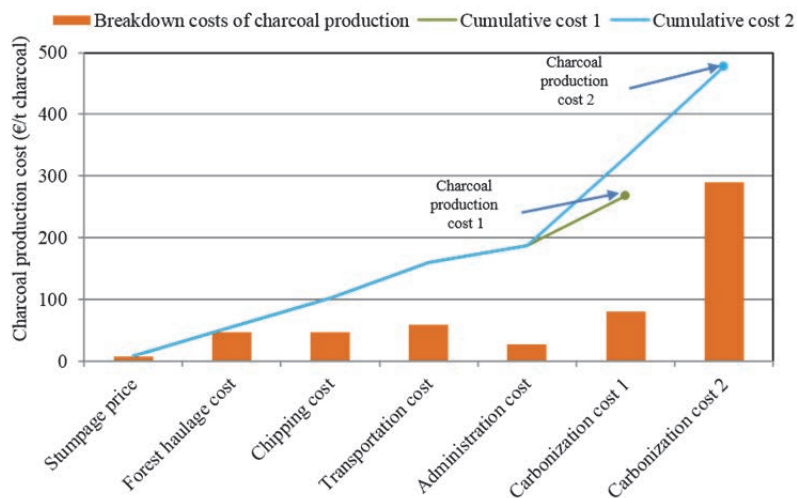


Fig. 14. Charcoal production costs in Finland (modified from Paper III, reprinted by permission of MPDI).

More thorough production cost analysis was conducted in Paper IV. In this research, more emphasis was placed on clarifying the production stage contribution to economics of the bioreducers. The total capital investment (TCI) costs derived from the plant sizing (50 MW dry biomass input) and literature sources were €₂₀₁₁12.6 million, €₂₀₁₁9.7 million and €₂₀₁₁38.0 million for charcoal,

torrefied wood and Bio-SNG production plants, respectively. Plant investment costs differ significantly from each other. Torrefaction and pyrolysis technologies are simpler than gasification and methanation technologies, which explains the difference in investment costs.

The production costs of three bioreducers, produced from logging residues (LR), small-diameter-wood (SDW), and stumps (ST), are presented in Fig. 15. Production costs were broken down to annualized capital investment cost, raw material, and other operating costs. The lowest production costs were with torrefied wood, €140–180/t. The production costs of charcoal ranged from €360/t to €490/t, depending on the feedstock. Bio-SNG had the highest production costs, €690–830/t.

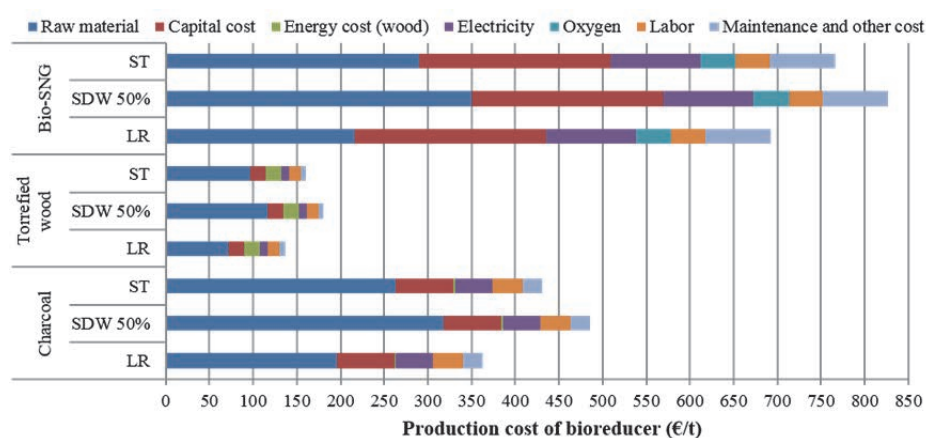


Fig. 15. Production costs of bioreducers (€/t) with base case plant size (Paper IV, reprinted by permission of Elsevier).

Raw material costs dominated in charcoal and torrefied wood production. The proportion of raw material costs in the total production cost of charcoal ranged from 54.0% to 65.4%, and from 52.8% to 64.4% in torrefied wood production. The proportion of raw material costs in Bio-SNG production was 31.3%–42.4%, and the proportion of capital costs was 26.5–31.6%. The proportion of transportation costs in total raw material costs is significant, ranging from 21% to 34% depending on the wood fraction. This has to be taken into account when considering the optimal size of bioreducer production plants. The production costs of bioreducers with different plant sizes are presented in Fig. 16. Depending on the cost structure of the thermochemical conversion plants, optimal bioreducer

production cost is attained with varying plant capacities. Investment costs in torrefied wood and charcoal production plants were considerably lower than with Bio-SNG production plants, which means that their production costs are more sensitive to raw material costs. Assumptions made in the economy of scale evaluation reveal that the lowest charcoal production costs would be attained when the plant capacity is 160 MW dry biomass input. However, increasing the capacity of charcoal plant above 100 MW (21.5 tDM/h) biomass input does not bring any considerable additional value to charcoal production costs. The investment costs of torrefaction plants were lower than charcoal production plants, and the optimal size of the plant is around 100 MW (21.5 tDM/h) biomass input. The production costs, however, do not increase significantly with larger capacities. This is because the supply area of energy wood was assumed to be a circle and the average transportation distance to increase only moderately. Due to higher investment costs of Bio-SNG plants, larger plant sizes are favored. The lowest unit cost is achieved with 515 MW (104.0 tDM/h) biomass input.

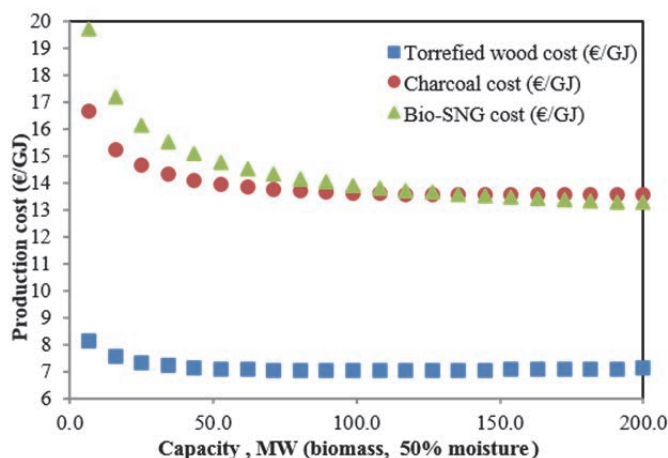


Fig. 16. Bioreducer production costs in relation to plant size (Paper IV, reprinted by permission of Elsevier).

By-product credits may play an important role in making bioreducers more feasible from an economic point of view. Depending on the valuation of the excess pyrolysis gas from charcoal production, which can be used e.g. in power production, selling pyrolysis gas at €20/MWh can contribute to up to 35% of annual operating costs. The production costs of charcoal with pyrolysis gas

credits subtracted are €250/t, €280/t and €380/t for LR, ST and SDW, respectively.

5.7 CO₂ reduction potential and mitigation costs of bioreducers in Finnish BF ironmaking

The economic feasibility and CO₂ reduction potential of using bioreducers instead of fossil-based reducing agents was further investigated in Paper III and Paper IV. Three fossil-based reductant replacement scenarios were evaluated in Paper III. In charcoal scenario 1, the competitiveness of charcoal against coke was investigated. In charcoal scenario 2, part of the injected oil was replaced with charcoal. In charcoal scenario 3, pulverized coal was replaced with charcoal injection. In the calculation, the CO₂ allowance price was also taken into account (Fig. 17). The detailed assumptions for the economic calculation can be found in Paper III. The charcoal price in a large-scale production plant was estimated to be around €400/t in Finland, assuming that revenues from the slow pyrolysis by-products would be fairly equivalent to the profit demand of the charcoal producer. In that case, the break-even carbon dioxide allowance price would be €16/tCO₂, €31/tCO₂ and €47/tCO₂ for the coke, oil and pulverized coal replacement scenarios, respectively.

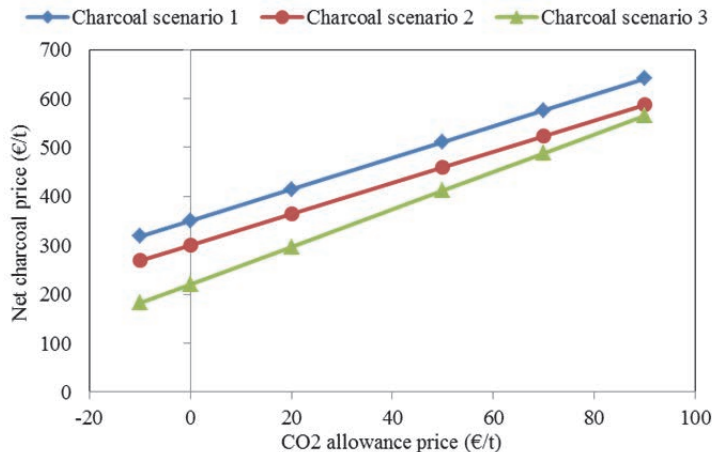


Fig. 17. Effect of CO₂ allowance price on the competitiveness of charcoal as reducing agent (Paper III, reprinted by permission of MPDI).

The fossil fuel replacement alternative closest to realization from the economic point of view would be partial coke replacement with lump charcoal charged from

the top of the blast furnace. It was pointed out in section 3.1 that only a small proportion of top-charged coke could be replaced with lump charcoal. In the case of Ruukki, with 2.0 million tonnes hot metal production, the annual direct CO₂ reduction potential would be 0.13 million tonnes compared to base case 1, which presents the current practice with coke and oil used as reducing agents. Partial replacement of oil would decrease direct CO₂ emissions by 0.09 million tonnes compared to base case 1. Base case 2, where solid pulverized coal was injected instead of oil, offered larger CO₂ reduction potential. In base case 2 the total direct CO₂ emissions in the blast furnace would increase compared to base case 1 because of the lower coke replacement ratio of pulverized coal than oil. The annual direct CO₂ reduction potential in charcoal scenario 3 compared to base case 1 was 0.99 million tonnes, and 1.15 million tonnes compared to base case 2.

The evaluation presented in Paper III considered only the direct CO₂ emission reduction potential. More in-depth analysis of the impact of bioreducer use on fossil CO₂ reduction and CO₂ mitigation costs in BF ironmaking was conducted by comparing the life cycle emissions and production costs of bioreducers and metallurgical coke (Paper IV). Again, production figures from Finnish steel producer Ruukki were used as a reference. In this assessment, the hot metal production rate was assumed to be 2.3 million tonnes annually, with coke consumption of 850,000 tonnes.

In the reference case, the CO₂ emissions from coke production were 573,000 tonnes annually, without taking into account the final conversion of coke carbon into carbon dioxide. Eventually, almost all the carbon is released into the atmosphere during the iron and steel production processes, which totals 2.7 million tonnes CO₂ emissions annually. The total CO₂ emissions from coke production and use stages add up to around 3.3 million tonnes annually.

Fossil-based CO₂ emissions could be reduced significantly by replacing metallurgical coke with bioreducers. CO₂ reduction can be facilitated in two ways. Firstly, CO₂ emissions are reduced in the reducing agent production stage, because production of bioreducers emits less CO₂ than production of coke. Secondly, CO₂ emissions from final conversion of carbon in reducing agents into carbon dioxide are lower, because carbon in the bioreducers is considered carbon-neutral. Annual coke consumption could be reduced from 851,000 tonnes to 575,000 tonnes, 740,000 tonnes, and 610,000 tonnes with charcoal, torrefied wood and Bio-SNG injection, respectively.

The emission reduction potential can be assessed with or without by-product credits. By-product credits come from utilizing by-products from bioreducer

production in power production. The highest global fossil CO₂ emission reduction was achieved with charcoal use. The CO₂ reduction potential without by-product credits was 0.99–1.01 million tonnes, and 1.29–1.39 million tonnes with by-product credits. Torrefaction does not yield by-products and the CO₂ reduction potential of torrefied wood was lower, 0.40 million tonnes per year. This is because the coke replacement ratio of torrefied wood is lower than with charcoal. Bio-SNG use decreased CO₂ emissions by 0.84–0.86 million tonnes annually without, and 0.89–0.97 million tonnes with by-product credits.

The CO₂ emission mitigation costs of different bioreducers were calculated by comparing the increase in reducing agent production costs with the CO₂ emission mitigation potential. The production cost of metallurgical coke was derived from the literature sources. Again, CO₂ mitigation costs were calculated with and without by-product credits. CO₂ mitigation costs without by-product credits were €33–69/tCO₂, €22–53/tCO₂, and €112–150/tCO₂ for charcoal, torrefied wood, and Bio-SNG, respectively. With by-product credits, the CO₂ mitigation costs were €26–54/tCO₂, €22–53/tCO₂, and €107–143/tCO₂ for charcoal, torrefied wood, and Bio-SNG, respectively. The global CO₂ reduction potential and mitigation costs of bioreducers are compared in Fig. 18.

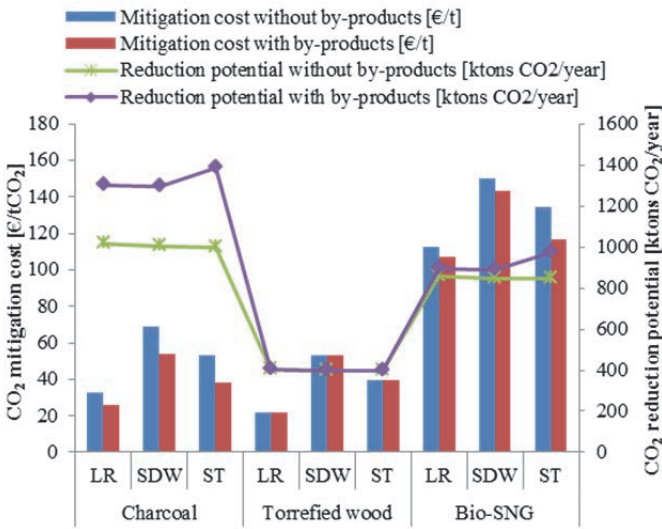


Fig. 18. Global CO₂ reduction potential and mitigation cost (Paper IV, reprinted by permission of Elsevier).

The results of the study (Paper IV) confirmed the findings in Paper III that indicated the low economic competitiveness of bioreducers compared to fossil-based reducing agents. Similar conclusions have been drawn by e.g. Norgate & Langberg (2009). High carbon taxes or other incentives would be needed to make bioreducers an economically competitive option in iron and steelmaking. However, comparison with innovative technologies, proposed e.g. in the context of the ULCOS program, shows that the economic competitiveness of switching fuels from fossil to renewable might be a viable option in the future. Tsupari *et al.* (2013) have estimated that the cost of emissions avoided globally with CCS in steel plants could be even above €100/tCO₂, which is higher than mitigation cost of using charcoal and torrefied wood as reducing agents.

5.8 Energy wood availability in Finland for bioreducer production

The range of suitable biomass feedstock is wide, from agricultural residues to wood; however, when considering large-scale use of biomass in the steel industry, the best alternative would be energy wood, at least in Finnish conditions. Current and future demand for forest chips in other industries is one of the main factors that will determine the possibility of utilizing wood-based biomass in iron- and steelmaking applications. The energy industry utilizes vast amounts of energy wood today. According to EU objectives for renewable energy, the amount will increase further in the coming years. Additionally, there are several other biomass projects that will increase the use of energy wood use in Finland

According to the analysis conducted in Paper III, the theoretical forest chip potential was 52.6–100 TWh, the techno-ecological potential 30.0–40.4 TWh, and the techno-economic potential 20.2–27 TWh. Differences in availability assessment figures are caused by varying assumptions concerning the annual fellings, distribution coefficients used, etc. In 2011, the forest chip use in energy production was 6.8 million m³, corresponding to some 13 TWh of energy. The anticipated use of forest chips in energy production in 2020 is some 25–28 TWh. In addition to energy production, there are several projects in Finland that would use energy wood as raw material. Two scenarios where alternative biomass projects would be realized were evaluated in addition to energy wood demand in energy production. In scenario 1, there would be one biodiesel production plant, four bio-oil plants and one Bio-SNG plant. The energy wood demand of these projects would be 6.9 TWh. In scenario 2, it was assumed that instead of one biodiesel plant (3 TWh), there would be three larger plants (12 TWh). In this

scenario, the energy wood demand of alternative biomass projects would be 15.9 TWh. The total energy wood demand considering the energy production use and scenario 1 would be 34.7 TWh and energy production use, and 43.7 TWh in scenario 2.

Comparing the anticipated energy wood demand in 2020 to energy wood availability, one can see that it exceeds the techno-economic potential, which is calculated from the viewpoint of energy production. However, when compared to the techno-ecological energy wood potential, it can be concluded that there is a possibility to sustainably increase the use of energy wood in several applications. Furthermore, it is unlikely that anticipated biomass projects will be realized by the year 2020. Finnish blast furnace-based steel production could be a major user of biomass. The annual charcoal need could be as high as 300,000 tonnes, which would result in a green wood demand of 2.35 million m³, which equates to more than 4.7 TWh in energy (Paper III). It is unlikely that the maximum amount of charcoal would be used in the blast furnaces, but the replacement of fossil-based reductants could still be substantial.

The availability and demand of energy wood fractions is bound to geographical location. There are 13 forestry centers in Finland, which are considered as analysis units in many forest chip potential evaluations. Forest chip potential and anticipated use in 2020 in energy production are not evenly distributed in Finland, which can be seen in Fig. 19. Forest chip potential is techno-ecological potential, which means that ecological restrictions are taken into account, but economic restrictions are ignored.

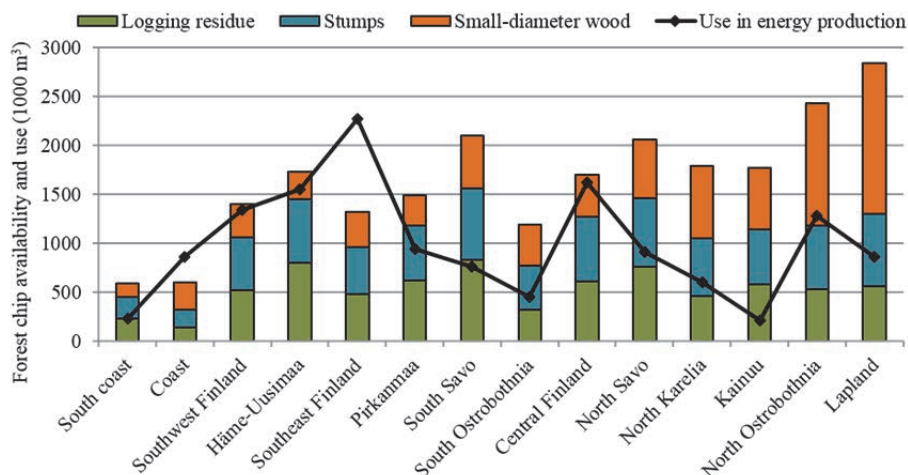


Fig. 19. Regional availability and use of forest chips (Paper III, reprinted by permission of MPDI).

Regional availability assessment suggested that availability of forest chips is negative in some forestry centers, whereas in others the availability is positive. These figures do not include the anticipated biomass projects, since their location is not certain. An interesting observation was that the availability of forest chips is higher in those forestry centers closest to the Finnish carbon steel producer (Lapland, North Ostrobothnia, Kainuu, North Savo). Demand for energy wood in these four regions is likely to remain stable, which encourages further studies of their utilization in alternative applications, e.g. as a reductant in iron- and steelmaking.

5.9 Evaluation of the reliability of the results

5.9.1 Blast furnace model

The blast furnace model gives quite reliable results in the base case simulation, compared to the reference plant values. Since the blast furnace process performance data of the reference plant cannot be distributed publicly, a comparison is made to the study by Kinnunen *et al.* (2011). The measured reductant rate with pellet operation in the reference plant was 472 kg/tHM, which is very close to the simulation results in the base case (469.0 kg/tHM). This

indicates that assumptions made in BF modeling can be held to be quite reliable when evaluating the amount of carbon needed to reduce and melt the iron oxides into hot metal. The heating value of the blast furnace top gas is slightly higher in the simulation case (3.52 MJ/Nm³), compared to the measured values (3.16 MJ/Nm³). This indicates that reduction of higher iron oxides into wüstite in the upper zone of the BF model is slightly more ineffective than in the actual process. The higher heating value means that the proportion of CO in top gas is higher in the simulation case.

The reliability of the results in the charcoal injection case (CC case 1 and CC case 2) is more difficult to assess, since there is no reference data available. However, since the equations used in the modeling of different blast furnace injectants are based on the known weight percentages of elements and known heat of combustion, the shift from base case to CC cases should result in reliable values. The input data concerning the raw material properties fed into the blast furnace is received directly from the reference plant chemical analysis data. Blast furnace heat losses and other process-specific data are also taken from the reference plant data.

5.9.2 Unit processes in gate-to-gate model

Unit process descriptions developed for the gate-to-gate steel plant model are based on industrial data and publicly available scientific literature. Thermodynamic and material property data (e.g. densities, enthalpies of reaction, heats of combustion, and heat capacities) have been obtained from the HSC Chemistry® database (Roine *et al.* 2007), which is integrated into the Factory simulation tool. The raw material data used in the calculations has been collected from the reference plant over several years and implemented in the Factory simulation tool. The reliability of the results of material and energy flow calculations can be held as trustworthy for the purpose they were developed. The main objective was to evaluate the amount of CO₂ emissions released inside the integrated steel plant in the reference case with oil injection and in cases where oil (CC case 1) and oil and part of the coke (CC case 2) are replaced with charcoal injection. The only publicly available source where the CO₂ emissions of the reference plant are described is Environmental Data Monitor (Ruukki 2014). Environmental data is provided for the years 2006–2013. The years 2012 and 2013 are comparable to the base case in the simulation model since the sintering plant was closed at the end of 2011. CO₂ emissions calculated per tonne of steel

and taken as an average for 2012 and 2013 are 1,676 kgCO₂/t steel. Simulation results for the base case CO₂ emissions with the gate-to-gate model were 1,780 kgCO₂/t hot rolled plate. The magnitude of the CO₂ emissions of the simulation results and publicly available information about the reference plant emissions are quite close to each other. The simulation conducted presents one possible operational practice, without optimization of the costs of CO₂ emissions. Several variables have an impact on the level of CO₂ emissions. Part of the coke used in the blast furnace could be purchased from an external producer, which would decrease the amount of coking coal needed in the coke plant and the CO₂ emissions produced. Also, the amount of injected reductant used has an impact on the plant site emissions.

5.9.3 Carbon footprint model

The carbon footprint model was basically divided into two separate calculation modules: forest chip production and thermochemical conversion modules, which were then combined. The forest chip production system included three different wood-based raw materials: logging residues, small-diameter wood, and stumps. The thermochemical conversion routes studied were slow pyrolysis, torrefaction and gasification plus methanation.

The forest chip production system model was based on the use of productivities (effective) and fuel consumptions of different forestry machines. In long-distance transportation, dedicated stump and chip trucks were assumed to be used. Logging residues and small-diameter wood were assumed to be chipped at the roadside, and stumps crushed at the point of use. The effective load was calculated based on either the maximum volume or mass, which depends on the moisture content and packing of the material. For chips produced from small-diameter wood and logging residues, the packing is assumed to be 0.45 cubic meters per loose cubic meter, and 0.25 for stumps. Data used in the supply chain fuel and energy consumption were mainly based on Finnish literature sources. Scientific literature was used as a source when taking into consideration the fuel and energy consumption and CO₂ emissions of electricity, diesel and fertilizer production.

The reliability of the input data concerning the forestry machines can be held as reliable, and the data describe the most common industrial forest chip supply chains in Finland. The effective productivities used for forestry machines have a great impact on the fuel and energy consumptions. In this work, technology-

specific productivities were used, which has to be taken into account when examining the results. The same chemical composition and heating value was assumed for all raw materials, i.e. logging residues, stumps, and small-diameter wood. In reality, there is variation in these parameters depending on the part of the wood and the wood species. Thermochemical conversion modules were modeled based on the available literature sources. These models are quite simple, but are based on data published in the scientific literature, which makes them trustworthy. The electricity consumption and other important process parameters of the thermochemical conversion plants are taken also from the literature, which adds to the reliability of the results.

5.9.4 Forest chip availability assessment

The forest chip availability assessment was based on available literature sources and the author's own calculations. Techno-ecological forest chip potential assessment in 2020 was done at the forestry center level based on forecasted round wood harvest level and accumulation coefficients (Kärh  *et al.* 2010). The small-diameter wood potential was taken from the work by K r h  *et al.* (2010). Forest chip potential evaluations are subject to many uncertainties. The anticipated round wood harvest level is affected by several factors, e.g. the development of the Finnish forest industry and the use of imported round wood, which in turn impacts the availability of logging residues and stumps. There is also variation in the coefficients between different literature sources concerning the yields of logging residues and stumps from the stemwood, recovery rates and ecological constraints. Many forest chip availability studies do not take pine and birch stumps into account, because their excavation is more difficult than with spruce stumps. In this assessment, pine and birch stumps are considered. The availability assessments of small-diameter wood have similar uncertainties than with logging residue and stump availability assessments. Despite these uncertainties, the availability assessment gives valuable information about the techno-ecological potential of using forest chips in reductant production.

6 Summary and conclusions

6.1 Summary

The application of biomass in iron and steelmaking processes can be a promising measure to decrease the amount of non-renewable resources used in steel production. Before the large-scale introduction of biomass in steel industry, several aspects have to be clarified. In this thesis, bioreducer use as a blast furnace reductant was investigated at different system levels.

In this work, the chemical and physical properties of bioreducers for blast furnace use were reviewed and compared to fossil-based reductants. The impact of bioreducers on the blast furnace process was evaluated based on the coke replacement ratios and blast furnace modeling. According to the blast furnace simulation results, the coke replacement ratio of charcoal is 0.97. The gate-to-gate CO₂ reduction potential when using biomass as a reductant in the blast furnace was calculated with the developed steel plant model. CO₂ emissions could be reduced by 15.4% at the plant scale by replacing oil injection with charcoal injection. Integrating charcoal production into an integrated steel plant makes it possible to decrease the environmental burden of steelmaking by utilizing the energetic off-gases from the pyrolysis process. According to the simulation results, CO₂ emissions in this scenario with gate-to-gate system boundaries would drop by 26.4%.

A life cycle model was developed to calculate the carbon footprint and energy return on investment of bioreducers from Finnish energy wood. The carbon footprint assessment conducted for torrefied wood, charcoal and Bio-SNG showed that there is substantial potential to decrease the fossil CO₂ emissions of steelmaking by utilizing bioreducers in the BF. The energy return on investment of all the bioreducers is well above one (3.1–6.2), which means that more energy is gained than has been used to produce the new unit of energy. The production cost analysis was conducted for bioreducers produced from Finnish energy wood. The calculated production costs were €140–180/t for torrefied wood, €360–490/t for charcoal, and €690–830/t for Bio-SNG. CO₂ mitigation costs for globally avoided CO₂ emissions when replacing coke with bioreducers were calculated based on the production cost analysis. CO₂ mitigation costs without by-product credits were €33–69/tCO₂, €22–53/tCO₂ and €112–150/tCO₂ for charcoal, torrefied wood, and Bio-SNG respectively.

The availability of domestic biomass in Finland (namely energy wood: logging residues, stumps and small-diameter wood) was evaluated at the national and forestry center levels. The assessment revealed that there is true potential to increase the use of forest chips beyond the traditional use in heat and power production in Finland.

The bioreducers examined in this thesis are summarized in Table 8 with an evaluation of their advantages and disadvantages.

Table 8. Comparison of bioreducers evaluated in this thesis.

Bioreducer	Torrefied biomass	Charcoal	Bio-oil	Syngas	Bio-SNG
Use in BF	Injection	Injection, top-charging	Injection	Injection	Injection
Coke replacement ratio in BF	~0.4	0.8–1.15	0.25–0.35	NA	0.9–1.15
Amount in BF (kg/tHM)	120	200	<100	NA	90–150
Metallurgical properties	Comparable to low-quality coal. Medium heating value. Low ash content. Low sulfur content.	Comparable to good-quality coal. High heating value. Low ash content. Low sulfur content.	Low heating value. High oxygen and moisture content.	The amount of N ₂ decreases in the BF → Facilitates CO ₂ capture. Low sulfur and alkali content.	Similar properties to natural gas. Low sulfur and alkali content.
Behavior in BF	Lowers the adiabatic flame temperature. Amount of gas increases	Increased productivity. Sufficient mass flow in injection might be challenging.	Lowers the adiabatic flame temperature.	The use of reducing gas injection is not proven at the large scale.	The injection of natural gas is common practice. Supercooling of the BF hearth.
Upgrading technology	Uncomplicated. No large-scale production units. High energy efficiency.	Technology is uncomplicated. Typically small-scale. Low carbon and energy yield of product.	Technologies are still at development stage. Commercial plants have been	Co-gasification of biomass and fossil-based feedstock. Use of low-quality biomass. Technologies	Co-gasification of biomass and fossil-based feedstock. High energy yield of product. Use of low-quality biomass.

Bioreducer	Torrefied biomass	Charcoal	Bio-oil	Syngas	Bio-SNG
			constructed. Can be integrated e.g. with CHP plant (example in Finland).	under development Oxygen/steam as gasification agent.	Technologies under development Oxygen/steam as gasification agent.
Carbon footprint without co-product credits in Finland (kg/t bioreducer)	106–122	214–267	Not evaluated	Not evaluated	368–426
Energy return on investment (Finland)	5.4–6.2	3.2–3.9	Not evaluated	Not evaluated	3.1–3.5
Global CO ₂ reduction potential (Finnish example) (M t/year)	0.40	0.99–1.01 (without by-product credits) 1.29–1.39 (with by-product credits)	Not evaluated	Not evaluated	0.84–0.86 (without by-product credits) 0.89–0.97 (with by-product credits)
CO ₂ mitigation cost (Finnish example) (€/tCO ₂)	22–53	33–69 (without by-product credits) 26–54 (with by-product credits)	Not evaluated	Not evaluated	112–150 (without by-product credits) 107–143 (with by-product credits)
Process integration	Use of waste heat from steel plant in biomass drying. Integration with energy production.	Use of waste heat from steel plant in biomass drying. Use of pyrolysis gas in energy and chemical production.	Use of waste heat from steel plant in biomass drying. Integration with energy production.	Use of waste heat from steel plant in biomass drying. Integration with energy production.	Use of waste heat from steel plant in biomass drying. Integration with energy production.

6.2 Concluding remarks

The main aim of this thesis was to gain knowledge of the feasibility of using bioreducers in Finnish blast furnace ironmaking. The main results of this thesis are as follows: Charcoal produced from wood-based biomass has the most

feasible metallurgical properties for BF injection. It has a high heating value and carbon content, a low ash content, and suitable physical properties. Torrefied wood could be part of the injection mix, but as a single injectant it has a heating value that is too low and an oxygen content that is too high. Syngas and Bio-SNG were also found to be suitable injectants, since their composition could be tuned to resemble that of fossil-based reductants already used in BF ironmaking.

Injection of charcoal to the blast furnace results in the largest replacement of fossil-based reductants. Charcoal has a high coke replacement ratio, higher than that of pulverized coals. Use of low-ash charcoal in blast furnace injection would result in decreasing slag and gas amounts, which leads to more efficient blast furnace operation. In the case where oil is replaced with charcoal produced outside the system boundaries, plant site impacts are modest. The calorific value of the blast furnace gas produced decreases to some extent, which impacts the output of the power plant by lowering the internally produced electricity. Integrating charcoal production into an integrated steel plant makes it possible to decrease the environmental burden of steelmaking by utilizing the energetic off-gases from the pyrolysis process.

The environmental feasibility of bioreducer use as a blast furnace reductant is positive. The carbon footprint of three bioreducers (torrefied wood, charcoal and Bio-SNG produced from Finnish energy wood) was considerably lower than with the fossil-based reductants coal and natural gas when indirect carbon stock change was not considered. According to the energy return on investment analysis, production of bioreducers with modern technologies from wood-based feedstock is feasible in Finland.

The economic comparison of bioreducers to fossil-based reductants showed that the bioreducer production costs are clearly higher than fossil-based reducing agents. In the case of wood-based bioreducers, the cost of the raw material is one of the largest contributors to the final bioreducer production cost. Despite the fact that bioreducers do not seem to be economically feasible today, they outperform the carbon capture and storage option, which involves heavy investments and transportation of CO₂ to storage areas.

The energy wood availability assessment revealed that there is a potential to use energy wood in new applications. There are regions in Finland where the anticipated use of energy wood in 2020 is lower than the techno-ecological energy wood potential. The use of energy wood is bound to regional use, because transportation of forest chips is not feasible over long distances. The regions with

the largest energy wood potential are close to Finnish steel plants, which encourages future energy wood availability studies.

This thesis provides valuable new knowledge for the steel industry to develop the production in a more sustainable direction from the perspective of global climate change. Methodologies related to industrial ecology (i.e. systems analysis and carbon footprint analysis) were used to improve the understanding of the impacts of using wood-based biomass in the steel industry as a reductant. The blast furnace and other gate-to-gate models developed are transferrable to other case studies where biomass is introduced into the steel production system with moderate changes. This thesis also has a strong case-specific view, since a considerable proportion of the data used is of Finnish origin. As a whole, this thesis provides new insights into the technical, environmental and economic opportunities for and barriers to the use of biomass as a reductant in the Finnish steel industry.

6.3 Recommendations for further research

First of all, there is a need to do further research at the unit process level (blast furnace) and also to investigate the pretreatment of bioreducers and the impact of bioreducers on off-gas cleaning systems. The behaviors of torrefied biomass and charcoal have been investigated by various means, but there is still a need to clarify, for instance, how the combustion behavior of bioreducers would impact gas flows inside the blast furnace. There are also changes in gas composition, which impact the reduction behavior of iron oxides in the furnace. Changes in gas flows and reduction behavior would be even more drastic if syngas or Bio-SNG were used as reductants. Pre-treatment of coal and solid bioreducers before injection into the blast furnace includes drying and pulverization. Because co-injection of fossil-based and biomass-based reducing agents would possibly be the most feasible option, there is a need to assess how the pre-treatment should be carried out. Whether coal and solid bioreducers can be ground at the same mills or whether it should be done separately needs to be clarified. Torrefied biomass and charcoal are also quite flammable materials, so their drying, storage and handling should be designed carefully. Injection trials (e.g. in experimental blast furnaces) could be conducted before industrial trials in modern, large-scale blast furnaces.

Secondly, the sustainability of the raw material supply should be carefully examined. Wood is one of the most suitable raw materials for bioreducer

production, but other biomass-based raw materials are also available. Increasing the use of biomass in energy production purposes or in reductant purposes should also mean that more emphasis is placed on selecting raw material sources that do not contribute considerably to carbon stock change through soils. It would also be beneficial if biomass upgrading processes could be kept to a minimum.

Finally, there is a need to develop feasible production platforms to produce bioreducers and possibly other, more valuable, products from biomass simultaneously. This would make sense from the economic point of view, but also from the environmental perspective. There could be several possibilities to integrate bioreducer production into the steel industry. Firstly, there is excess heat available in integrated steel plants, which could be used in biomass drying. Low temperature heat sources include e.g. district heat water and hot stove off-gases. These two process integration possibilities could be realized with reasonably low changes in current infrastructure. By utilizing excess heat in biomass drying, there could be fuel savings in bioreducer production plants.

A significant amount of thermal energy is bound up in iron- and steelmaking slags in the form of sensible heat. The temperatures of iron- and steelmaking slags are high: the temperature of blast furnace slag is around 1,500 °C. This waste heat could be recovered and used in the thermochemical conversion of biomass.

One additional innovative approach could be the utilization of blast furnace gases as a raw material to produce valuable fuels and chemicals by Fischer-Tropsch or methanol synthesis. If a bioreducer is used as a blast furnace injectant, part of the blast furnace gas could be considered carbon-neutral. Ultimately, the goal should be finding synergies between biomass refining and the metallurgy industry to maximize the potential of material and energy exchanges, as well as to minimize logistic challenges. Building integrated metallurgy and biomass refining plants would also contribute to realizing industrial ecology in practice.

References

- Ahmed HM, Viswanathan N & Björkman B (2013) Composite Pellets – A Potential Raw Material for Iron-Making. *Steel Res Int* 84: 1–14.
- Alam A, Kilpeläinen A & Kellomäki S (2010) Potential Energy Wood Production with Implications to Timber Recovery and Carbon Stocks Under Varying Thinning and Climate Scenarios in Finland. *Bioenergy Res* 3(4): 362–372.
- Antal Jr MJ & Gronli M (2003) The Art, Science, and Technology of Charcoal Production. *Ind Eng Chem Res* 42(8):1619–1640.
- Babich AI, Gudenau HW, Mavrommatis KT, Froehling C, Formoso A, Cores A & Garcia L (2002) Choice of technological regimes of a blast furnace operation with injection of hot reducing gases. *Revista de Metalurgia* 38(4): 288–305.
- Babich A, Senk D & Fernandez M (2010) Charcoal Behaviour by Its Injection into the Modern Blast Furnace. *ISIJ Int* 50: 81–88.
- Bailis R, Rujanavech C, Dwivedi P, de Oliveira Vilela A, Chang H & de Miranda RC (2013) Innovation in charcoal production: A comparative life-cycle assessment of two kiln technologies in Brazil. *Energy Sustain Dev* 17(2): 189–200.
- Birat JP, Borlée J, Lavelaine H, Négro P, Meijer K, van der Stel J & Sikstrom P (2012) ULCOS program: an update 2012. *Proc 4th International Conference on Process Development in Iron and Steelmaking (SCANMET IV)*, Luleå, Sweden.
- Birat, JP (2009) Steel and CO₂ – the ULCOS program, CCS and Mineral carbonation using Steelmaking Slag, *Proc 1st International Slag Valorisation Symposium*, Leuven, Belgium.
- Birat JP (2011) CO₂-lean steelmaking: ULCOS, other international programs and emerging concepts. *Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry*, Düsseldorf, Germany.
- Brent GF, Allen DJ, Eichler BR, Petrie JG, Mann JP & Haynes BS (2012) Mineral Carbonation as the Core of an Industrial Symbiosis for Energy-Intensive Minerals Conversion. *J Ind Ecol* 16(1): 94–104.
- Bridgewater AV (2003) Renewable fuels and chemical by thermal processing of biomass. *Chem Eng J* 91(2-3): 87–102.
- Bridgewater AV (2012) Review of fast pyrolysis of biomass and product upgrading. *Biomass Bioenerg* 38: 68–94.
- Burchart-Korol D (2013) Life cycle assessment of steel production in Poland: a case study. *J Clean Prod* 54: 235–243.
- Bürgler T & Di Donato A (2008) Biomass Gasification for DRI production. *Revue de Metallurgie* 106: 429-433
- Bürgler T, Schuster S, Nograting H, Ferstl A, Pillmair G & Schuster E (2011) Portfolio of reducing agents for CO₂-lean ironmaking at voestalpine. *Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry*, Düsseldorf, Germany.

- Burnham A, Han J, Clark CE, Wang M, Dunn JB & Palou-Rivera I (2012) Life-Cycle Greenhouse Gas Emissions of Shale Gas, Natural Gas, Coal, and Petroleum. *Environ Sci Technol* 46(2):619–627.
- Channiwala SA & Parikh PP (2002) A unified correlation for estimating HHV of solid, liquid and gaseous fuels. *Fuel* 81(8): 1051–1063.
- Chen WH & Wu JS (2009) An evaluation on rice husks and pulverized coal blends using a drop tube furnace and a thermogravimetric analyzer for application to a blast furnace. *Energy* 34(10): 1458–1466.
- Chen WH, Du SW, Tsai CH & Wang ZY (2012) Torrefied biomasses in a drop tube furnace to evaluate their utility in blast furnaces. *Bioresour Technol* 111: 433–438.
- Chertow MR (2000) Industrial Symbiosis: Literature and Taxonomy. *Annu Rev Energy Environ* 25: 313–337.
- Collins Dictionary (2013) English Dictionary – Pioneers in dictionary publishing since 1819. URI: <http://www.collinsdictionary.com/dictionary/english>. Cited 2014/1/8.
- Crelling JC (1989) Coal as a material. In: Marsh H (ed) *Introduction to Carbon Science*. London, Butterworth & Co: 260–275.
- Czernik S & Bridgewater AV (2004) Overview of Applications of Biomass Fast Pyrolysis Oil. *Energy Fuels* 18(2): 590–598.
- Danloy G, van der Stel J & Schmöle P (2008) Heat and mass balances in the ULCOS Blast Furnace. *Proc of the 4th Ulcoss seminar*.
- de Castro JA, da Silva AJ, Sasaki Y & Yagi J (2011) A Six-phases 3-D Model to Study Simultaneous Injection of High Rates of Pulverized Coal and Charcoal into the Blast Furnace with Oxygen Enrichment. *ISIJ Int* 51(5): 748–758.
- de Castro JA, de Mattos Araújo G, de Oliveira da Mota I, Sasaki Y & Yagi J (2013) Analysis of the combined injection of pulverized coal and charcoal into large blast furnaces. *J Mater Res Technol* 2(4): 308–314.
- Ertem ME & Özdabak A (2005) Energy balance application for Erdemir Coke Plant with thermal camera measurements. *Appl Therm Eng* 25: 423–433.
- Diez MA, Alvarez R & Fernández M (2012) Biomass derived products as modifiers of the rheological properties of coking coals. *Fuel* 96: 306–313.
- Diez MA & Borrego AG (2013) Evaluation of CO₂ - reactivity patterns in cokes from coal and woody biomass blends. *Fuel* 113: 59–68.
- Environmental Data Monitor (2014) Ruukki Corporate Responsibility. URI: <http://www.ruukki.com/Corporate-responsibility/Environment/Environmental-data-monitor>. Cited 2014/1/20.
- Fagernäs L, Kuoppala E, Tiilikkala K & Oasmaa A (2012) Chemical Composition of Birch Wood Slow Pyrolysis Products. *Energy Fuels* 26(2): 1275–1283.
- Fan J, Kalnes TN, Alward M, Klinger J, Sadehvandi A & Shonnard DR (2011) Life cycle assessment of electricity generation using fast pyrolysis bio-oil. *Renewable Energy* 36(2): 632–641.
- Garcia R & Freire F (2014) Carbon footprint of particleboard: a comparison between ISO/TS 14067, GHG Protocol, PAS 2050 and Climate Declaration. *J Clean Prod* 66: 199–209.

- Fortum (2013) Fortum's bio-oil plant commissioned in Joensuu - first of its kind in the world. URI: <https://www.fortum.fi/en/mediaroom/Pages/fortums-bio-oil-plant-commissioned-in-joensuu-first-of-its-kind-in-the-world.aspx>. Cited 2014/1/22.
- Gaunt J & Lehmann J (2008) Energy balance and emissions associated with biochar sequestration and pyrolysis bioenergy production. *Environ Sci Technol* 42(11): 4152–4158.
- Geerdes M, Toxopeus H & van der Vliet C (2009) *Modern Blast Furnace Ironmaking: an Introduction*, (2nd edition). IOS Press, Amsterdam, The Netherlands.
- Gibbs D, Deutz P (2007) Reflections on implementing industrial ecology through eco-industrial park development. *J Clean Prod* 15(17): 1683–1695.
- Goncalves LJ, Manella R, da Silva FR (2012) The use of coke and charcoal in Aperam South America. 6th International Congress on the Science and Technology of Ironmaking - ICSTI.
- Hacatoglu K, McLellan PJ & Layzell DB (2010) Production of Bio-Synthetic Natural Gas in Canada. *Environ Sci Technol* 44(8): 2183–2188.
- Hakkila P (2004) Developing technology for large-scale production of forest chips. Wood Energy Technology Programme 1999–2003. Final report, technology programme report 6/2004, Tekes.
- Hanrot F, Sert D, Delinchant J, Pietruck R, Bürgler T, Babich A, Fernández, M, Alvarez R & Diez MA (2009) CO₂ Mitigation for Steelmaking using Charcoal and Plastics Wastes as Reducing Agents and Secondary Raw Materials, Proc 1st Spanish National Conference on Advances in Materials Recycling and Eco-Energy, Madric, Spain.
- Hanrot F, Sert D, Babich A, Pitrucek R, Fernandez M, Diez MA, Bürgler T & Delinchant-Lété J (2011) Short term CO₂ mitigation for steelmaking (SHOCOM). Contract no. RFSR-CT-2005-00001; 1 July 2005–30 June 2008.
- Helle H, Helle M, Saxén H & Pettersson F (2009) Mathematical Optimization of Ironmaking with Biomass as Auxiliary Reductant in the Blast Furnace. *ISIJ Int* 49(9): 1316–1324.
- Hooley PL, Bodén A, Wang C, Grip CE, Jansson B (2010) Design and Application of a Spreadsheet-based Model of the Blast Furnace Factory. *ISIJ Int* 50(7): 924–930.
- Hooley L, Tobiesen A, Johns J, Santos S (2013) Techno-economic study of an integrated steelworks equipped with oxygen blast furnace and CO₂ capture. *Energy Procedia* 37: 7139–7151.
- Hu C, Han X, Li, Z, Zhang C (2009) Comparison of CO₂ emission between COREX and blast furnace iron-making system. *J Environ Sci (Suppl 1)* 21: S116–S120.
- IEA (2007) *Tracking industrial energy efficiency and CO₂ emissions*. Paris, France, International Energy Agency, 324 pp.
- IEAGHG (2013) *Overview to the Current State and Development of CO₂ Capture technologies in the Ironmaking Process*. 2013/TR3, April 2013.
- Iosif AM, Hanrot F, Birat JP & Ablitzer D (2010) Physicochemical modelling of the classical steelmaking route for life cycle inventory analysis. *Int J Life Cycle Assess* 15(3): 304–310.

- ISO/TS 14067: Greenhouse Gases e Carbon Footprint of Products – Requirements and Guidelines for Quantification and Communication (Technical Specifications). International Organization for Standardization, Geneva, 2013.
- ISO 14040: Environmental Management e Life Cycle Assessment – Principles and Framework. International Organization for Standardization, Geneva, 2006.
- ISO 14044: Environmental Management - Life Cycle Assessment – Requirements and Guidelines. International Organization for Standardization, Geneva, 2006.
- Jahanshahi S, Xie D, Pan Y, Ridgeway P & Mathieson J (2011) Dry Slag Granulation with Integrated Heat Recovery. Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry, Düsseldorf, Germany.
- Jelinski L, Graedel T, Laudise R, McCall D & Patel C (1992) Industrial ecology: concepts and approaches. Proc National Academy of Sciences 89: 793–797.
- Johansson MT, Söderström M (2011) Options for the Swedish steel industry – Energy efficiency measures and fuel conversion. Energy 36:191–198.
- Kasai A & Matsui Y (2004) Lowering of Thermal Reserve Zone Temperature in Blast Furnace by Adjoining Carbonaceous Material and Iron Ore. ISIJ Int 44(12): 2073–2078.
- Karlsson M & Wolf A (2008) Using an optimization model to evaluate the economic benefits of industrial symbiosis in the forest industry. J Clean Prod 16(14): 1536–1544.
- Kinnunen K, Paananen T & Lilja J (2011) Modelling and simulation of Blast Furnace Process for switch from Sinter to Pellet Operation. Metec InSteelCon 2011, Proc 4th International Conference on Modelling and Simulation of Metallurgical Processes in Steelmaking, Düsseldorf, Germany.
- Knop K, Hallin M, Burström E (2008) ULCORED SP 12 Concept for minimized CO₂ emissions. Proc 4th Ulcos seminar.
- Konishi H, Ichikawa K & Usui T (2010) Effect of Residual Volatile Matter on Reduction of Iron Oxide in Semi-charcoal Composite Pellets. ISIJ Int 50(3): 386–389.
- Kumar A, Jones DD & Hanna MA (2009) Thermochemical Biomass Gasification: A Review of the Current Status of the Technology. Energies 2(3): 556–581.
- Kärhä K, Elo J, Lahtinen P, Räsänen T, Keskinen S, Saijonmaa P, Heiskanen H, Strandström M & Pajuoja H (2010) Kiinteiden puupolttoaineiden saatavuus ja käyttö Suomessa vuonna 2020. Publications of the Ministry of the Employment and the Economy of Finland, Energy and the Climate 66/2010.
- Larsson M (2004) Process Integration in the Steel Industry – Possibilities to Analyse Energy Use and Environmental Impacts for an Integrated Steel Mill. Doctoral Thesis. Luleå Univ of Technology. Dept of Applied Physics and Mechanical Engineering. Div of Energy Engineering.
- Larsson M, Görling M, Grönkvist S, Alvfors (2013) Bio-methane upgrading of pyrolysis gas from charcoal production. Sustain Energy Technol Asses 3: 66–73.
- Link J (2008) IRMA – Flowsheet Model Examples of Application. Proc 4th Ulcos seminar.

- Liski J, Repo A, Känkänen R, Vanhala P, Seppälä J, Antikainen R, Grönroos J, Karvosenoja N, Lähtinen K, Leskinen P, Paunu VV & Tuovinen JP (2011) Metsäbiomassan energiakäytön ilmastovaikutukset Suomessa (forest bioenergy: greenhouse gas emissions and climate impacts in Finland), The Finnish, Environment, vol. 5.
- Luengen HB, Peters M, Schmöle P (2011) Iron Making in Western Europe. Metec InSteelCon 2011, Proc 6th European Coke and Ironmaking Congress, Düsseldorf, Germany.
- Lundgren J, Ekblom T, Hultberg C, Larsson M, Grip CE, Nilsson L & Tunå R (2013) Methanol production from steel-work off-gases and biomass based synthesis gas. Appl Energy 112: 431–439.
- Machado JGMS, Osório E, Vilela ACF, Babich A, Senk D & Gudenau HW (2010a) Reactivity and Conversion Behaviour of Brazilian and Imported Coals, Charcoal and Blends in view of their Injection into Blast Furnaces. Steel Res Int 81(1): 9–16.
- Machado JGMS, Osório E, Vilela CF (2010b) Reactivity of Brazilian Coal, Charcoal, Imported Coal and Blends Aiming to Their Injection into Blast Furnaces. Mat Res 13(3):287–92.
- MacPhee JA, Gransden JF, Giroux L & Price JT (2009) Possible CO₂ mitigation via addition of charcoal to coking blends. Fuel Process Technol 90:16–20.
- Maidell M, Pyykkönen P & Toivonen R (2008) Metsäenergiapotentiaalit Suomen maakunnissa. Pellervo Economic Research Institute Working Papers. No 106, 2008.
- Makkonen HT, Heino J, Laitila L, Hiltunen A, Pöyliö E, Härkki J (2002) Optimisation of steel plant recycling in Finland: dusts, scales and sludge. Resour Conserv Recycl 35:77–84.
- Mathews JA & Tan H (2009) Biofuels and indirect land use change effects: the debate continues. Biofuels Bioprod Biorefin 3(3): 305–317.
- Mathieson JG, Truelove JS & Rogers H (2005) Toward an understanding of coal combustion in blast furnace tuyere injection. Fuel 84(10): 1229–1237.
- Mathieson JG, Rogers H, Somerville M, Ridgeway P & Jahanshahi S (2011) Use of Biomass in the Iron and Steel Industry – An Australian Perspective. Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry, Düsseldorf, Germany.
- Mathieson JG, Rogers H, Somerville MA & Jahanshahi S (2012) Reducing Net CO₂ Emissions Using Charcoal as a Blast Furnace Tuyere Injectant. ISIJ Int 52(8): 1489–1496.
- McKendry P (2002) Energy production from biomass (part 3): gasification technologies. Bioresour Technol 83(1): 55–63.
- Meijer K, Denys M, Lasar J, Birat JP, Still G & Overmaat B (2009) ULCOS: ultra-low CO₂ steelmaking. Ironmaking Steelmaking 36(4): 249–251.
- Meijer K, Guenther C & Dry RJ (2011) HIsarna Pilot Plant Project. Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry, Düsseldorf, Germany.

- Metla (2013) Metsätilastotiedote 15/2103. Metsähakkeen käyttö uuteen ennätykseen vuonna 2012: 8,3 miljoonaa kuutiometriä (in Finnish). URI: <http://www.metla.fi/metinfo/tilasto/julkaisut/mtt/2013/puupolttoaine2012.pdf>. Cited 2014/2/17.
- Miao X & Wu Q (2004) High yield bio-oil production from fast pyrolysis by metabolic controlling of *Chlorella protothecoides*. *J Biotechnol* 110(1): 85–93.
- Midrex (2013) World Direct Reduction Statistics 2012. URI: http://www.midrex.com/handler.cfm/cat_id/153/section/company. Cited 2013/12/13.
- Ministry of Employment and the Economy (2010) Kohti vähäpäästöistä Suomea. Presentation of Minister Mauri Pekkarinen 2010. URI: http://www.tem.fi/files/26643/UE_lo_velvoitepaketti_Kesaranta_200410.pdf. Cited 2014/1/20.
- Miwa T, Okuda H, Osame M, Watakabe S & Saito K (2011) COURSE50-CO₂ Ultimate Reduction in Steelmaking process by innovative technology for cool Earth 50: CO₂ emission reduction technology in Japan. Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry, Düsseldorf, Germany.
- Mohan D, Pittman Jr. CU & Steele PH (2006) Pyrolysis of Wood/Biomass for Bio-oil: A Critical Review. *Energy Fuel* 20(3): 848–889.
- Montiano MG, Diaz-Faes E, Barriocanal C & Alvarez R (2014) Influence of biomass on metallurgical coke quality. *Fuel* 116: 175–182.
- Murphy DJ & Hall CAS (2010) Year in review–EROI or energy return on (energy) invested. *Ann N Y Acad Sci* 1185: 102–118.
- Ng KW, MacPhee JA, Giroux L & Todoschuk T (2011) Reactivity of bio-coke with CO₂. *Fuel Process Technol* 92(4): 801–804.
- Ng KW, Giroux L, MacPhee T & Todoschuk T (2010) Direct Injection of Biofuel in Blast Furnace Ironmaking. AISTech Conference Proc, Pittsburgh, PA.
- Nogami H, Yagi J & Sampaio RS (2004) Exergy Analysis of Charcoal Charging Operation of Blast Furnace. *ISIJ Int* 44(10): 1646–1652.
- Nomura S, Ayukawa H, Kitaguchi H, Tahara T, Matsuzaki S, Naito M, Koizumi S, Ogata Y, Nakayama T & Abe T (2005) Improvement in blast furnace reaction efficiency through the use of highly reactive calcium rich coke. *ISIJ Int* 45(3): 316–324.
- Noldin JH (2011) Energy Efficiency and CO₂ Reduction in the Brazil Steel Industry. Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry, Düsseldorf, Germany.
- Norgate T & Langberg D (2009) Environmental and Economic Aspects of Charcoal Use in Steelmaking. *ISIJ Int* 49(4): 587–595.
- Norgate T, Haque N, Somerville M & Jahanshahi S (2012) Biomass as a Source of Renewable Carbon for Iron and Steelmaking. *ISIJ Int* 52(8): 1472–1481.
- Ooi TZ, Thompson D, Anderson DR, Fisher R, Fray T, Zandi M (2011) The effect of charcoal combustion on iron-ore sintering performance and emission of persistent organic pollutants. *Combust Flame* 158(5): 979–987.

- Paananen T, Kinnunen K, Lilja J & Pisilä E (2011) Experience of full scale research test with 100% pellet blast furnace operation in Ruukki. Proc 6th European Coke and Ironmaking Congress, Düsseldorf, Germany.
- Peacey JG & Davenport WG (1979): The Iron Blast Furnace, Pergamon Press, Oxford.
- Peacocke GVC, Bridgewater AV & Brammer JC. Techno-economic assessment of power production from the Wellman and BTG fast pyrolysis process, in: Bridgewater AV & Boocock DGB (Eds.), Thermal and Chemical Biomass Conversion, CPL Press, Newbury, Berkshire, UK, 2006.
- Phanphanich M & Mani S (2011) Impact of torrefaction on the grindability and fuel characteristics of forest biomass. *Bioresour Technol* 102(2): 1246–1253.
- Piketty MG, Wichert M, Fallot A & Aimola L (2009) Assessing land availability to produce biomass for energy: The case of Brazilian charcoal for steel making. *Biomass Bioenerg* 33(2): 180–190.
- Plevin RJ, O'Hare M, Jones AD, Torn MS & Gibbs HK (2010) Biofuels and indirect land use change effects: the debate continues. *Environ Sci Technol* 44: 8015–8021.
- POSCO (2011) Carbon Report 2011 – Toward a Sustainable Society. URI: <http://www.iea.org/media/weowebiste/ebc/POSCOCarbonReport2011.pdf>. Cited 2014/1/3.
- Poveromo JJ (2013) ICSTI-12 A report on the international ironmaking conference. *Steel Times International*. URI: <http://www.steeltimesint.com/issue-archive/view/march-2013>. Cited 2013/1/10.
- Prins MJ, Ptasinski KJ, Janssen FJJG (2006) More efficient biomass gasification via torrefaction. *Energy* 31(15): 3458–3470.
- Pöyry Energy (2007) Puupolttoaineiden kysyntä ja tarjonta Suomessa vuonna 2020–Päivitetty tilannekatsaus. Pöyry Energy Oy, 2007.
- Rasul MG, Tanty BS, Mohanty TB (2007) Modelling and analysis of blast furnace performance for efficient utilization of energy. *Appl Therm Eng* 27(1): 78–88.
- Raugei M, Fullana-i-Palmer P & Fthenakis V (2012) The energy return on energy investment (EROI) of photovoltaics: Methodology and comparisons with fossil fuel life cycles. *Energy Policy* 45: 576–582.
- Repo A, Tuomi M & Liski J (2011) Indirect carbon dioxide emissions from producing bioenergy from forest harvest residues. *GCB Bioenergy* 3(2):107–115.
- Roberts KG, Gloy BA, Joseph S, Scott NR & Lehmann J (2010) Life Cycle Assessment of Biochar Systems: Estimating the Energetic, Economic, and Climate Change Potential. *Environ Sci Technol* 44(2):827–833.
- Roine A, Lamberg P, Mansikka-aho J, Björklund P & Kotiranta T (2007) HSC Chemistry® v. 6.12, Outotec Research Oy, Pori.
- Ruukki (2014) Environmental Data Monitor, Corporate responsibility report 2013, Environmental responsibility. URI: <http://www.ruukki.com/Investors/Corporate-responsibility-report-2013/Environmental-responsibility/Environmental-data-monitor>. Cited 2014/10/3.

- Seager TP & Theis TL (2002) A uniform definition and quantitative basis for industrial ecology. *J Clean Prod* 10: 225–235.
- Scott DS, Majerski P, Piskorz J, Radlein D (1999) A second look at fast pyrolysis of biomass – the RTI process. *J Anal Appl Pyrol* 51: 23–37.
- Shabangu S, Woolf D, Fisher EM, Angenent LT, Lehmann (2014) Techno-economic assessment of biomass slow pyrolysis into different biochar and methanol concepts. *Fuel* 117(part A): 742–748.
- Shen YS, Guo BY, Yu AB & Zulli P (2009) A three-dimensional numerical study of the combustion of coal blends in blast furnace. *Fuel* 88: 255–263.
- Sjolie HK (2012) Reducing greenhouse gas emissions from households and industry by the use of charcoal from sawmill residues in Tanzania. *J Clean Prod* 27: 109–117.
- Slaby S, Andahazy D, Winter F, Feilmayer C & Bürgler T (2006) Reducing Ability of CO and H₂ of Gases Formed in the Lower Part of the Blast Furnace by Gas and Oil Injection. *ISIJ Int* 46(7): 1006–1013.
- Sokka L, Lehtoranta S, Nissinen A, Melanen M (2010) Analyzing the Environmental Benefits of Industrial Symbiosis. *J Ind Ecol* 15:137–155.
- Steele P, Puettman ME, Penmetsa VK & Cooper JE (2012) Life-Cycle Assessment of Pyrolysis Bio-Oil Production. *Forest Product Journal* 62(4): 326–334.
- Steubing B, Zaf R & Ludwig C (2011) Life cycle assessment of SNG from wood for heating, electricity, and transportation. *Biomass Bioenerg* 35(7): 2950–2960.
- Suopajarvi H & Fabritius T (2012) Effects of Biomass Use in Integrated Steel Plant–Gate-to-gate Life Cycle Inventory Method. *ISIJ Int* 52(5): 779–787.
- Suopajarvi H & Fabritius T (2013) Towards More Sustainable Ironmaking—An Analysis of Energy Wood Availability in Finland and the Economics of Charcoal Production. *Sustainability* 5(3): 1188–1207.
- Suopajarvi H, Pongrácz E & Fabritius T (2013) The potential of using biomass-based reducing agents in the blast furnace: A review of thermochemical conversion technologies and assessments related to sustainability. *Renew Sustain Energy Rev* 25: 511–528.
- Suopajarvi H, Pongrácz E & Fabritius T (2014) Bioreducer use in Finnish blast furnace ironmaking – Analysis of CO₂ emission reduction potential and mitigation cost. *Appl Energy* 124: 82–93.
- Takeda K, Anyashiki T, Sato T, Oyama N, Watakabe S & Sato M (2011) Recent Developments and Mid- and Long-Term CO₂ Mitigation Projects in Ironmaking. *Steel Res Int* 82(5): 512–520.
- Thomas S, McKnight SJ, Serrano EJ, Maslejova A, Zelinsky R, Tomás J & Vlasic P (2011) Laboratory evaluation of biomass usage for coke and sinter production. *Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry*, Düsseldorf, Germany.
- Tsupari E, Kärki J, Arasto A, Pisilä E (2013) Post-combustion capture of CO₂ at an integrated steel mill – Part II: Economic feasibility. *Int J Greenhouse Gas Control* 16:278–286.

- Turkdogan ET (1996) *Fundamental of steelmaking*. The Institute of Materials, London, United Kingdom.
- Uslu A, Faaij APC & Bergman PCA (2008) Pre-treatment technologies, and their effect on international bioenergy supply chain logistics. *Techno-economic evaluation of torrefaction, fast pyrolysis and pelletisation*. *Energy* 33(8): 1206–1223.
- van der Meijden CM, Veringa HJ & Rabou LPLM (2010) The production of synthetic natural gas (SNG): A comparison of three wood gasification systems for energy balance and overall efficiency. *Biomass Bioenerg* 34(3): 302–311.
- Van der Stel J, Louwerse G, Sert D, Hirsch A, Eklund N & Pettersson M (2013) Top gas recycling blast furnace development for ‘green’ and sustainable ironmaking. *Ironmaking Steelmaking* 40(7): 483–489.
- van der Stelt MJC, Gerhauser H, Kiel JHA, Ptasinski KJ (2011) Biomass upgrading by torrefaction for the production of biofuels: A review. *Biomass Bioenerg* 35(9): 3748–3762.
- Vehec J (2011) AISI CO₂ Breakthrough Program—A Technology-driven Approach to Climate Change. *Metec InSteelCon 2011, Proc 1st International Conference on Energy Efficiency and CO₂ Reduction in the Steel Industry*, Düsseldorf, Germany.
- Wiedmann T & Minx J (2008) A Definition of 'Carbon Footprint'. In: C.C. Pertsova, *Ecological Economics Research Trends: Chapter 1*, pp. 1-11, Nova Science Publishers, Hauppauge NY, USA. https://www.novapublishers.com/catalog/product_info.php?products_id=5999. Cited 2014/14/1.
- Wijayanta AT, Alam MS, Nakaso K, Fukai J, Kunimoto K & Shimizu M (2014) Combustibility of biochar injected into the raceway of a blast furnace. *Fuel Process Technol* 117: 53–59.
- Wiklund CM, Pettersson F & Saxén H (2012) Optimal Resource Allocation in Integrated Steelmaking with Biomass as Auxiliary Reductant in the Blast Furnace. *ISIJ Int* 52(1): 35–44.
- Wiklund CM, Pettersson F & Saxén H (2013) Optimization of a Steel Plant with Multiple Blast Furnaces Under Biomass Injection. *Metall Mater Trans B* 44(2): 447–458.
- World Steel Association (2014a) World crude steel output increases by 3.5% in 2013, press release. URI: <https://www.worldsteel.org/media-centre/press-releases/2014/World-crude-steel-output-increases-by-3-5--in-2013.html>. Cited 2014/3/10.
- World Steel Association (2014b) World steel in figures 2014. URI: <http://www.worldsteel.org/dms/internetDocumentList/bookshop/World-Steel-in-Figures-2014/document/World%20Steel%20in%20Figures%202014%20Final.pdf>. Cited 2014/9/24.
- World Steel Association (2011) Fact Sheet, Steel and raw materials, May 2011. URI: <http://www.worldsteel.org/publications/fact-sheets.html>. Cited 2013/12/16.
- Worrell E, Neelis M, Price L, Galitsky C & Zhou N (2008) World Best Practice Energy Intensity Values for Selected Industrial Sectors. Lawrence Berkeley National Laboratory, Berkeley, CA pp. 5–17.

- Worrell E, Blinde P, Neelis M, Blomen E & Masanet E (2010) Energy Efficiency Improvement and Cost Saving Opportunities for the U.S. Iron and Steel Industry—An ENERGY STAR® Guide for Energy and Plant Managers. Ernest Orlando Lawrence Berkeley National Laboratory. URI: www.energystar.gov/ia/business/industry/Iron_Steel_Guide.pdf. Cited 2014/14/1.
- Xu C & Cang DQ (2010) A brief overview of low CO₂ emission technologies for iron and steelmaking. *J Iron Steel Res Int* 17(3): 1–7.
- Yellishetty M, Mudd GN, Ranjith PG, Tharumarajah A (2011) Environmental life-cycle comparisons of steel production and recycling: sustainability issues, problems and prospects. *Environ Sci Pol* 14(6): 650–663.
- Ylitalo E (2013) (ed) *Metsätalastollinen vuosikirja 2013*. Metsäntutkimuslaitos, Vammala, Finland.
- Yokoyama H, Higuchi K, Ito T & Oshio A (2012) Decrease in Carbon Consumption of a Commercial Blast Furnace by Using Carbon Composite Iron Ore. *ISIJ Int* 52(11): 2000–2006.
- Zhang Q, Chang J, Wang T & Xu Y (2007) Review of biomass pyrolysis oil properties and upgrading research. *Energy Convers Manag* 48(1): 87–92.
- Zhang X, Stromman AH, Solli C & Hertwich EG (2008) Model-Centered Approach to Early Planning and Design of an Eco-Industrial Park around an Oil Refinery. *Environ Sci Technol* 42: 4958–4963.
- Zhang W (2010) Automotive fuels from biomass via gasification. *Fuel Proc Technol* 91(8): 866–876.
- Zhang H, Dong L, Li H, Fujita T, Ohnishi S & Tang Q (2013) Analysis of low-carbon industrial symbiosis technology for carbon mitigation in a Chinese iron/steel industrial park: A case study with carbon flow analysis. *Energy Policy* 61: 1400–1411.
- Zhou L, Hu S, Li Y, Jin Y & Zhang X (2012) Modeling and Optimization of a Coal-Chemical Eco-industrial System in China. *J Ind Ecol* 16(1): 105–118.
- Ziebig A, Lambert K & Szega M (2008) Energy analysis of a blast-furnace system operating with the Corex process and CO₂ removal. *Energy* 33(2): 199–205.

Original publications

- I Suopajarvi H, Fabritius T (2012) Effects of biomass use in integrated steel plant – gate-to-gate life cycle inventory method. *ISIJ Int* 52(5):779–787.
- II Suopajarvi H, Pongrácz E, Fabritius T (2013) The potential of using biomass-based reducing agents in the blast furnace: A review of thermochemical conversion technologies and assessments related to sustainability. *Renew Sustain Energy Rev* 25:511–528.
- III Suopajarvi H, Fabritius T (2013) Towards More Sustainable Ironmaking – An Analysis of Energy Wood Availability in Finland and the Economics of Charcoal Production. *Sustainability* 5(3): 1188–1207.
- IV Suopajarvi H, Pongrácz E, Fabritius T (2014) Bioreducer use in Finnish blast furnace ironmaking – Analysis of CO₂ emission reduction potential and mitigation cost. *Appl Energy* 124: 82–93.

Reprinted with permission from ISIJ International (I), Elsevier (II, IV) and MDPI (III).

Original publications are not included in the electronic version of the dissertation.

497. Myllylä, Teemu (2014) Multimodal biomedical measurement methods to study brain functions simultaneously with functional magnetic resonance imaging
498. Tamminen, Satu (2014) Modelling the rejection probability of a quality test consisting of multiple measurements
499. Tuovinen, Lauri (2014) From machine learning to learning with machines : remodeling the knowledge discovery process
500. Hosio, Simo (2014) Leveraging Social Networking Services on Multipurpose Public Displays
501. Ohenoja, Katja (2014) Particle size distribution and suspension stability in aqueous submicron grinding of CaCO_3 and TiO_2
502. Puustinen, Jarkko (2014) Phase structure and surface morphology effects on the optical properties of nanocrystalline PZT thin films
503. Tuhkala, Marko (2014) Dielectric characterization of powdery substances using an indirectly coupled open-ended coaxial cavity resonator
504. Rezazadegan Tavakoli, Hamed (2014) Visual saliency and eye movement : modeling and applications
505. Tuovinen, Tommi (2014) Operation of IR-UWB WBAN antennas close to human tissues
506. Vasikainen, Soili (2014) Performance management of the university education process
507. Jurmu, Marko (2014) Towards engaging multipurpose public displays : design space and case studies
508. Namal, Suneth (2014) Enhanced communication security and mobility management in small-cell networks
509. Huang, Xiaohua (2014) Methods for facial expression recognition with applications in challenging situations
510. Ala-aho, Pertti (2014) Groundwater-surface water interactions in esker aquifers : from field measurements to fully integrated numerical modelling
511. Torabi Haghighi, Ali (2014) Analysis of lake and river flow regime alteration to assess impacts of hydraulic structures
512. Bordallo López, Miguel (2014) Designing for energy-efficient vision-based interactivity on mobile devices

Book orders:

Granum: Virtual book store

<http://granum.uta.fi/granum/>

UNIVERSITY OF OULU P.O. Box 8000 FI-90014 UNIVERSITY OF OULU FINLAND

ACTA UNIVERSITATIS OULUENSIS

S E R I E S E D I T O R S

A SCIENTIAE RERUM NATURALIUM

Professor Esa Hohtola

B HUMANIORA

University Lecturer Santeri Palviainen

C TECHNICA

Postdoctoral research fellow Sanna Taskila

D MEDICA

Professor Olli Vuolteenaho

E SCIENTIAE RERUM SOCIALIUM

University Lecturer Veli-Matti Ulvinen

F SCRIPTA ACADEMICA

Director Sinikka Eskelinen

G OECONOMICA

Professor Jari Juga

EDITOR IN CHIEF

Professor Olli Vuolteenaho

PUBLICATIONS EDITOR

Publications Editor Kirsti Nurkkala

ISBN 978-952-62-0705-6 (Paperback)

ISBN 978-952-62-0706-3 (PDF)

ISSN 0355-3213 (Print)

ISSN 1796-2226 (Online)

UNIVERSITY of OULU
OULUN YLIOPISTO

