The Feasibility of Torrefaction for the Co-Firing of Wood in Pulverised-Fuel Boilers

David Agar

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David Agar
David Agar  
M.Sc. 2004

Laboratory of Inorganic Chemistry  
Department of Chemical Engineering  
Åbo Akademi University

Supervisors  
Professor Mikko Hupa  
Åbo Akademi University

Professor Margareta Wihersaari  
Åbo Akademi University

D.Sc. Nikolai DeMartini  
Åbo Akademi University

Opponent and reviewer  
Associate Professor Peter Arendt Jensen  
Technical University of Denmark

Reviewer  
Professor Risto Soukka  
Lappeenranta University of Technology

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This was the first of many delightful days never to be forgotten

- Charles Darwin, The Voyage of the Beagle (1845)
Preface

The work embodied in this thesis was carried out in the Department of Chemistry at the University of Jyväskylä (JYU) and at the Laboratory of Inorganic Chemistry at the Department of Chemical Engineering at Åbo Akademi University. A vital part of this research evolved from collaboration with the Spanish National Renewable Energy Centre (CENER) and the Finnish Technical Research Centre (VTT). The main funding sources for this research were The Fortum Foundation, The Johan Gadolin Scholarship Programme, Åbo Akademi University and the University of Jyväskylä. Secondary funding was provided by the Finnish Doctoral Programme in Environmental Science and Technology.

I would like to thank my supervisors for their guidance and support. This journey all started with a telephone call to Professor Margareta Wihersaari in December 2008. Since that day, she has been an endless source of encouragement and support. Through Margareta I was fortunate enough to have Professor Mikko Hupa as a supervisor for the second half of the research period. I am extremely grateful to him for the pleasant discussions we shared on the nature and direction of the work. His creative guidance has enabled me to narrow the focus of the research. I am indebted to D.Sc. Nikolai DeMartini for his detailed comments and suggestions regarding the thesis, the manuscripts for Paper IV and V and for many other practical matters. His timely correspondence and flexibility have made working at a distance possible.

Though my time at the Process Chemistry Centre was limited, I thank everyone at OOK for the amiable atmosphere. Additionally, I thank the following individuals for their collaboration and contributions: Peter Bachman, Luis Bezerra, Niklas Vähä-Savo, Tooran Khazraie, Johan Werkelin, Emil Vainio, Magnus Perander and Oskar Karlström.

Many colleagues at JYU have influenced this research in some way over the last six years. A warm thanks to Leena Karjalainen, Miia Jämsén, Jussi Maunukseta, Jan Kunnas, Esa Alakoski, Arjo Heisola, Prasad Kaparaju, Marina Himanen, Heli Ratia, Sahoo Tarini, Noora Nuutinen and Jenni Pällysaho. A special thanks to Professors Jan
I would like to thank the personnel of the Biomass Energy Department at CENER for their kind hospitality during my two-month stay in Sarriguren. It was a pleasure to work with Inés, Javier, David, Edwardo and especially Blanca and Goizeder who also introduced me to the beautiful Navarre region. I also thank my fellow explorer and connoisseur of fine bread, Gonzalo, for opening his home to me and guiding me across so many wonderful landscapes – may our paths cross again soon!

Lastly, I would like to thank my parents for their continued support and Marta and Axel for the loving encouragement, happiness and laughter.

Jyväskylä, November 2015

David Agar
Abstract

Torrefaction is the partial pyrolysis of wood characterised by thermal degradation of predominantly hemicellulose under inert atmosphere. Torrefaction can be likened to coffee roasting but with wood in place of beans. This relatively new process concept makes wood more like coal. Torrefaction has attracted interest because it potentially enables higher rates of co-firing in existing pulverised-coal power plants and hence greater net CO$_2$ emission reductions.

Academic and entrepreneurial interest in torrefaction has skyrocketed in the last decade. Research output has focused on the many aspects of torrefaction – from detailed chemical changes in feedstock to globally-optimised production and supply scenarios with which to sustain EU emission-cutting directives. However, despite its seemingly simple concept, torrefaction has retained a somewhat mysterious standing. Why hasn’t torrefied pellet production become fully commercialised? The question is one of feasibility.

This thesis addresses this question. Herein, the feasibility of torrefaction in co-firing applications is approached from three directions. Firstly, the natural limitations imposed by the structure of wood are assessed. Secondly, the environmental impact of production and use of torrefied fuel is evaluated and thirdly, economic feasibility is assessed based on the state of the art of pellet making. The conclusions reached in these domains are as follows.

Modification of wood’s chemical structure is limited by its naturally existing constituents. Consequently, key properties of wood with regards to its potential as a co-firing fuel have a finite range. The most ideal benefits gained from wood torrefaction cannot all be realised simultaneously in a single process or product. Although torrefaction at elevated pressure may enhance some properties of torrefied wood, high-energy torrefaction yields are achieved at the expense of other key properties such as heating value, grindability, equilibrium moisture content and the ability to pelletise torrefied wood. Moreover,
pelletisation of even moderately torrefied fuels is challenging and achieving a standard level of pellet durability, as required by international standards, is not trivial. Despite a reduced moisture content, brief exposure of torrefied pellets to water from rainfall or emersion results in a high level of moisture retention. Based on the above findings, torrefied pellets are an optimised product.

Assessment of energy and CO$_2$-equivalent emission balance indicates that there is no environmental barrier to production and use of torrefied pellets in co-firing. A long product transport distance, however, is necessary in order for emission benefits to exceed those of conventional pellets. Substantial CO$_2$ emission reductions appear possible with this fuel if laboratory milling results carry over to industrial scales for direct co-firing.

From demonstrated state-of-the-art pellet properties, however, the economic feasibility of torrefied pellet production falls short of conventional pellets primarily due to the larger capital investment required for production. If the capital investment for torrefied pellet production can be reduced significantly or if the pellet-making issues can be resolved, the two production processes could be economically comparable. In this scenario, however, transatlantic shipping distances and a dry fuel are likely necessary for production to be viable.

Based on demonstrated pellet properties to date, environmental aspects and production economics, it is concluded that torrefied pellets do not warrant investment at this time. However, from the presented results, the course of future research in this field is clear.

Keywords: Torrefaction, feasibility, co-firing, pellets, pulverised-fuel, economics, wood, carbon dioxide emissions, pelletisation
Svensk sammanfattning

Torrefiering innebär partiell pyrolys av trä och karaktäriseras av termisk nedbrytning av i synnerhet hemicellulosa i en inert atmosfär. Torrefiering kan liknas vid rostning av kaffebönor men med bönorna utbytta mot trä. Detta relativt nya koncept omvandlar trä till en produkt som påminner om kol. Torrefiering har väckt intresse för materialets egenskaper möjliggör en högre grad av biomaterial vid samförbränning i befintliga kolkraftverk, med lägre CO\textsubscript{2} emissioner som följd.


Den här avhandlingen tar sig ann frågan om genomförbarhet vid samförbränning från tre olika infallsvinklar. Först behandlas de begränsningar som härrör sig till träråvarans struktur, sedan utreds miljöpåverkan av både produktion och användning av torrefierat bränsle och till sist undersöks ekonomiska aspekter kring pelletering med tillgänglig teknik. I det följande presenteras slutsatserna från dessa tre delområden.

Vilka förändringar som kan göras i träets struktur begränsas av materialets kemiska sammansättning. Detta betyder att centrala egenskaper hos trä begränsar den potentiella utnyttjandegraden vid samförbränning. Alla de fördelar som uppnås med torrefiering av trä kan inte samtidigt realiseras i en process eller produkt. Trots att torrefiering vid förhöjt tryck kan ge större energiutbyte och gynna några av egenskaperna hos trä, sker detta på bekostnad av andra
centrala egenskaper så som förhöjt värmeverde, fukthalt, malbarhet och möjligheterna att tillverka pellets av materialet. Redan att tillverka pellets av måttligt torrefierad råvara är en utmaning, och att uppnå den hållbarhet som internationella standarder kräver är inte en helt trivial uppgift. Trots att fukthalten sänkts i torrefierade pellets, resulterar redan en kort tids utsatthet för exempelvis regn i att fukthalten förhöjs avsevärt. Baserat på iakttagelserna ovan kan man konstatera att torrefierade pellets är en optimerad produkt.

Bedömningen av energi- och koldioxidekvivalentbalanser indikerar att det finns inga miljöhinder för en tillverkning och användning av torrefierade pellets vid samförbränning. Långa transportavstånd krävs däremot för att fördelarna med mindre utsläpp för transport av torrefierade pellets ska övervinna de för konventionell pellets. En avsevärd minskning av koldioxidutsläpp kan dock möjligen uppnås med detta bränsle ifall resultaten från malningen i laboratorieskala går att förverkliga i industriell skala vid direkt samförbränning.

Utgående från tillgänglig teknik och demonstrerade pelletegenskaper, förlorar torrefierad pellets ekonomiskt gentemot konventionell pellets, huvudsakligen på grund av högre kapitalkostnader inom produktionen. Ifall uppskattade kapitalkostnader för tillverkningen av torrefierade pellets kunde sänkas märkbart, eller om utmaningarna relaterade till pellettillverkningen kunde lösas, så skulle de två processerna bli ekonomiskt jämförbara. I detta scenario förutsätts ett torrt bränsle och transatlantiska transportavstånd om produktionen skall vara livskraftig.

Baserat på demonstrerade pelletegenskaper, miljöaspekter och produktionskostnader kan man dra slutsatsen att investeringar i pelletproduktion med torrefierad träbränsle inte för närvarande är berättigade.

Sökord: torrefiering, genomförbarhet, samförbränning, pellets, pulvriserat bränsle, ekonomi, trä, koldioxidutsläpp, pellettillverkning
Publications

The following publications are included in this thesis:


IV. Agar, David; Gil, Javier; Sanchez, David; Echeverria, Inés; Wihersaari, Margareta. Torrefied versus conventional pellet production - A comparative study on energy and emission balance based on pilot-plant data and EU sustainability criteria. Applied Energy 138; 621-630 (2015).

V. Agar, David; DeMartini, Nikolai; Hupa, Mikko. The influence of elevated pressure on the torrefaction of wood. Submitted to Energy and Fuels.

VI. Agar, David. Torrefied versus conventional pellet production - A comparative economic analysis based on the state of the art. Submitted to Biomass and Bioenergy.
Author contributions

Paper I: David Agar was the main author of this paper and calculated the mass and energy balance values presented. He arranged and participated in the site visit to the Topell torrefaction plant in Duiven.

Paper II: David Agar was the main author of this paper. He collected the reviewed data from literature, performed the calculations for derived values presented and wrote the analysis and interpretations of the results.

Paper III: David Agar was the main author of this paper and carried out equilibrium moisture content (EMC) measurements using saturated-salt solutions for presented isothermal EMC curves. He performed calculations on pellet properties using experimental data and was the main contributor to analysis and interpretation of the results.

Paper IV: David Agar was the main author of this paper and derived the presented relations for determining physical quantities in production. He calculated the comparative energy and emission balances in addition to other calculations. David Agar was the main contributor to the analysis and interpretation of the results.

Paper V: David Agar was the main author of the paper. He was responsible for the spreadsheet processing of experimental data, performed the calculations and did most of the CHNS analysis on the samples. David Agar was the main contributor to the analysis and interpretations of the results.

Paper VI: David Agar was the sole author of this paper.
Nomenclature

AOR  Angle of repose
ar As received
C  Carbon mass per cent in dry sample (%)
C_D  Depreciation cost, linear (M €)
C_f  Financing cost (M €)
C_t  Future cash flow (M €)
C_s  Scrap value (M €)
C_CO2  Cost of cutting CO_2 through co-firing (€ t^{-1})
C_0  Capital investment (M €)
CFD  Computational fluid dynamics
CRF  Capital recovery factor
DP  Degree of polymerisation
E  CO_2-equivalent emissions from fuel production (g)
E_{el}  Electricity from tonne-of-coal equivalent of pellets (MJ)
EC_{el}  CO_2-equivalent emissions from electricity (g MJ_{el}^{-1})
EC_{F(el)}  CO_2-equivalent fossil fuel comparator 198 (g MJ_{el}^{-1})
EMC  Equilibrium moisture content (%)
e_{ec}  emissions from raw material extraction (g)
e  annualised emissions from carbon stock changes (g)
e_p  emissions from processing (g)
e_d  emissions from transport and distribution (g)
e_u  emissions from the fuel in use (g)
e_{sca}  emission savings from soil carbon accumulation (g)
e_{ccs}  emission savings from carbon capture and storage (g)
e_{ccr}  emission savings from carbon capture and replacement (g)
H  Hydrogen mass per cent in dry sample (%)
HGI  Hardgrove Grindability Index
HV  Heating value (MJ kg^{-1})
HHV  Higher heating value at constant pressure (MJ kg^{-1})
HHV_V  Higher heating value at constant volume (MJ kg^{-1})
HHV_0  Higher heating value of untreated sample (MJ kg^{-1})
HHV_T  Higher heating value of torrefied sample (MJ kg^{-1})
h  Pellet price (€ t^{-1})
i  Rate of financing (unitless)
LHV  Lower heating value (MJ kg^{-1})
LHV_{ar}  lower heating value, as received (GJ t^{-1})
M  Moisture content (%)
MEC  Minimum explosible concentration
m_{CO2}  Mass of CO_2 reductions (t)
m_{pellets}  Mass of pellets to replace tonne-of-coal equivalent energy (t)
N  Nitrogen mass per cent in dry sample (%)
NER Net Energy Ratio
NPV Net present value (€)
n Duration of financing (a)
O Oxygen mass per cent in dry sample (%)
P Production amount (t a⁻¹)
PTGR Pressurised thermogravimetric reactor
q_input Thermal energy equivalent input in fuel production (MJ)
q_output Thermal energy equivalent output of produced fuel (MJ)
RC Annual cash flow (€)
RP Profit before tax
RED Renewable Energy Directive of the European Union
r Internal rate of return
Sₜₐ Emission savings from generated electricity
T Torrefaction temperature (°C)
t Torrefaction time (s)
T₀ Depreciation period (a)
Vₜₒᵣ Volume of torrefied pellets produced (m³)
Vₜₒᵣ Volume of conventional pellets produced (m³)
Yₐ Carbon yield of torrefaction, dry mass basis (%)
Yₑ Energy yield of torrefaction, dry mass basis (%)
Yₘ Solid mass yield of torrefaction, dry mass basis (%)
ΔH Heat of reaction (kJ)
ΔHₜ Relative higher heating value increase (%)
Δh Price of replacing coal with pellets (€)
ρ Bulk density (kg m⁻³)
σ Energy density (MJ m⁻³)
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Publications
1. Introduction

European Union Energy Policy Objectives

The European Union (EU) relies on energy imports for roughly half of its gross inland energy consumption. The EU fuel import dependency was 53.8% in 2011 [1]. EU energy policy has two main objectives: to reduce emissions of greenhouse gases by member states and to provide more secure inland sources of energy.

The adoption of renewable energy (RE) sources is one important mechanism to achieve policy objectives. The EU is committed to increasing the share of renewable energy to 20% by 2020 [2] and to 27% by 2030 with the aim of reducing domestic emissions to 40% of 1990 levels [3].

For many member states the most feasible path to realising significant increases in renewable energy production in this time period is the increased use of biomass fuels through co-combustion at existing power plants. Despite strong political motivation for using these resources in energy production, cost-effective utilisation is usually a prerequisite condition.

Finnish Energy Policy Objectives

With some 75% of the country having forest cover, Finland is a land of forests. Annual wood growth is approximately 104 million cubic metres of which some 68% (2012) is harvested [4]. Extracting this natural resource generates a cascade of material flows and by-products a large portion of which eventually end up as fuel.

Finland has set its own national target of achieving 38% of its energy production from renewables by 2020 [5]. Finnish use of wood-derived fuels is already fairly developed (Figure 1), especially in utilising by-products from its forest industry in heat and power production [6]. EU and Finnish energy policy objectives are sufficient motivation to expand national use of wood fuels.
Fluidised-bed and grate combustion boilers which are common technologies in Finland will play a large role in expanding biomass use. These plants are well-suited to this task but are not the subject of this thesis. Finland also has several coastal pulverised-fuel power plants which burn imported coal. There is a strong political desire to burn wood in these plants and reduce net CO\(_2\) emissions. However, without significant new investments in plant operations, the existing plant design allows co-firing of only trivial amounts of wood (5-10% energy basis). This is primarily due to the differences in energy density and the milling requirements of wood and coal.

Using a thermal treatment process known as torrefaction, a new type of wood pellets (torrefied pellets) can be produced which have properties more like that of coal. The improved milling behaviour of torrefied wood is supposed to allow significantly greater co-firing amounts (perhaps up to 50%) using existing plant design.

Both academic and commercial interest in torrefaction has skyrocketed in the last decade. This is reflected in the output of peer-reviewed journal publications on the topic – seen in Figure 2 as the
number of publication returns from a ScienceDirect\(^1\) search using torrefaction as the search term.

![Graph showing the number of peer-reviewed journal publications between 2005-2014 based on the search term “torrefaction” from ScienceDirect.](image)

Figure 2: The number of peer-reviewed journal publications between 2005-2014 based on the search term “torrefaction” from ScienceDirect.

Much of the interest in torrefaction can be traced back to a pair of technical reports published in 2005 by the Energy Research Centre of The Netherlands (ECN). Although these were not the first studies on torrefaction, they were among the first to suggest the use of torrefied wood in co-firing applications with the aim of reducing net carbon dioxide emissions [7, 8]. Several properties of torrefied wood were presented in the reports. Additionally, a process of combining torrefaction and pelletisation, known as the TOP Process (i.e. **T**orrefaction and **P**elletisation) was described. Conventional wood pellets – produced usually from pelletised sawdust – were compared to their torrefied counterpart. Commercial production of both pellet types was also compared using an economic analysis the results of which concluded that torrefied pellet production is clearly an attractive prospect.

\(^1\) www.sciencedirect.com (2 February 2015)
Using conventional pellets as a basis of comparison with torrefied pellets is sensible because the former industry is already globally well established. Global pellet production is estimated (2012) at 15 million tonnes annually [9] with imported amounts to the EU reaching 4.3 million tonnes in 2012 [10].

The basic details of how torrefied pellet production differs from conventional wood pellets are illustrated in Figure 4, in which three different production processes are shown. The first process is wood pellet production from dry saw dust. The second is the same process but using wet saw dust. The dry mass of saw dust feedstock is the same for all three processes. The third process is based on the ECN TOP process. Mass and energy flows are given in the figure with total thermal energy content of produced pellets (in Terawatt hours) on the right of the figure.
Figure 4. A comparison of three processes: conventional pellet production from sawdust, conventional pellets produced from wet sawdust and TOP pellets produced from wet sawdust. The dry mass of sawdust feedstock is the same for all three processes (Figure from Paper I based on data from [7] and [11]).

Comparing the two latter processes, the TOP process has several expected benefits which are itemised in Table 1. The produced pellets, known as TOP pellets, have a total energy content of 0.32 TWh. This is less than that of conventional pellets (0.36 TWh) because of the mass loss from feedstock during torrefaction. Recycling of torrefaction gases, however, allows a lower heat input. TOP pellets also have a lower moisture content which equates to greater net heating value. Overall, the TOP process produces a more concentrated form of fuel –
enabling more energy to be packed into a cubic metre of pellets. This has clear benefits in transporting and storage of the fuel.

Taken together with other benefits, the TOP process represents a shift towards greater sustainability compared to conventional pellets because it enables greater emission reductions using existing coal power plant technology.

Table 1: The expected benefits with regard to sustainability of torrefaction technology for fuel-pellet production (Paper I).

<table>
<thead>
<tr>
<th>Feature of Torrefaction Technology</th>
<th>Expected Benefit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Improved milling behaviour</td>
<td>+ displacement of fossil coal use + reduction of electricity in production</td>
</tr>
<tr>
<td>Expansion of feedstock choice</td>
<td>+ increase utilisation of biomass + less dependence on conventional feedstock + improved investment security of plant</td>
</tr>
<tr>
<td>Increased energy density of pellets</td>
<td>+ less transport fuel per energy unit + extend feasible transport distances</td>
</tr>
<tr>
<td>Enhanced storage properties</td>
<td>+ reduced infrastructure investment + less losses due to storage</td>
</tr>
<tr>
<td>Drier less volatile fuel</td>
<td>+ greater combustion stability/ efficiency</td>
</tr>
<tr>
<td>Recycling of torrefaction gases</td>
<td>+ heating energy derived from feedstock</td>
</tr>
</tbody>
</table>

As a potential mechanism for large CO₂ emission reductions, torrefaction has attracted a great deal of interest. Research output has focused on the many physical and chemical details of torrefaction. Meanwhile, several companies have been developing the technology, for example, Topell Energy of the Netherlands constructed the world’s first large-scale commercial plant in 2011 (Figure 3). Despite these activities, there has been relatively little academic focus on the economic and environmental benefits of torrefaction. This is a technology trying to emerge and such studies are an important part of this process.
2. Objectives of the Thesis

The purpose of this thesis is to address the following question: Is production of pellets from torrefied wood for co-firing in existing pulverised-coal power plants a venture worth pursuing?

This is a question of feasibility, which herein is defined as a three-component measure:

- Technical feasibility of wood torrefaction (chemical and physical limitations)
- Environmental feasibility (energy and greenhouse gas emission balance)
- Economic feasibility (profitability analysis)

In order to evaluate each of these measures, the following objectives have been set:

- Describe observed chemical and physical changes occurring through torrefaction of wood.
- Identify the characteristics of torrefied wood – so-called key properties – which influence the success of co-firing at pulverised-fuel power plants.
- Quantify the range of these key properties and benefits gained through torrefaction.
- Compare the CO$_2$ footprint of torrefied pellet production to conventional wood pellets.
- Evaluate the economic potential of torrefied fuel production based on state-of-the-art.

The scope of this research is necessarily wide-ranging. It draws extensively on scientific literature while also contributing to the current body of knowledge regarding torrefaction and torrefied pellet production. The thesis makes the following assumptions with regard to scope:

- The focus is on torrefaction of woody biomass from temperate tree species
- It considers torrefaction and pelletisation of wood (i.e. torrefied pellet production)
- Only so-called “dry torrefaction” is considered
Only stand-alone production is considered (i.e. thermal integration of torrefaction or torrefied pellet production with other industrial processes is not considered).

Only existing pulverised-coal power plants are considered for co-firing.

This work is not a technological evaluation of any one torrefaction reactor technology.

This work identifies limits in capital investment amounts required for feasible torrefied pellet production.

It is desirable to reduce net CO₂ emissions from the energy sector.

This thesis is based on six publications referred to with the Roman numeral I through VI. **Paper I** is a qualitative introduction to the subject of torrefaction and accordingly, its contents are summarised in Chapter 1. In **Paper II**, three key physical properties of torrefied wood, regarding its use in co-firing in pulverised-fuel boilers, are identified. Data on these properties are reviewed from recent literature and compared to the expected benefits of torrefaction as described in **Paper I**. Content from **Paper II** has been expanded on and used in Chapter 4. Experimentally determined physical properties of six different types of torrefied pellets were determined in **Paper III**. Selected parts of this **Paper III** appear in Chapters 1, 4 and 5 and the main results are discussed in Chapter 6. **Paper IV** utilises data from a torrefaction pilot plant in order to compare the energy and emission balance of production with conventional pellets. The pilot plant and its torrefaction concept are described in Chapter 4. The results from **Paper IV** are presented in Chapter 6. **Paper V** describes a study on how elevated pressure influences the torrefaction of wood. The apparatus in this work is described in Chapter 5 and the results are presented in Chapter 6. **Paper VI** utilises pellet property data from **Paper III** and torrefaction process data from **Paper IV** in an economic evaluation of torrefied pellet production. Methods and results from **Paper VI** are presented in Chapter 5 and Chapter 6 respectively.
3. Background

This chapter provides the necessary background information relevant to the thesis topic and objectives. First some basic aspects of fuels and carbon dioxide emissions are described followed by an overview of relevant lignocellulosic resources in Finland. More detailed information is then given regarding the natural structure of wood and the general properties of wood fuels. Lastly, coal and pulverised-coal boilers are described and the concept of co-firing wood using this technology is explained.

3.1 Combustion of Wood and CO₂ Emissions

In a combustion process, the following are the main combustion reactions and their approximate heats of reaction which take place between the fuel and oxygen [12].

\[ C(s) + O_2(g) \rightarrow CO_2(g) \quad \Delta H = -393 \text{ kJ} \]
\[ CO(g) + \frac{1}{2} O_2(g) \rightarrow CO_2(g) \quad \Delta H = -283 \text{ kJ} \]
\[ 2H(g) + \frac{1}{2} O_2(g) \rightarrow H_2O(g) \quad \Delta H = -242 \text{ kJ} \]

The products of these reactions are heat, gaseous carbon dioxide and water vapour. The heating value of a fuel depends on the heat of reaction and, for wood and coal, mostly determined by the amount of carbon in the fuel. Furthermore, the quality of a fuel is commonly described through proximate analysis which expresses the per cent amounts of moisture, ash, volatile matter and fixed carbon (or char) in the fuel. Fuels with high fixed carbon content are difficult to ignite due to low volatility. Fuels with a high volatile matter content, however, ignite relatively easily but burn out more quickly. Wood and coal are good examples of two fuels at these aforementioned extremes. Dry wood can have fixed carbon content of some 15% and volatile content of 85% while the amounts in hard coal can be the reverse of these values.
Fossil fuels, such as coal, petroleum and natural gas, are large stores of carbon formed through decomposition of various organisms over millions of years. This carbon was absorbed from the atmosphere of the past. Consequently, fossil fuel use in energy production today is the main source of anthropogenic carbon dioxide emissions [13].

Carbon dioxide emissions from the combustion of wood releases the carbon dioxide absorbed by the tree during its growth. Although there are also some emissions associated with wood fuel production [14-17], sustainably grown wood is considered to be mostly carbon-neutral. Therefore, replacing fossil fuels with wood reduces net anthropogenic CO₂ emissions to the atmosphere.

### 3.2 Lignocellulosic Resources

**Forest Chips**

Forest chips is a general term for chipped or crushed woody material whose raw material originates from forest stands [18]. Size reduction is accomplished with specialised machinery and chip sizes generally range from one to three centimetres in length. Chips are further classified according to which part of the tree is used. For example, log chips are made from stem wood without branches and with or without bark. Whole-tree chips are made from the entire above-ground tree including stem, bark, branches, needles or leaves. In addition to these types, there are also stump chips, chips from small-sized trees removed during forest thinning operations and logging residue chips.

Logging residue chips are produced from branches, tree tops and undergrowth when final timber harvest operations are carried out. Extractable forest residues represent the largest single potential for forest-based fuels in Finland, estimated at 10 million cubic meters annually - or about four million tonnes annually [19]. This figure is based on the assumption that residues amount to 15% of stem wood mass for coniferous trees and 10% for deciduous. Since logging residues are a by-product of a commercial timber harvest, they are mostly composed of the common commercial species (spruce, pine and birch).
Logging residues are already widely harvested and used in Nordic countries at grate-fired and fluidised-bed power plants both of which are designed for heterogeneous wet solid fuels. Additionally, studies suggest that the utilisation of residues from Nordic forests is more environmentally beneficial than leaving them on the forest floor and can lead to maximum benefit when offsetting fossil coal use at power plants [20, 21].

Conventional Wood Pellet Production

Wood pellets are a densified fuel produced by compressing sawdust and cutter shavings (predominantly from spruce and pine species [11]) into cylindrical pellets having standard diameters of 6 and 8 mm. Due to their low moisture content and packaging wood pellets have a LHV range of 16.7-17.9 MJ kg\(^{-1}\) [22]. Global production of wood pellets has grown rapidly since the turn of the century and is estimated at 15 million tonnes annually (2010) [9]. The raw materials used to produce pellets are most often by-products of large wood-processing industries. Consequently, wood pellet production and feedstock availability has traditionally relied mainly on the productivity of the wood-processing sector. Integration of pellet making at wood-processing sites is therefore a common venture because by-products can be utilised on-site with minimal transport and a non-specialised labour force. Strong growth in pellet production has increased interest in other feedstock material such as tree bark and logging residues [23, 24] although there have been some challenges in adapting these [25].

Despite on-going scientific research in this sphere, pellet making is still more of an art than it is an exact science. Pellet producers use trial and error methods to determine successful procedures and recipes for production. To make full use of the feedstock available to them, producers may modify mixtures according to feedstock type and seasoning, wood species and moisture content to ensure that their product complies with production standards (EN 14961-2).

At industrial scales, wood pellets are commonly produced using ring-die pellet presses. Additives are not usually required to achieve good pellets because the lignin content of the wood feedstock provides adhesion. When the moisture content of the feedstock is optimal (15-20\%) compression and extrusion through the die heats the feedstock leading to the so-called softening of lignin [26]. Exceeding the glass

11
transition temperature for a sufficient portion of the polymers in
wood is necessary in order to achieve a sufficient bonding area [27].
Steam can also be added to the mixture to enhance this effect. Freshly
pressed pellets are consequently hot (90°C) and proper cooling and re-
hardening of lignin plays a critical role in pellet stability and strength
[11]. The moisture content of feedstock and the length of die channels
are two parameters that can be varied in pellet making to achieve
good quality pellets [11, 28]. During steady-state operation, die
temperature and compressional force with tend to some equilibrium
values which cannot be controlled in commercial ring-die pellet
presses [29].

Binding agents are sometimes added to feedstock to improve the
durability of wood pellets. Examples of common agents include
starch, sulfonated lignin, flour [30] and caustic soda [31]. European
quality standards (EN 14961-2) permit a maximum additive amount
of two per cent, on a mass basis, for pellets used in heating
applications. Although binding agents are often by-products of other
industrial processes, their use adds to the production costs of wood
pellets [32].

Wood pellets are packaged in large sacks (1-1.5 m$^3$) or sold in bulk to
large-scale consumers. The transport of pellets is done using
conventional covered lorries and those with pneumatic unloading.
Being susceptible to moisture wood pellets are stored in dry locations
which are protected from rainfall and rapid fluctuations in humidly
and temperature. The moisture content range of Finnish pellets from
the manufacturer is 7-10% [11].

Pellet production is most cost-effective when feedstock is cheap and
requires no drying. However, drying costs are minimised when heat
can be supplied through the combustion of wet feedstock [32, 33].
The economy of scale favours large pellet plants in the case of stand-
alone production.

Environmental Impact of Wood Pellets and Chips

Studies of on the environmental impact of wood pellets have looked
at energy input and CO$_2$-equivalent emission [34]. Primary energy
input for Canadian wood pellets delivered to Sweden is 7.2 GJ t$^{-1}$ and
CO$_2$-equivalent emissions from their production and transport is 29 g
MJ\(^{-1}\). These figures assume feedstock material is used as a fuel in the drying process. Over one third of input energy from Canadian pellets stems from transatlantic shipping via the Panama Canal to Stockholm and in total 39% of the energy content of these pellets is consumed in their manufacture and transport. Based on these values, pellets use in Swedish space heating would realise an emissions saving of about 60% compared to coal. This savings assumes a boiler efficiency of 80%.

Upgrading wood into pellets requires greater energy input than producing wood chips. For comparison, Swedish production of logging residue chips is found to have an energy input range of only 0.4-0.5 GJ t\(^{-1}\) [35]. Additionally, chip production from loose (non-bundled) residue collection required the lowest primary energy input (21-27 kJ MJ\(^{-1}\)) of several studied production scenarios and generated the least amount of CO\(_2\)-equivalent air emissions (1.5-1.9 g MJ\(^{-1}\)). The above values assume that the full lower heating value (LHV) can be realised in combustion. This is often not the case since residue chips are not typically dried like the feedstock used to make wood pellets.

When considering the difference between wood pellets and logging residue chip production it is noted that the pellet production study takes into account emissions and energy input from the final felling of timber from which pellet feedstock is derived (including the hauling of stem wood a distance of 110 km) whereas the above residue chip production does not include this operation [35].

3.3 Chemical and Physical Structure of Wood

Wood is a naturally complex material composed of cells whose continual division process results in the growth of the tree. The physical and chemical properties of wood depend strongly on the species but significant inter-species differences also exist due to growing location, available moisture, soil type and nutrients. Two wood classes are commonly distinguished by the name hardwood (Gymnospermae) and softwood (Angiospermae). Moreover, wood properties also vary between different parts of the same tree due to differentiated cell structure in the formation of heartwood, stem, cambium, bark, root and crown [36].
Wood is composed of cellulose, polyoses (hemicelluloses), lignin, extractives and minerals (Figure 5). Cellulose, hemicellulose and lignin are polymers of graduated complexities – super molecules formed from long chains of hydrocarbon molecules. The number of molecules in the chain is known as the degree of polymerisation (DP). The molecular structure of wood can be visualised as a skeleton of cellulose supported by a hemicellulose matrix and glued together with coatings of lignin which provide mechanical strength to wood.

Cellulose

Celluloses are homopolysaccharides composed of repeating β-D-glucopyranose units. Wood cellulose has a degree of polymerisation in the range of 10 000. Temperate hardwood species have a cellulose content of 38-51 % whereas for softwoods the range is 33-42 % [36].

Hemicellulose

Hemicelluloses are heteropolysaccharides whose branched polymers have a relatively low molecular weight and DP in the range of 80-425 [37]. Hemicelluloses can be classified further to be of five main types (Figure 5) whose relative amounts vary between hard and softwoods and to a lesser extent between species. The five types are xylans, mannans, glucans, galactans and pectins [38]. The hemicellulose content of the tree species presented in Table 2 is 26-33% for softwoods and 19-35% for hardwoods. The main hemicellulose of hardwoods is glucuronoxylan forming 15-30 % of the wood with glucomannan (mannans) forming 2-5% [36]. In softwoods however mannans are the primary hemicellulose whose fraction in temperate species has a range of 14-20% followed by arabinoglucuronoxylan fraction of 5-10% [36]. For simplicity glucuronoxylans and arabinoglucuronoxylan are often labelled as xylans and glucomannan and galactoglucomannans as mannans.

Lignin

Lignin has a DP of approximately 15 000 [36, 38]. The lignin content of temperate wood species has a range of 21-32% in which softwoods have the upper end of this range and hardwoods the lower end [36]. Hardwood species contain a greater portion of cellulose and
hemicellulose but less lignin and extractives than do softwoods (Table 2).

Extractives

The organic low-molecular-weight matter in wood can be extracted using solvents. Therefore these compounds are called extractives. Non-polar solvents yield lipophilic while polar solvents yield hydrophilic extractives respectively. Extractives are concentrated in resin canals (oleoresin), parenchyma cells (fats and waxes) and in heartwood (phenols). These different chemical compounds make up roughly 1 to 5% of the wood [36].

Minerals

The small amounts of metals and elements such as phosphorus, silicon in wood remain as ash after incineration at high temperature. The ash content of temperate wood has a range of 0.2-0.5% and the ash consists mainly of inorganic compounds of potassium, calcium and magnesium [38].

Figure 5. Classification of wood contents based on analysis showing hemicelluloses (adapted from [38]).
Table 2. Chemical composition of selected temperate wood species (adapted from [36])

<table>
<thead>
<tr>
<th>Common name</th>
<th>extractives</th>
<th>lignin</th>
<th>cellulose</th>
<th>mannan</th>
<th>xylose</th>
<th>other polysaccharides</th>
<th>residual constituents</th>
<th>total hemicellulose</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Softwoods</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Abies balsamea</td>
<td>Balsam fir</td>
<td>2.7</td>
<td>29.1</td>
<td>38.8</td>
<td>17.4</td>
<td>8.4</td>
<td>2.7</td>
<td>0.9</td>
</tr>
<tr>
<td>Pseudotsuga menziesii</td>
<td>Douglas fir</td>
<td>5.3</td>
<td>29.3</td>
<td>38.8</td>
<td>17.5</td>
<td>5.4</td>
<td>3.4</td>
<td>0.0</td>
</tr>
<tr>
<td>Tsuga canadensis</td>
<td>Eastern hemlock</td>
<td>3.4</td>
<td>30.5</td>
<td>37.7</td>
<td>18.3</td>
<td>6.5</td>
<td>2.9</td>
<td>0.5</td>
</tr>
<tr>
<td>Juniperus communis</td>
<td>Common juniper</td>
<td>3.2</td>
<td>32.1</td>
<td>33.0</td>
<td>16.4</td>
<td>10.7</td>
<td>3.2</td>
<td>1.4</td>
</tr>
<tr>
<td>Pinus radiata</td>
<td>Monterey pine</td>
<td>1.8</td>
<td>27.4</td>
<td>37.4</td>
<td>20.4</td>
<td>8.5</td>
<td>4.3</td>
<td>0.4</td>
</tr>
<tr>
<td>Pinus sylvestris</td>
<td>Scots pine</td>
<td>3.5</td>
<td>27.7</td>
<td>40.0</td>
<td>16.0</td>
<td>8.9</td>
<td>3.6</td>
<td>0.3</td>
</tr>
<tr>
<td>Picea abies</td>
<td>Norway spruce</td>
<td>1.7</td>
<td>27.4</td>
<td>41.7</td>
<td>16.3</td>
<td>8.6</td>
<td>3.4</td>
<td>0.9</td>
</tr>
<tr>
<td>Picea glauca</td>
<td>White spruce</td>
<td>2.1</td>
<td>27.5</td>
<td>39.5</td>
<td>17.2</td>
<td>10.4</td>
<td>3.0</td>
<td>0.3</td>
</tr>
<tr>
<td>Larix sibirica</td>
<td>Siberian larch</td>
<td>1.8</td>
<td>26.8</td>
<td>41.4</td>
<td>14.1</td>
<td>6.8</td>
<td>8.7</td>
<td>0.4</td>
</tr>
<tr>
<td><strong>Hardwoods</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Acer rubrum</td>
<td>Red maple</td>
<td>3.2</td>
<td>25.4</td>
<td>42.0</td>
<td>3.1</td>
<td>22.1</td>
<td>3.7</td>
<td>0.5</td>
</tr>
<tr>
<td>Acer saccharum</td>
<td>Sugar maple</td>
<td>2.5</td>
<td>25.2</td>
<td>40.7</td>
<td>3.7</td>
<td>23.6</td>
<td>3.5</td>
<td>0.8</td>
</tr>
<tr>
<td>Fagus sylvatica</td>
<td>Common beech</td>
<td>1.2</td>
<td>24.8</td>
<td>39.4</td>
<td>1.3</td>
<td>27.8</td>
<td>4.2</td>
<td>1.3</td>
</tr>
<tr>
<td>Betula verrucosa</td>
<td>Silver birch</td>
<td>3.2</td>
<td>22.0</td>
<td>41.0</td>
<td>2.3</td>
<td>27.5</td>
<td>2.6</td>
<td>1.4</td>
</tr>
<tr>
<td>Betula papyrifera</td>
<td>Paper birch</td>
<td>2.6</td>
<td>21.4</td>
<td>39.4</td>
<td>1.4</td>
<td>29.7</td>
<td>3.4</td>
<td>2.1</td>
</tr>
<tr>
<td>Alnus incana</td>
<td>Grey alder</td>
<td>4.6</td>
<td>24.8</td>
<td>38.3</td>
<td>2.8</td>
<td>25.8</td>
<td>2.3</td>
<td>1.4</td>
</tr>
<tr>
<td>Eucalyptus camaldulensis</td>
<td>River red gum</td>
<td>2.8</td>
<td>31.3</td>
<td>45.0</td>
<td>3.1</td>
<td>14.1</td>
<td>2.0</td>
<td>1.7</td>
</tr>
<tr>
<td>Eucalyptus globulus</td>
<td>Blue gum</td>
<td>1.3</td>
<td>21.9</td>
<td>51.3</td>
<td>1.4</td>
<td>19.9</td>
<td>3.9</td>
<td>0.3</td>
</tr>
<tr>
<td>Gmelina arborea</td>
<td>Yemane</td>
<td>4.6</td>
<td>26.1</td>
<td>47.3</td>
<td>3.2</td>
<td>15.4</td>
<td>2.5</td>
<td>0.9</td>
</tr>
<tr>
<td>Acacia mollissima</td>
<td>Black wattle</td>
<td>1.8</td>
<td>20.8</td>
<td>42.9</td>
<td>2.6</td>
<td>28.2</td>
<td>2.8</td>
<td>0.9</td>
</tr>
<tr>
<td>Ochroma lagopus</td>
<td>Balsa</td>
<td>2.0</td>
<td>21.5</td>
<td>47.7</td>
<td>3.0</td>
<td>21.7</td>
<td>2.9</td>
<td>1.2</td>
</tr>
</tbody>
</table>
3.4 Pulverised-Coal and the Direct-Firing System

Power plants are designed according to the specifications of the fuel to be used. Fluidised-bed and grate combustion boilers are both well-established technologies in the Nordic countries. They are especially attractive because the boiler design allows fuel flexibility and routine multi-fuel operation. For instance, fuels which have high water content, a heterogeneous composition and low heating values can be utilised in fluidised-bed boilers to reach a high portion of the overall fuel mix. It is important to note that burning torrefied fuels in these plants is also fully possible. However, it is not of interest in torrefaction research since such plants have been designed to use untreated or low-value biomass fuels without upgrading.

Coal continues to be the staple source in global electricity production. This is because of its relative abundance, low cost, and widespread geographical distribution. Not surprisingly, many power plants are designed exclusively to burn coal, the majority of the boilers being burner-fired units using pulverised coal. Pulverised-coal boiler technology is almost a century old and remains in widespread use because it is low cost, suitable for many coal types (bituminous and sub-bituminous) and it enables more complete combustion of carbon while making possible fast changes in fuel feeding in response to load variations. Globally, a great number of existing power plants are coal-fired plants which utilise pulverised-fuel technology. In 2006, the EU-25 had 155 GW\textsubscript{e} of coal-fired power plants in operation or in planning stages \cite{39}; the total installed electricity capacity in 2005 (EU-27) was 751 GW\textsubscript{e} \cite{1}. Despite the fact that some 40\% of these were more than 30 years old, the remaining number still represents a huge market for coal in Europe.

Coals are classified by their rank, a classification which includes lignite, sub-bituminous, bituminous and anthracite. The fixed carbon content of coal increases with rank and the amount of volatile matter decreases. U.S. coals, for instance, have a higher heating value range of 16.7-33.2 MJ kg\textsuperscript{-1} \cite{40}. Typical Polish and Russian coals used in Finnish power plants have a volatile matter content of 28-30\%, a moisture content of 8-13\%, 8-12\% ash and a higher heating value of 25 MJ kg\textsuperscript{-1} \cite{41}.
Mined coal comes in different sizes. According to ASTM D 310 these approximate diameters include run-of-mine coal (20 cm), lump coal (13 cm), egg coal (13 × 5 cm) and nut coal (5 × 3 cm). Coal is usually sorted to have uniform particle size before delivery to the power plant where it is often stored in heaps outdoors. The maximum size of coal intended for pulverising mills is about 3 cm. Larger sized particles require crushing at the power plant before pulverisation. Crushing is usually carried out using a ring crusher or hammer mill [40]. Coal is then fed to the pulveriser from a bunker via a feeder.

Several types of pulverisers exist and size reduction is accomplished by impact, attrition, crushing, or combinations of these actions [40]. After pulverisation the fuel is conveyed in pipes pneumatically by a primary air fan to the furnace burners. The dried fuel suspended in primary air is mixed in excess of stoichiometric requirements for combustion (15 to 40% excess air) with hot combustion air from a preheater before entering the burner nozzle and passing through a coal impeller to the furnace. Small ignition jets in the burner ignite the fuel until temperature in the furnace is high enough to sustain the burner flame [40].

Successful combustion in pulverised-fuel systems requires fuel with a large surface-to-volume ratio but also a minimum amount of larger particles to achieve high combustion efficiency [40]. If particles are too large, however, there will not be sufficient time for complete combustion or burn out. Optimal particle size depends on fuel volatility. The higher the fixed carbon content of the coal, the finer the particle size needs to be [40]. Suitable particle size for most coal types is below 100 µm [42]. Typically, 80% of particles pass through a sieve opening of about 75 µm and all other particles pass through a 300 µm sieve opening [40].

The Hardgrove Grindability Index (HGI) is a common measure to describe how difficult it is to grind a given coal to the suitable aforementioned particle size. Although the index does not exhibit any step changes and is of a continuous nature, it is not of a linear function of coal rank [43]. HGI is a useful measure of the capacity of a certain mill. The 100% capacity of a mill is normally set at a HGI value of 50. The higher the HGI of a coal, the easier it is to grind so that a fuel with a low HGI lowers the capacity of a given mill. As the HGI is determined through an empirical test, is not correlated to any physical
property of coal nor is it generally applicable for other fuels such as wood. Energy requirements for pulverising different coals ranks have a range 7-36 kWh t⁻¹ [42].

3.5 Direct Co-Firing of Wood with Coal

Pulverised-fuel plants are not designed to burn biomass but in the simplest case co-firing small amounts of wood is possible if it can be pulverised and handled like coal. In direct co-firing, as opposed to parallel firing or the use of a gasified fuel, the pre-processed wood is mixed with coal upstream of the coal feeders. The blend is then pulverised and the powdered fuel is pneumatically carried to the burners in the furnace.

Co-firing of wood with coal is generally less challenging than agricultural residues or crops grown for use in energy production [44]. Dry wood has a higher heating value range of 18-21 MJ kg⁻¹ [18]. However, wood fuels can have a high moisture content which effectively halves this range. Due to their low bulk density, wood fuels also have a low energy density. Because of plant design these differences in properties affect operation and performance of coal plants using wood [44]. One consequence is that the capacity of coal mills is significantly reduced for wood. For example, when milling wood chips in an industrial vertical roller mill, the maximum capacity (mass throughput) observed is 0.25 t h⁻¹ while that of coal is 2.2 t h⁻¹ [45]. This is a mass-flow difference of almost nine times. Furthermore, the capacity difference in terms of fuel energy throughput is even greater due to the differences in the heating values of wood and coal². This is the primary reason why the direct co-firing of untreated wood fuels is limited to some 5-10% on an energy basis in pulverised-fuel plants [44].

² Even considering relatively dry wood with LHV of 16 MJ kg⁻¹ and typical coal with LHV of 25 MJ kg⁻¹, the fuel energy throughput for wood is (16 MJ kg⁻¹)(250 kg h⁻¹) = 4000 MJ h⁻¹ and for coal (25 MJ kg⁻¹)(2200 kg h⁻¹) = 55000 MJ h⁻¹; a difference of almost 14 times.
Handling and Friability

Before combustion takes place, wood fuels are delivered to the fuel yard of the power plant where they are put in storage. Long-term storage of biomass is usually not practiced because biological activity can lead to decomposition and loss of fuel quality. This is especially true of wet biomass with a small particle size.

Wood is typically in the form of larger chips and, at modern plants, is milled in two stages before use [44]. Due to the mill capacity limitations mentioned above, the milling of wood to particle sizes required for pulverised-coal boilers is energy intensive and is best carried out with mills dedicated for wood.

Frictional forces between particles of fuel have practical implications for conveying and feeding systems. Flow properties of blends of coal and wood depend on the form and size of the particles. For inhomogeneous biomass fuels, the combination of low bulk density and non-uniform particle size can cause fuel to bridge in conveying lines thereby forming plugs of material in pulverisers or channel bends [44].

Sawdust with an average particle size of 100 μm behaves much like coal in wood-coal blends when the wood fraction is 10% or less (mass basis). Whereas wood-coal blends with the same fraction of wood chips (2 x 3 cm$^2$) are more susceptible to flow stoppage [46]. Ball mills fed with wood-coal blends with up to a 9.5% mass fraction of moist sawdust (LHV 13.5 MJ kg$^{-1}$) have been reported to operate reasonably well with only minor effects on resulting coal fineness; there was an upward shift in particle-size distribution – leading to slightly lower (0.3%) boiler efficiency [47]. The same shift in distribution was observed using 2.5% mass fraction of wet (50-65% moisture as received) pine sawdust (LHV 5.5-7 MJ kg$^{-1}$) [48].

Energy requirements for pulverising lignocellulosic biomass have a typical range of 20-150 kWh t$^{-1}$ [42, 49]. The final particle size for coal is much smaller than for wood however and this range of energy requirement takes this into consideration [42].
Risk of Incomplete Combustion

Adequate size reduction is important in pulverised fuel plants because if the carbon in the fuel is not completely oxidised, combustion efficiency is reduced. Having high volatile matter content, particles of wood can be much larger than coal particles and still be sufficiently oxidised in the boiler [18]. Burners designed for wood powders require a particle size below 1 mm [42] and wood particles larger than 3 mm in co-firing applications significantly increase the probability of incomplete combustion [44]. Furthermore, wood char is highly reactive due to its porous structure and presence of catalytic metals such as potassium. Consequently, ensuring complete burn out of wood char in co-firing is normally not an issue even when particles sizes are much larger than coal [50]. Optimal burner design and their location in the boiler, however, may be different for wood fuels.

Influence of Wood Fuels on Emissions of SO\textsubscript{x} and NO\textsubscript{x}

The co-firing of wood also lowers the emissions of sulphur and nitrogen oxides (NO\textsubscript{x}). One source of NO\textsubscript{x} emissions in pulverised-fuel combustion is formation from the nitrogen in the fuel [51]. Fuel blends of wood and coal produce less NO\textsubscript{x} emissions than from coal alone. This reduction stems from the low nitrogen content of wood. Similarly, emissions of sulphur dioxide depend on the sulphur content of the fuel and are reduced with co-firing wood [44].

Ash

Some 80% of total ash material from pulverised-fuel plants is in the form of fly ash collected from the system and the remaining 20% is in bottom ash or slag [44]. Wood-coal blends will generally reduce the amount of fly and bottom ash simply because of the low ash content (less than 1%) of wood. This reduction effect can be large depending on the ash content of the wood species and coal [44]. As a commercial by-product used in concrete production, fly ash from wood-coal combustion is not seen to significantly degrade its usefulness in this application [44, 52, 53].
Wood Pellets in Co-Firing

Since their arrival on the market conventional wood pellets have mostly been used in small-scale domestic heating. The desire to lower CO₂ emissions, however, is leading to their use for co-firing with coal at large pulverised-fuel power plants. Co-firing of wood pellets in Finland has been carried out by blending pellets with coal at the fuel yard after which the fuel mixture (with up to 7% pellets on an energy basis) is pulverised using existing mills [11]. Wood pellets offer improvements over co-firing with wood chips. These include a greater bulk and energy density; and typically lower moisture content and hence higher heating value as received. Studies indicate that the throughput of existing coal mills is also slightly enhanced for pellets compared to wood chips due to the disintegration of pellets back into the particles from which they were formed [45, 54]. The energy required for the milling of wood pellets is estimated to be 40 kWh t⁻¹ and upwards [54].
4. Torrefaction

The purpose of this chapter is to define torrefaction and describe relevant findings from research pertaining to its use as a co-firing fuel. The torrefaction process concept is then illustrated using an existing pilot plant as an example to demonstrate the production of pellets made of torrefied wood. The final section describes key physical properties changes in wood through torrefaction.

The word torrefaction comes from French (torréfaction) meaning to roast and is used in connection with the roasting of coffee beans. In the 1980s, Bourgeois and Doat had an interest in torrefaction as an industrial process for producing a more energy-efficient fuel to replace wood charcoal as well as a suitable reducing agent in metallurgy and gasification fuel [55]. Today, however, interest in torrefied wood stems from its potential as a co-firing fuel – to offset fossil coal use – and mitigate carbon dioxide emissions. Currently several technology developers are working to commercialise torrefied fuel production.

The main idea of torrefaction is simple - the mild roasting of wood in inert atmosphere improves its fuel properties. Energy is required for roasting, but this energy expenditure is justified if handling and combustion properties can be improved significantly. During roasting, part of the wood mass is devolatised (Figure 6). These vapour-phase products contain chemically-bound energy and ideally these can be further utilised through combustion and heat integration in the torrefaction system. The rationale behind torrefaction is that the energy yield of the solid product is greater than the mass yield – the result is an increase in the heating value of the solid.
4.1 Thermal Decomposition of Wood

Torrefaction takes place in the temperature range in which the hemicellulose component of wood has the greatest rate of decomposition. Whereas fast pyrolysis and cellulose reactions at higher temperature have been historically well-studied, there are far fewer detailed investigations of wood torrefaction. Torrefaction is usually carried out under inert atmosphere, although some studies show that low levels of oxygen can be more viable and even beneficial to the process [56, 57].

The nominal decomposition temperature of each wood component correlates, in a very general sense, with its degree of polymerisation. For example, hemicellulose, cellulose and lignin have progressive DP values of 80-425, 10 000 and 15 000 [36].

Thermogravimetric (TG) mass-loss curves of three isolated wood components are shown in figure 7 as a function of time. Each component sample (~6 mg) was heated (10°C min$^{-1}$) in stages (red line, right-hand axis) from room temperature to 290°C in nitrogen (50 ml min$^{-1}$) using a laboratory thermobalance (Perkin Elmer Pyris Diamond TG/DTA). The curves have been normalised to mass
fraction of samples. In the figure, xylan (the principle constituent in hardwood hemicellulose) shows a rapid and early degradation between the 60-70 min. mark where its mass fraction is reduced by 50%. Note, however, that both cellulose and lignin have been significantly decomposed by some 10% and 20% respectively during this time frame.

![Figure 7. TG curve mass loss versus time at 290°C of cellulose (blue), xylan (green) and lignin (violet) (This work).](image)

Torrefaction increases the carbon content and decreases the hydrogen and oxygen content of wood. For example, early results by Girard and Shah from the torrefaction of pine chips over the temperature range of 250-350°C resulted in chars with a carbon content increase from 47 to 66% with increasing temperature, a hydrogen content decrease from 6.3 to 5.2% and an oxygen content decrease from 45 to 27% [58].

Describing the thermal decomposition of wood by the behaviour of its components is an approximation which neglects degrees of interaction. This has been mostly justified in predicting pyrolysis behaviour of wood [59, 60]. Thermal decomposition not only degrades polymer chains, creating species with lower degrees of polymerisation, it also leads to re-polymerisation and formation of complex structure from simpler constituents with mineral matter.
acting as catalysts for these reactions [61, 62]. In the above figure, significant decomposition of cellulose and lignin begins before the target torrefaction temperature is reached. This has also been observed in torrefaction of birch wood where the onset of cellulose and lignin degradation occurs at 255°C and 240°C respectively [63].

Torrefaction temperature and time directly affect mass yield. Yields are also influenced by wood species. Species-dependent differences can partly be explained by the differing proportions of wood components in addition to composition of hemicellulose. Of the hemicelluloses, xylans are more reactive than mannans. Xylans are the main hemicellulose type in hardwoods, while mannans are the main type in softwoods. This is one factor in different mass-loss rates observed for hardwoods and softwoods [64]. The concentration of certain metals bound to organic sites in the material also has an influence. Potassium, sodium and magnesium, for instance, have been shown to affect the rate of mass loss and char formation during the torrefaction of wood [61, 62, 65]. This finding supports the view that the thermal decomposition behaviour of wood is more than the sum of its components.

The yield of solid product decreases with torrefaction temperature and time. The vapour-phase products, as a consequence, increase and can be separated into condensable and non-condensable fractions. The non-condensable fraction consists of low molecular weight gases, mostly carbon dioxide (CO₂) and carbon monoxide (CO), but also small amounts of hydrogen (H₂) and methane (CH₄) [66, 67]. In addition to water, the condensable fractions include acetic acid (CH₃COOH), methanol (CH₃OH), lactic acid (C₂H₄OHCOOH), formic acid (CH₂O₂), furfural and hydroxyl acetone [66, 67].

Pyrolysis of Untreated Versus Torrefied Fuels

Pyrolysis curves of untreated (blue) and torrefied (red) Finnish logging residues are shown in Figure 8. The milled (75-80% of particles less than 0.5 mm) samples (~8 mg) were heated (10°C min⁻¹) in nitrogen (50 ml min⁻¹) using a laboratory thermobalance (Perkin Elmer Pyris Diamond TG/DTA). The curves are derivatives of the mass fraction of the samples. Torrefaction conditions of the latter sample were 290°C and a torrefaction time of 30 minutes using the same thermobalance.
The differences between the two curves in the figure demonstrate the changes brought about through torrefaction. During the initial heating of the samples, moisture is removed corresponding to the two hillocks on the left of the figure. The torrefied sample, having lower equilibrium moisture content, has less moisture to lose as reflected by the relative heights of the hillocks. Secondly, as temperature exceeds 200°C, the more reactive components in the untreated sample begin to devolatise. Because these components have already been degraded from the torrefied sample, its curve is essentially non-reactive until about 250°C, after which point its rate of increase surpasses the untreated sample. Consequently, the peak reactivity of the torrefied sample occurs at lower temperature, to the left of the other peak; a shift of some 30°C to the left. The shoulder on the left of the blue curve is attributed to hemicellulose fraction of the sample while the peak itself is attributed to cellulose [44]. Thirdly, after peak devolatisation has occurred, the rates are essentially constant – indicated that the remaining sample fraction (proportionally greater for the torrefied sample) is mostly char. This portion of the curve is associated with lignin which mainly degrades at high temperature [44].

Figure 8. Differential of mass-loss curves during pyrolysis of untreated and torrefied (290°C, 30 min.) Finnish logging residues (This work).
Summarising the behaviour shown in the figure, the torrefied sample is a less volatile fuel, having greater fixed carbon content: the fuel properties of the sample have moved a step closer to those of coal.

4.2 Torrefaction Process Concepts

From a feasibility perspective, an ideal torrefaction process would operate effectively, have a low cost and generate minimal environmental impacts. Worldwide, there are currently many different reactor technologies and processes used in pilot-plants [68]. An ideal reactor would maximise transfer of heat from source to feedstock within a narrow range of temperature [69].

Heat transfer can be realised using a thermal gas or liquid and recently microwave radiation is being investigated [70-72]. Hot fluids can be in direct contact with the feedstock or indirectly with heat transfer occurring through the walls and components of the reactor vessel. Direct contact enables a higher rate of heat transfer but means that the feedstock and thermal fluid streams are combined. Indirect heating allows separation of these two streams. Torrefaction can be a batch process or be of a continuous nature. Continuous-flow reactor systems are inherently more complex but have advantages in industrial processes.

The vapour-phase reaction products from torrefaction contain energy both as a high-temperature gas and as non-oxidised products of gases such as CO, H₂ and CH₄. The concentration of these gases is small, however. Many torrefaction processes aim to utilise this energy through heat integration. The concept of autothermal operation has been put forward as the process operating conditions when the total heat demand of drying and torrefaction of feedstock is supplied by the energy content of the vapour-phase products [8]. The effectiveness of heat recovery, therefore, is a critical factor in the thermal efficiency of the process. Within the context of torrefaction, the vapour-phase products are known as torgas and in modelling work, have been assumed to have a heating value range of 5.3-16.2 MJ N⁻¹m⁻³ depending on the torrefaction conditions [8]. The technical realisation of autothermal operation, however, has not yet been documented.
The process concept of the National Renewable Energy Centre (CENER) provides a good example of a torrefaction process. CENER operates a pilot-scale torrefaction process at its Second Generation Biofuels Centre in the Navarra Region of Spain. The facility has a capacity to process up to 500 kg h\(^{-1}\) of biomass feedstock [73]. CENER’s proposed torrefaction concept is shown in Figure 9.

Figure 9. Schematic diagram of the torrefaction process concept under development at the Spanish National Renewable Energy Centre (CENER) (Paper IV).

The process is heat integrated. The combustible vapour-phase products generated in the reactor are treated in a thermal oxidiser taking advantage of energy in an attached boiler to heat up the thermal fluid. To optimise reactor throughput, it is convenient to dry the biomass before torrefaction. Flue gases from the boiler at temperatures higher than 300°C are fed to the drier. Additional fuel could be necessary for drying, depending on wood moisture content and target torrefaction degree.

The core of the process equipment is the torrefaction reactor. It is a cylindrical horizontal reactor with an agitator shaft using specially designed elements. It aids efficient transport of feedstock and radial mixing inside the reactor. Reactor heating is carried out indirectly through the hot reactor walls, the actively heated shaft tube and the actively heated internal shaft elements using thermal oil as heat transfer fluid.

The torrefaction reactor can be scaled up to industrial sizes. This is
based on a reactor model developed at CENER. The main features of the model can be found in Paper IV. Figure 10 gives the thermal balance for an industrial production plant producing 40 000 tonnes of torrefied wood pellets annually. This balance is based on the CENER concept and pilot plant data.

![Thermal Balance Diagram](image)

Figure 10. The thermal balance of scaled-up industrial torrefaction plant for annual production of 40 000 tonnes torrefied biomass (Paper IV).

The thermal balance of the CENER process is shown in Figure 10 for an annual production rate of 40 000 tonnes torrefied pellets. The thermal efficiency of the process is 86%. The mass and energy yields of torrefaction are 80 and 90% respectively\(^3\). Among other influencing

\[^3\] Using the information from Figure 10, the input dry mass of feedstock is 6234 kg h\(^{-1}\) (i.e. 12 058 kg h\(^{-1}\) – 0.483*12 058 kg h\(^{-1}\)) and the output dry mass torrefied feedstock 4982 kg h\(^{-1}\) (i.e. 5179 kg h\(^{-1}\) – 0.038*5179 kg h\(^{-1}\)).

\[Y_m = \frac{4982 \text{ kg h}^{-1}}{6234 \text{ kg h}^{-1}} = 0.80\]
factors, the efficiency depends directly on the net heating value of the product and inversely on the moisture content of feedstock. Thermal losses stem from flue gas, product cooling, radiative heat loss and ash amount to 14%. Other inputs to the process (not shown in the Figure) include nitrogen gas, used in purging conveying lines to minimise ignition risks, air for drying and electricity used in process control, conveying feedstock and pelletisation.

4.3 Key Properties Influencing Feasibility

Key properties of torrefied wood are defined as those properties which undergo change through torrefaction and may have some influence on the use of torrefied wood in co-firing. These have been compiled from existing research. Property changes can have a positive or a negative impact on feasibility. This section discussed each property in detail and, based on research findings, describes its influence on fuel properties of torrefied wood.

4.3.1 Mass and Energy Yield (Heating Value Change)

The mass yield $Y_m$ and energy yield $Y_E$ of torrefaction depend on the initial $m_0$ and final $m_T$ mass of the feedstock and the initial $HHV_0$ and final $HHV_T$ higher heating values (dry basis) and are defined as:

(1) $Y_m(\%) = 100 \left( \frac{m_T}{m_0} \right)^{-1}$

(2) $Y_E(\%) = Y_m(HHV_T)(HHV_0)^{-1}$

The mass and energy balance of torrefaction is important because it determines the heating value of the solid product and the required volume of feedstock for each unit of produced fuel. Selected experimental results on mass and energy yields, heating value and torrefaction conditions are given in Table 3. For wood materials the

\[ Y_E = (0.80) \frac{20.7 \text{ MJ kg}^{-1}}{18.5 \text{ MJ kg}^{-1}} = 0.90 \]

Extra thermal power is being put into the torrefaction process by natural gas and combustion of extra feedstock to generate heat. These thermal power inputs are used in drying and torrefying the feedstock so that moisture is removed and heating value improves accordingly.
The range of mass and energy yields are 61-82 and 73-92\% respectively. The corresponding yields for agro biomass are 25-76 and 29-81\%. The relative increase in higher heating value $\Delta \text{HHV}$ is calculated using the following equation:

$$\Delta \text{HHV} (\%) = 100 \left( \frac{\text{HHV}_T - \text{HHV}_0}{\text{HHV}_0} \right)$$

In Table 3, the $\Delta \text{HHV}$ for wood has a range of 7-21\%. The volatile matter and fixed carbon content of the materials are also in the table if they were reported. The number of significant digits in the table corresponds to those given in the original reference. Experimental uncertainties were for the most part not given.

Table 3. Selected experimental results on torrefaction of biomass raw materials from recent literature. $\Delta \text{HHV}$ is the relative increase the higher heating value after torrefaction. Symbols used refer to (a) LHV was used if HHV was not given, (b) Estimated from on-screen graphical data, (c) Calculated value using heating value (LHV or HHV), (d) as received (not daf), (e) Calculated from given data on ultimate analysis and (n/a) Data not available (Adapted from Paper II).

<table>
<thead>
<tr>
<th>Material</th>
<th>Mass Yield (%)</th>
<th>Energy Yield (%)</th>
<th>HV (MJ kg$^{-1}$)</th>
<th>$\Delta \text{HHV}$ ($%$)</th>
<th>T (ºC)</th>
<th>t (min.)</th>
<th>Volatiles (%)</th>
<th>Fixed Carbon (%)</th>
<th>Ref.</th>
</tr>
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<tr>
<td>Willow</td>
<td>78.6$^b$</td>
<td>91.9$^e$</td>
<td>17.7/20.7 (LHV)</td>
<td>16.9</td>
<td>270</td>
<td>15</td>
<td>n/a</td>
<td>n/a</td>
<td>[66]</td>
</tr>
<tr>
<td>Beech</td>
<td>73.8$^b$</td>
<td>88.1$^e$</td>
<td>17.0/20.3 (LHV)</td>
<td>19.0</td>
<td>280</td>
<td>30</td>
<td>n/a</td>
<td>n/a</td>
<td>[66]</td>
</tr>
<tr>
<td>Willow</td>
<td>79.8</td>
<td>85.8</td>
<td>20.2/21.4 (HHV)</td>
<td>7.00</td>
<td>270</td>
<td>30</td>
<td>79.3</td>
<td>18.6</td>
<td>[67]</td>
</tr>
<tr>
<td>Willow</td>
<td>81.6</td>
<td>89.9</td>
<td>19.0/21.8 (HHV)</td>
<td>10.2</td>
<td>290</td>
<td>10</td>
<td>72.4$^e$</td>
<td>23.9$^e$</td>
<td>[74]</td>
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<td>Wheat straw</td>
<td>71.5</td>
<td>78.2</td>
<td>18.9/20.7 (HHV)</td>
<td>9.52</td>
<td>270</td>
<td>30</td>
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<td>26.5</td>
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<td>Rice straw</td>
<td>36.6</td>
<td>39.9$^c$</td>
<td>17.1/18.7 (HHV)</td>
<td>9.11</td>
<td>300</td>
<td>30</td>
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<td>n/a</td>
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<tr>
<td>Rape stalk</td>
<td>25.3</td>
<td>29.1$^c$</td>
<td>18.8/21.6 (HHV)</td>
<td>15.1</td>
<td>300</td>
<td>30</td>
<td>n/a</td>
<td>n/a</td>
<td>[75]</td>
</tr>
<tr>
<td>Lobolly pine</td>
<td>74.2</td>
<td>83.1</td>
<td>19.55/21.80 (HHV)</td>
<td>11.5</td>
<td>275</td>
<td>80</td>
<td>83.0</td>
<td>16.4</td>
<td>[76]</td>
</tr>
<tr>
<td>Lobolly pine</td>
<td>60.5</td>
<td>73.2</td>
<td>19.55/23.56 (HHV)</td>
<td>20.5</td>
<td>300</td>
<td>80</td>
<td>82.3</td>
<td>17.0</td>
<td>[76]</td>
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<td>Miscanthus</td>
<td>75.7</td>
<td>81.0</td>
<td>20.27/21.6 (HHV)</td>
<td>6.98</td>
<td>290</td>
<td>10</td>
<td>63.8$^e$</td>
<td>32.6$^e$</td>
<td>[74]</td>
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<td>Eucalyptus</td>
<td>80</td>
<td>90</td>
<td>19.4/22.2 (HHV)</td>
<td>14.4</td>
<td>240</td>
<td>30</td>
<td>75.4</td>
<td>21.8$^e$</td>
<td>[77]</td>
</tr>
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<td>Pine chips</td>
<td>73</td>
<td>87</td>
<td>18.5/21.8 (HHV)</td>
<td>18.2</td>
<td>275</td>
<td>30</td>
<td>76.4</td>
<td>23.3</td>
<td>[78]</td>
</tr>
<tr>
<td>Southern yellow pine residues</td>
<td>70</td>
<td>82</td>
<td>18.0/22.0 (HHV)</td>
<td>17.2</td>
<td>275</td>
<td>30</td>
<td>71.4</td>
<td>26.7</td>
<td>[78]</td>
</tr>
<tr>
<td>Willow</td>
<td>n/a</td>
<td>n/a</td>
<td>17.6/21.0 (LHV)</td>
<td>19.3</td>
<td>300</td>
<td>10</td>
<td>n/a</td>
<td>n/a</td>
<td>[79]</td>
</tr>
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</table>
4.3.2 Bulk Density and Energy Density of Torrefied Wood

By reducing nominal particle size and inter-particle spaces, the bulk density of wood (poplar and pine) and bark (pine) can be increased significantly for particle size ranges less than one centimetre [42]. Similar increases are achievable with agricultural straws and grasses [49]. Bulk density of real fuels is also a function of moisture. For example, a cubic metre of wet saw dust has more mass than a cubic metre of dry saw dust.

Energy density $\sigma$ describes how much energy can be stored and transported in a certain space. In general it is calculated by taking the product of a fuel’s heating value $HV$ and bulk density $\rho$ and expressed in units of GJ m$^{-3}$.

\[
\sigma = HV \cdot \rho
\]

Both heating value and bulk density are functions of moisture, so that $\sigma$ is also a function of moisture. Any value of energy density, therefore, only has meaning when it is calculated with $HV$ and $\rho$ values that corresponds to the same level of moisture for the fuel in question. It follows that $\sigma$ can have both higher (gross) and lower (net) values like heating value itself.

Wood fuels have a low energy density compared to fossil fuels like coal and oil. Moreover, the energy density of wood – for example, wood chips – cannot be appreciably improved through torrefaction. This is because the gain in $HV$ through torrefaction is offset by the reduced $\rho$ of torrefied wood due to the incurred mass loss.

Wood charcoal is a well-known example of how wood, even when completely devolatised, occupies approximately the same volume of space. To further demonstrate this fact, consider a one-metre solid cubic of dry wood having 20% mass loss and 20% increase in HHV through torrefaction\(^4\). The energy density of the torrefied cubic is less than in the untreated case.

\[^4\] 1 cubic metre untreated birch (dry basis)
Bulk density = 410 kg m$^{-3}$
This also holds for milled wood. For example, saw dust from North American species of spruce, pine and fir has a range of gross energy density 3.4 to 4.2 GJ m\(^{-3}\) after torrefaction while the combined dry and wet untreated range is 3.7 to 4.3 GJ m\(^{-3}\) [80].

Despite this, however, a slight improvement in \(\sigma\) has been demonstrated but necessarily requires a small particle size and consequently a greater energy input for milling. For example, an energy density range of 7.3 to 8.2 GJ m\(^{-3}\) for torrefied pine and logging residue chips exceeded their untreated range of 6.6 to 7.0 GJ m\(^{-3}\) [78].

### 4.3.3 Pellet Production from Torrefied Wood

From the above remarks, it is evident that the ability to pelletise torrefied wood is a key property for its use in co-firing because with it, the amount of fuel energy in a volume of space can be increased. Pelletisation is therefore desirable for improving both \(\sigma\) and also flow properties of torrefied wood.

Most torrefaction research to date is based on laboratory-scale studies which necessarily neglect pelletisation. It is true that private companies and national research organisations have been pelletising torrefied material for many years, but analysis of their methods and results have not been openly available.

The few studies available on pelletisation using a single-pellet press and industrial-scale pelletisers show that torrefied wood is more challenging to pelletise than untreated wood. The best evidence supporting this claim was reported by Larsson et al. who found that spruce wood torrefied at temperatures 270°C and 300°C for 16.5 minutes required 100% more energy to pelletise than untreated spruce

\[
\text{HHV} = 21 \text{ MJ kg}^{-1} \\
\text{Energy density} = (21 \text{ MJ kg}^{-1})(410 \text{ kg m}^{-3}) = 8610 \text{ MJ m}^{-3}
\]

1 cubic metre torrefied birch (dry basis)

\[
\text{Bulk density} = (410 - (0.20 \times 410)) = 328 \text{ kg m}^{-3} \\
\text{HHV} = 25.2 \text{ MJ kg}^{-1} \\
\text{Energy density} = (25.2 \text{ MJ kg}^{-1})(328 \text{ kg m}^{-3}) = 8266 \text{ MJ m}^{-3}
\]
Measurements made in the pressing of individual pellets have shown similar results but have been able to discern between energy used in compaction of feedstock and that used in extrusion of the formed pellet from the die. Li et al. describe a 29-71% and 280-530% increase in compaction and extrusion energy respectively for a spruce-pine-fir (SPF) mix of sawdust torrefied between 260-300°C for 10-90 minutes [82]. Taken together this represents an increase of at least 50%. A reduction of feedstock plasticity – as described by Li et al. – creates more frictional forces among wood particles and between feedstock and walls of die channels. The latter force was observed to increase some 10-30 times for spruce and wheat straw [83]. Moreover, enhanced friction also causes bridging of feedstock in the conveyor system and, contrary to experiences in conventional pellet making, is worsened by the addition of water [81]. The role of water in pelletisation has thus been shown to change because of changes in the properties of the torrefied wood. These changes include the loss of OH groups and the lowering of moisture content which affects inter-particle surfaces.

Based on their work, both Larsson and Li et al. recommend a higher die temperature to improve pelletisation success and reduce energy consumption. The latter author suggests that this is likely necessary in order to get mid-molecular weight lignin polymers sufficiently involved in inter-particle bonding. Low weight lignin polymers – the ones implicated in conventional pellet bonding – are absent being the first to decompose during torrefaction [82]. Despite the abovementioned recommendation by Larsson, a further study by Segerström and Larsson found, in fact, the opposite result: that die temperature was negatively correlated to both pellet bulk density and durability [29]. Furthermore, by using a modified (stationary) ring-die press, Larsson and Rudolfsson found that actually, low die temperature (30°C) was best for ensuring good quality of pellets made of reed canary grass [84]. Their two indicators of pellet quality in this study were bulk density and durability.

Peng et al. carried out pelletisation of individual torrefied pellets using elevated die temperature from 170°C to 260°C. By use of a heated die unit, the authors found that the hardness of pellets does indeed increase with increasing die temperature. They found that at 240°C and above hardness values were equal to or exceeded those of the control pellets (composed of untreated wood). Energy
consumption and required compression pressures were lower when using higher die temperature. Yet despite these positive findings, Larsson et al. claim that conclusions reached in single-pellet-press studies are not valid in industrial scale studies [84].

Realising a higher die temperature with commercial pellet mills would require modification in design of mills including temperature regulation mechanisms and possibly an addition heat source. In addition to the cooling of produced pellets, the risk of ignition also needs consideration. Finally, greater frictional forces within the die during operation with torrefied wood will lead to greater wear and maintenance of the mill. These issues will change some aspects of pellet production but what is clear is that much more research is needed to clarify the above observations.

4.3.4 Bulk Density and Energy Density of Torrefied Pellets

There are few studies documenting the bulk and energy density of torrefied pellets produced at semi-industrial scales. Pellets produced by Larsson et al., described above, had a bulk density range of 683–713 kg m\(^{-3}\) with moisture content range of 1-5% as received [81]. These pellets had an energy density range of 12.3-14.7 GJ m\(^{-3}\).

4.3.5 Friability

The friability of a material is a measure of how easily it can be milled. Torrefaction increases the friability of wood and, thus, lowers the energy required to mill it. The loss of the hemicellulose matrix through torrefaction is thought to be responsible for this change and the milling energy of torrefied wood has been shown to have a linear dependence on the hemicellulose content [85]. The increase in friability of torrefied wood is also accompanied by a shift to smaller particle sizes, in the size distribution after milling, as the degree of torrefaction increases [77, 86].

Experimental measurements of the energy requirements of grinding for four woody samples are summarised in Table 4. The energy needed for all samples was greatly reduced after torrefaction. The
used grinding energy (52–150 kWh t⁻¹, d₅₀=93–460 µm) was reduced by 68–89% compared to untreated wood [78, 87].

Table 4. Experimental results on the grindability of torrefied wood. Symbols used refer to (a) Change in grinding energy \( \Delta E_g = 100\left(\frac{E'_g}{E_g}\right) \) (b) The heating values (expressed as MJ kg⁻¹) used in this calculation were Spruce 19.0 [88], Pine 18.46, Logging residues 18.79 [78] and Beech 17.0 [87]. (c) Values calculated by fitted equation from reference at 25°C (Paper II).

<table>
<thead>
<tr>
<th>Material, T (°C)</th>
<th>t (min.)</th>
<th>Initial particle size</th>
<th>Final particle size</th>
<th>( E_g ) (kWh t⁻¹)</th>
<th>( E'_g ) (kWh t⁻¹)</th>
<th>( \Delta E_g )%</th>
<th>Heating value fraction%</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beech 280</td>
<td>5</td>
<td>2-4 mm</td>
<td>( d_{50}=140 ) µm</td>
<td>840</td>
<td>90</td>
<td>-89</td>
<td>2 (LHV)</td>
<td>[87]</td>
</tr>
<tr>
<td>Spruce 280</td>
<td>5</td>
<td>2</td>
<td>( d_{50}=93 ) µm</td>
<td>750</td>
<td>150</td>
<td>-80</td>
<td>2.8 (HHV)</td>
<td>[87]</td>
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<tr>
<td>Pine 275</td>
<td>30</td>
<td>20.94-70.59 x 15.08-</td>
<td>( d_{50}=279 ) µm</td>
<td>241</td>
<td>52.0</td>
<td>-78</td>
<td>1.0 (HHV)</td>
<td>[78]</td>
</tr>
<tr>
<td>Logging residues</td>
<td>275</td>
<td>39.70 x 1.88-4.94 mm</td>
<td>( d_{50}=468 ) µm</td>
<td>242</td>
<td>78.0</td>
<td>-68</td>
<td>1.5 (HHV)</td>
<td>[78]</td>
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</tbody>
</table>

The utility of the Hardgrove Grindability Index for coal has led to it being applied to torrefied wood. One investigation found that the grinding behaviour of willow and its particle-size distribution, after torrefaction at 290°C for 60 minutes, is within the boundaries of reference coals having a HGI range of 32 to 92 [74]. It is noted, however, that these torrefaction conditions are rather severe and under milder conditions HGI was not a reliable indicator of friability.

If the friability of torrefied wood is equal to that of coal, then torrefied wood could be passed through existing coal mills at the power plant. This would permit greater co-firing rates without the need for additional investments. Unfortunately, industrial-scale energy requirements for pulverising torrefied wood and pellets are not well documented. Industrial-scale size reduction uses larger mills but despite the difference in scale, the relative differences in required energy in Table 4 are likely to remain. Based on the aforementioned work on conventional wood pellets [89], the milling energy requirements of torrefied pellets are thought to be less than torrefied wood chips but this has yet to be observed.
4.3.6 Equilibrium Moisture Content

The moisture content of fuel stored at constant temperature and relative humidity (RH) will over time approach a dynamic equilibrium which is characteristic for that fuel. This level is known as the equilibrium moisture content (EMC).

Moisture in wood exists in what is characterised as three different forms. Free water is that which evaporates easily from the surface of wood structure - this is the familiar form of moisture in the macro world. In addition, capillary water (or non-bonded water) is present in small pores within wood fibres. Here the vapour pressure can be very low due to the pressure gradient between the external wood surface and inner structure. As such, water trapped in pores does not experience low relative humidity conditions that may exist during drying of the external surface and is difficult to evaporate. The third form of water in wood is that which is absorbed and bonded - via hydrogen bonding – to chemical binding sites that exist on wood polymers [90]. Bonding takes place on free OH groups. These groups are more numerous in hemicellulose and (non-crystalline) cellulose components [91].

OH-binding sites are present in carboxylic acid groups chiefly located in the uronic acids of the hemicellulose component [92]. A correlation between the number of carboxylic groups and the EMC of wood has been shown [92]. Therefore, thermal decomposition of these groups through torrefaction lowers the number of free sites and consequently, the EMC of wood. If complete removal of OH-binding sites could be achieved, capillary water is always present in wood. It represents a kind of minimum moisture level which is characteristic of wood [91].

EMC is an important property of a fuel because it directly influences the net heating value. The net heating value, called the lower heating value as received \(LHV_{ar}\) (MJ kg\(^{-1}\)) is found by the following relation in which \(HHV_V\) (MJ kg\(^{-1}\)) is the higher heating value measured at constant volume, \(M\) (%) is the amount of moisture and \(H\) (%), \(O\) (%) and \(N\) (%) are the content of elemental hydrogen, oxygen and nitrogen in the fuel - expressed in dry mass per cent (EN 14918).

\[
LHV_{ar} = \{HHV_V - 2122 \cdot H - 0.8 \cdot [O + N]) \cdot (1 - 0.01M) - 24.43M
\]
It is important to emphasise that EMC values have no meaning without knowing temperature and relative humidity at which the fuel is kept. Additionally, reported values need to include the basis of the measurements (i.e. wet or dry basis). The differences in EMC for untreated and torrefied wood are at a maximum under high relative humidity and minimum for low RH. At constant temperature, the values of EMC as a function of RH form continuous isotherms. EMC isotherms based on measurements of beech wood are presented in Figure 11.

Figure 11: Measured isothermal curves for equilibrium moisture content (wet basis) of untreated (at 8 and 22°C) and torrefied beech stem wood (at 7 and 22°C). The EMC of pellet Sample 7 at 23°C and 85% RH is also shown (Paper III).

Additionally, Table 5 summarises these and other EMC observations of torrefied wood from literature.
Table 5. EMC results from literature (adapted from Paper II).

<table>
<thead>
<tr>
<th>Wood species</th>
<th>T (°C)</th>
<th>t (min.)</th>
<th>RH (%)</th>
<th>Ambient T (°C)</th>
<th>EMC, untreated (%)</th>
<th>EMC, torrefied (%)</th>
<th>ΔEMC (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spruce</td>
<td>260</td>
<td>80</td>
<td>75</td>
<td>23</td>
<td>12</td>
<td>9</td>
<td>-3</td>
<td>[76]</td>
</tr>
<tr>
<td>Spruce</td>
<td>300</td>
<td>(80)</td>
<td>11</td>
<td>23</td>
<td>3.5</td>
<td>2.2</td>
<td>-1.3</td>
<td>[90]</td>
</tr>
<tr>
<td>Beech</td>
<td>273</td>
<td>45</td>
<td>75</td>
<td>22</td>
<td>12.1</td>
<td>7.0</td>
<td>-5.1</td>
<td>III</td>
</tr>
<tr>
<td>Beech</td>
<td>273</td>
<td>45</td>
<td>11</td>
<td>22</td>
<td>4.6</td>
<td>1.9</td>
<td>-2.7</td>
<td>III</td>
</tr>
<tr>
<td>Spruce/Pine/Fir mix</td>
<td>300</td>
<td>10-15</td>
<td>90</td>
<td>30</td>
<td>20.7</td>
<td>12</td>
<td>-8.7</td>
<td>[82]</td>
</tr>
<tr>
<td>Spruce/Pine/Fir mix</td>
<td>300</td>
<td>60</td>
<td>90</td>
<td>30</td>
<td>19</td>
<td>10</td>
<td>-9.0</td>
<td>[93]</td>
</tr>
</tbody>
</table>

4.3.7 Moisture Content from Non-EMC Conditions

Moisture content is frequently reported on an as-received basis but from the aforementioned points, these values contain no real precision. Nonetheless, the moisture content of torrefied pellets under non-EMC conditions has been reported by Larsson et al. Their pellets produced from torrefied spruce (270-300°C) were measured to have a moisture content range of 0.8-3.9% after cooling [94].

4.3.8 Resistance to Rainfall and Water Immersion

Pellets that are susceptible to moisture may undergo swelling and disintegrate into particle form. Even short term exposure to water affects the structural integrity of pellets. Currently, there are no standards to determine the resistance of torrefied pellets to moisture nor does this topic appear in literature. Two non-standard methods of resistance to moisture, however, have been devised and are described in Chapter 5.

4.3.9 Mechanical Durability of Torrefied Pellets

The mechanical durability of fuel pellets is a standardised measure (EN 15210) which gives an indication of the fuel’s ability to retain its form during transport and handling processes without going to pieces. The measurement procedure requires a 500-gram sample of pellets. The pellets are placed in a box and tumbled at 50 ± 2 revolutions per minute for a total of 500 rotations. The sample is then
sieved and the fraction of pellets that remain intact is expressed as a per cent value.

The limited number of durability studies on torrefied pellets is a reflection of the few existing pelletisation studies. Two studies have measured durability after pelletisation on the semi-industrial scale.

Durability and hardness of torrefied Norway spruce pellets were found to be problematic and in all cases below the normative minimum (95%) of conventional wood pellets. This study reported a durability range of 82.8% to 90.3% for samples torrefied at 270 and 300°C and having a range of die temperatures [94]. ECN made no mention of durability problems with torrefied pellets [7]. On the contrary, the report finds that the durability of torrefied pellets is better than conventional pellets. This implies a durability rating higher than 98% which is routinely achieved with commercially produced wood pellets. Pellet hardness, a non-standardised measure, is stated to be between 50% and 200% greater than conventional pellets. Additionally, the improved hardness of torrefied larch-wood pellets is reported to be 250%.

4.3.10 Fines Content and Dust

Due to the aforementioned shift of the particle-size distribution for torrefied wood, average particle size from milling decreases when higher torrefaction temperature or longer residence times are used. For eucalyptus wood particles, this is shown to be primarily from the shortening of needle-shaped particles to those of a more spherical nature [77]. This is seen in optical photomicrographs of torrefied material. Extending this finding to the smallest of wood particles undergoing torrefaction is a sufficient explanation for the increase in fine dust seen in handling, conveying and grinding of torrefied material [95].

At large particle sizes, explosion risks associated with handling torrefied wood appears no more hazardous than those from untreated wood dust. However, for particles sizes below 150 micrometres, the minimum explosible concentration (MEC) for torrefied wood
decreases indicating that explosion risks are significantly enhanced [96]. Moreover, torrefied wood dust is far more reactive than coal dust and exhibits MECs three times lower than those of coal [96]. It is not known, whether these differences in dust reactivity during co-firing would necessitate additional safety measures at coal plants. For example, the elimination of dust formation and possible ignition sources may become more stringent.

4.3.11 Conveying Properties

The increase in inter-particle friction of torrefied wood suggests that its conveying properties may be affected. For example, qualitative observations that torrefied wood feedstock has flow problems in the pellet-making process have been made [81]. Additionally, the author himself, in a project described in Paper III, has taken part in pellet-making demonstrations using torrefied beech and can confirm that feedstock blockages were more frequent than in pelleting using the untreated feedstock. However, it is not clear if the flow of feedstock in pellet making is directly comparable to the handling and conveying operations in co-firing. No data on conveying properties of torrefied wood or torrefied pellets has been published to date.

The more spherical nature of torrefied wood particles supports the view that, as particles, torrefied wood should flow more easily. The angle of repose (AOR) of a granular material is a measure of its resistance to flow. Laboratory investigations of AOR of milled wood in conical piles have shown that an increase in the torrefaction degree corresponds to a smaller AOR – meaning that piles of torrefied wood are lower than those of untreated wood [85]. Torrefaction of Norway spruce at 260°C and above reduces the AOR to approximately 67° from the untreated value of 69°. A more significant change occurs after torrefaction at 310°C when the AOR was found to be some 63°. Therefore, these results show that torrefaction enhances flow.

5 The Minimum Explosible Concentration MEC (g m\(^{-3}\)) for TM 7 compared to Raw 7 are 26:37, 32:44 and 52:56 for particle sizes below 150 micrometres [Table 2][96] – this indicates that for small particle size, torrefied biomass is more explosive. But at particle sizes greater than 150 um, raw wood is more explosive.
4.3.12 Combustion Behaviour of Torrefied Wood

Before the interest in torrefied wood for co-firing, it was thought it could replace wood charcoal use in cooking. Girard et al. reported on the benefits of cooking with torrefied wood which, compared to untreated wood, was easier to ignite and smoked less while showing a similar overall combustion time [58]. The main limitation of torrefied wood, however, was reported to be the length of its ember phase of burning – only one third of that of charcoal – which made it unattractive in practice.

Although torrefaction reduces the amount of volatile content in wood, it is still high compared to coal. For large-scale combustion, this means that torrefied wood particles still ignite more easily than coal particles of the same size. The increase in char content may increase the carbon content in fly ash as compared to wood particles of the same size when burned in similar burner and furnace.

Studies on how torrefaction influences char properties have been carried out on wood and non-wood biomass. Torrefied wood char was reported to be less reactive than untreated wood char [97] and studies on non-wood biomass suggest that any influence on oxidation reactivity is minor [98, 99].

Laboratory studies using a methane air flame and a high-speed camera showed that the ignition times (observed visually and assumed to be independent of particle mass) of both combustible volatiles and char of torrefied wood decrease as a function of torrefaction temperature [67]. The total time needed for complete combustion, however, increased some two or three times for particles between 0.5 and 1.5 mg. In other words, the combustion behaviour of torrefied wood is a step away from untreated wood. This indicates that required burnout times for particles are extended through torrefaction which suggest optimal particle size could be slightly lower than those for untreated wood [50] but otherwise not an issue.

Modelling Combustion of Torrefied Wood

CFD modelling results have shown that 100% substitution is possible for a coal plant without loss of boiler efficiency [100]. In addition, NOx emissions would be significantly reduced. The volatile and char
content of the modelled torrefied wood used in the study were 62 and 31% respectively, with a LHV of 18 MJ kg\(^{-1}\). These properties corresponded to an approximate dry mass yield of 80%.

Other CFD simulations of co-firing torrefied wood suggest that up to 50% mass-based substitution (about 40% energy basis) of coal is feasible for a modelled 275 MW (thermal) pulverised-coal furnace (corner-firing type with 12 coal burners on three levels) \[95\]. The torrefied wood had a volatile content of 83% and char content of 17%, with a LHV of 18.4 MJ kg\(^{-1}\). This study concludes that co-firing combustion efficiencies will be comparable to the pure-coal case but with a reduction of about 40% in the total amount of ash. An increase in unburnt carbon in the fly ash, owing to coal mill performance, is also predicted. Both combustion efficiency and the fly ash issue can be improved by using dedicated mills for torrefied fuels so that a better particle-size distribution can be achieved. Additionally, the results of the study predict a reduction in NO\(_x\) emissions and, despite model uncertainties, see no drastic rise of CO in stack emissions. A higher co-firing rate was not considered in this study.

**Pilot-Scale**

The unpublished results from co-firing trials with coal in a 0.5 MW boiler have been described \[86\]. This work indicated that combustion efficiency remains constant or increases slightly with a mass-based co-firing rate of 30-70% torrefied pellets. A reduction in coal-mill performance - larger particles and less small particles - was observed with increasing fraction of torrefied wood. The torrefied pellets used had a LHV of 18.9 MJ kg\(^{-1}\). Trials using 100% torrefied wood were not presented. These trials are not described in literature.

**4.3.13 Gasification Behaviour of Torrefied Wood**

The use of torrefied pellets as a gasification feedstock has been investigated. When using an industrial-scale air-blown gasifier (300 kg/h feed) a number of differences have been observed compared to the gasification of conventional pellets with the same apparatus \[101\]. Torrefied pellets tended to combust rather than gasify, which was thought to be due to low moisture content and volatile matter. Higher temperature was produced in the gasifier when using torrefied wood.
compared to untreated – although it is unclear if this was a result of a difference in the initial state of the gasifier. The gasification rate for torrefied wood was half that of conventional pellets. Moreover, the rate of tar collection from gasification of torrefied pellets was also reduced by some 75%. The study concluded that, compared to sawdust and conventional pellets, effective and stable gasification was difficult to achieve using torrefied pellets.

Although it is not relevant to stand-alone production of torrefied wood fuels, torrefaction as a pre-treatment process in wood gasification has been studied. Results showed that if vapour-phase products from torrefaction could be thermally integrated with the gasification process, then the efficiency of gasification could be improved [79].

4.3.14 Resistance to Biological Degradation

Bacteria and fungi colonise wood in the natural environment causing decomposition in the long-term. Micro-biological organisms utilise easily consumable substances in wood which are those that also undergo degradation in torrefaction. Therefore, torrefied wood exhibits better resistance to degradation than it does in its natural state. The period of effective storage of wood is then prolonged through torrefaction. Initial results of studies on this subject clearly indicate an enhance resistance to biological decay under conditions of high humidity [102]. By reducing biological decomposition (both aerobic and anaerobic) of stored fuel, carbon dioxide and methane emissions to the air are also reduced.

4.3.15 Chlorine Content Reduction

There is some indication that torrefaction of wood reduces the chlorine content [62, 103, 104]. Keipi et al. have observed up to a 90% reduction in chlorine concentration through torrefaction (260°C) of eucalyptus and Shoulaifar et al. also report a reduction for birch wood at 280°C. As chlorine can lead to problems during combustion, this would be a considerable advantage of torrefaction of high-Cl feedstock, for example cereal straws. However, as wood generally has very low chlorine content these findings are not applicable herein.
4.4 Torrefaction at Elevated Pressure

Many industrial thermochemical processes utilise elevated pressure to enhance economic attractiveness. Pressurised reactors can increase rates of chemical reactions leading to improved efficiency and smaller size while enabling higher throughput of products. The use of pressurised reactors, however, can also complicate process designs and increase investments costs.

Existing pilot and semi-industrial-scale torrefaction reactors, most of which are designed as a continuous process, operate at or slightly above atmospheric pressure. As in other industrial processes, the influence of elevated pressure on torrefaction is of interest for possible benefits it may bring to the feasibility of this process. In particular, high-pressure torrefaction is likely to alter mass and energy yields and elemental composition thereby affecting heating value and other key properties already described.

Past investigations of wood pyrolysis at elevated pressure are almost exclusively above 300°C. The influence of pressure on torrefaction might be expected to be similar to that on pyrolysis at higher temperature; for example, charcoal yields can be improved using elevated pressure [105]. Below 300°C, however, only two studies using elevated pressure have been described and these used batch reactors with no flow conditions.

In their study, Wannapeera et al. investigated torrefaction of a tropical wood using elevated pressures of 1 to 4 MPa, temperatures of 200-250°C and a torrefaction time of 30 minutes. They found that the total carbon content increased progressively with torrefaction temperature and reactor pressure from 50 to 62% [106]. The greatest calculated higher heating value (HHV) was 25.8 MJ kg⁻¹ from torrefaction at 250°C and 4 MPa. A HHV increase of 26.5% is reported which corresponds to a mass and energy yield combination of 74.4 and 94.1% respectively. Nhuchhen et al. studied torrefaction of cylindrical poplar samples (50.5 mm, ø 19 mm) in a batch reactor using nitrogen pressures up to 0.6 MPa [107]. They report a HHV and mass yield range of 19.5-26.0 MJ kg⁻¹ and 56-92% respectively using torrefaction temperatures of 220-300°C and 15-35 min. residence times. Deposition
of tars on the torrefied wood was also observed in this study and their size increased with pressure.

In these studies, no flow through the reactors meant that vapour-phase pyrolysis products remained in contact with the solid sample. As a consequence, the influence of pressure and vapour-phase residence time on solid yields cannot be differentiated in their results.
5. Methodology

In this section, the quantitative and qualitative methods used in this study are presented. These include: on-site research visits and demonstrations, standardised methods of pellet characterisation, non-standardised methods of pellet characterisation, laboratory CHNS analysis, laboratory-scale torrefaction at elevated pressure, environmental impact assessment (RED) and economic analysis of pellet production.

5.1 On-Site Visits and Demonstrations

A great deal of practical knowledge on torrefaction and pellet production was gained through discussions during on-site visits and demonstrations at a variety of venues (Table 6). Most of these experiences were realised or initiated through national and international bioenergy projects. The following table summarises some of the most influential activities.

Table 6. The author’s participation in torrefaction-related on-site visits and demonstrations.

<table>
<thead>
<tr>
<th>Activity</th>
<th>Location/Facilities</th>
<th>Project or initiative</th>
</tr>
</thead>
<tbody>
<tr>
<td>Guided tour of the world’s first commercial torrefied pellet plant (21.6.2011)</td>
<td>Topell Energy, Duiven, The Netherlands</td>
<td>Own</td>
</tr>
<tr>
<td>Study trip (17-28.10.2011) Navarra Region, Spain</td>
<td>BIOCLUS</td>
<td></td>
</tr>
<tr>
<td>Site visit to commercial torrefaction plant construction (27.10.2011)</td>
<td>Thermya, Urieta, Spain</td>
<td>Own supported by CENER</td>
</tr>
<tr>
<td>Two-month research visit (4-5.2012)</td>
<td>CENER pilot plant, Sarriguren, Spain</td>
<td>Own supported by Jyväskylä University and CENER</td>
</tr>
<tr>
<td>Bench-marking visit to torrefaction pilot plant (14-17.11.2011)</td>
<td>Andritz, Frohnleiten, Austria</td>
<td>BIOCLUS</td>
</tr>
</tbody>
</table>
5.2 Standardised Fuel-Pellet Characterisation

Existing standardised methods of fuel pellet characterisation were used when possible (Paper III and IV). These standards were defined for common solid biofuels which includes conventional wood pellets. The applicable standards included the determination of calorific value (EN 14918), the determination of bulk density (EN 15103), the determination of moisture content (EN 14774) and the mechanical durability of pellets (EN 15210). The pellets sample types in Paper III were chosen based on preliminary thermogravimetric experiments and single-pellet-press trials using torrefied feedstock.

5.3 Non-Standardised Fuel-Pellet Characterisation

Non-standardised methods were used to determine some properties of fuel pellets and these included the determination of equilibrium moisture content and moisture uptake. Pellet hardness was also determined in Paper III but is not described herein.

5.3.1 Equilibrium Moisture Content Determination

EMC determination for torrefied pellets utilised a condensing dryer as described in Paper III. The determination of EMC of torrefied wood samples (non-densified) made use of super-saturated solutions of salts to provide static conditions of RH [108]. This method is also described in Paper III.

5.3.2 Moisture Uptake from Rainfall

The improved resistance to moisture uptake shown by torrefied wood makes outdoor storage of torrefied fuels an interesting possibility. However, no standardised method exists for testing pellet susceptibility to moisture uptake from rainfall or immersion in water. Therefore, the following methods were devised to test pellet susceptibility.
A 1 kg sample of pellets was placed on a 450 mm diameter Retsch 3.15 mm sieve. The amount was sufficient to cover the bottom of the sieve with one layer of pellets. The sieve was placed over a container. Simulated rainfall was realised through use of a spray bottle fitted with multi-orifice nozzle. In total, 400 g of water was sprayed over each sample during a one-hour period. Runoff water drained through the sieve and was collected in the container beneath. The mass of water not absorbed by the pellets could then be determined. The total amount of water corresponds to rainfall of 2.5 mm per hour – a level of rainfall intensity which occurs in Finland statistically once every decade.

5.3.3 Moisture Uptake from Water Immersion

Pellets were also subjected to a water immersion test as described in Paper III. A 500 g sample of each pellet type was placed in a filtration bag which was then submersed for 15 minutes in a five-litre container of water. By weighing the quantity of water after the immersion period, the amount of water absorbed by the pellet sample could be known.

5.4 CHNS Analysis

CHNS analysis of wood samples (Paper V) was done using a FLASH 2000 Series Organic Elemental Analyser manufactured by Thermo Scientific. Sample sizes ranged from approximately 1.4 to 2.3 mg for all measurements. Averages of measurements performed in triplicate are presented. The oxygen content was calculated based on the difference between the sum of other elements and one hundred per cent. The ash content of the samples was not determined.

5.5 Environmental Impact of Pellet Production

The aim of the environmental impact assessment was to estimate two quantities: the use of energy input for an operation, known as primary energy use, and the amount of generated CO₂-equivalent air emissions. The Renewable Energy Directive (RED) provides sustainability criteria by which alternate production methods can be compared based on their CO₂-equivalent air emissions to the
environment [2]. The use of these criteria for determination of emissions from two different products from the same raw material negates the need of analysing the entire life-cycle of both products. It simplifies comparison by focusing on relative differences between two products. This is sensible for well-established industries such as the wood pellet industry. Further information on RED methodology can be found in Paper IV.

5.5.1 CO$_2$-Equivalent Emission Balance

Equation 6 is used to calculate the total emissions $E$ from the production of the solid biomass fuel before energy conversion. This is simply the sum of the emissions from harvesting ($e_c$), land-use changes ($e_l$), processing ($e_p$), transport ($e_t$) and utilisation ($e_u$) with emission savings from any soil carbon accumulation ($e_{sca}$) or carbon capture ($e_{ccs}$ and $e_{ccr}$) subtracted.

$$
E = e_c + e_p + e_p + e_t + e_u - e_{sca} - e_{ccs} - e_{ccr}
$$

The total emissions from the final energy commodity $EC_{el}$ (i.e. electricity) are found by dividing Equation 6 by the efficiency of electrical generation $\eta_{el}$ at the plant where the fuel is combusted.

$$
EC_{el} = \frac{E}{\eta_{el}}
$$

The use of the biofuel results in an emission savings $S_{el}$ when compared to the use of fossil fuel. The extent of this savings is found by using Equation 8 in which the fossil fuel comparator $EC_{F(el)}$ is used.

$$
S_{el} = \frac{EC_{F(el)} - EC_{el}}{EC_{F(el)}}
$$

5.5.2 Energy Balance and Net Energy Ratio

When comparing the production of two or more fuels, the concept of net energy ratio (NER) is helpful. This value is the ratio of the thermal
energy output $q_{\text{output}}$ extracted from the produced fuel to $q_{\text{input}}$ the energy used in its production.

\[ NER = \frac{q_{\text{output}}}{q_{\text{input}}} \]

5.5.3 Energy and Emission Factors

To calculate energy and CO$_2$-equivalent emission balance of a process, the contribution of individual consumables is assessed. An energy factor describes the amount of primary energy represented in the consumable substance. The CO$_2$-equivalent emission factor describes the amount of emissions attributed to the consumable. The two factors can be thought of as energy and emission footprint. The following factors were used in calculating energy input and emission output from the production scenario.

Table 7. Energy and CO$_2$-equivalent emission factors of production scenarios inputs and outputs. Energy and emission factors are taken from Official Journal of the European Union except for water and nitrogen values which are from Ecoinvent Database. N.A. indicates Not Applicable (Paper IV).

<table>
<thead>
<tr>
<th>Input/ output (unit)</th>
<th>Energy factor (kJ)</th>
<th>CO$_2$-equivalent (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>diesel (L)</td>
<td>41,597</td>
<td>3,143</td>
</tr>
<tr>
<td>electricity (kWh)</td>
<td>9,819</td>
<td>465</td>
</tr>
<tr>
<td>natural gas (kWh)</td>
<td>4,061</td>
<td>243</td>
</tr>
<tr>
<td>utility water (m$^3$)</td>
<td>5,337</td>
<td>263</td>
</tr>
<tr>
<td>nitrogen gas (kg)</td>
<td>9,189</td>
<td>436</td>
</tr>
<tr>
<td>methane (kg)</td>
<td>N.A.</td>
<td>25000</td>
</tr>
</tbody>
</table>

5.5.4 Pellet Production and Supply Chain

The scenario used for this study compares the energy and CO$_2$-equivalent emission balance from the production of conventional pellets to that of torrefied pellets. In the base case of this study, pellet
production takes place in Finland and end use of the pellets in co-firing takes place in Spain. This represents an intra-European production and use scenario. In the second case, production takes place in western Canada which is representative of a long-distance transport distance and where significant production and export of conventional pellets is already a well-established enterprise. Both pellet types are produced from the same feedstock; logging residues from final timber harvest. The system considered starts from the logging residue harvesting operation and ends with the direct co-firing of pellets with fossil coal (Figure 12) – the final felling operation is outside the system boundary.

Figure 12. The four stages of the pellet production and supply chain are represented by the larger cells. Separate stage operations are depicted in the order they occur. Drying and torrefaction operations are described in Figure 10. The final felling operation of forest at the site from which logging residues are collected is outside the system boundary (Paper IV).

The results from the study of environmental impact are presented in Chapter 6.
5.6 Economic Analysis of Torrefied Pellet Production

Despite a decade of torrefaction research, the economic feasibility of torrefied pellet production is still a topic of considerable uncertainty. This is partly due to differences in reactor and process technologies currently under development. Understandably, proprietary interests of those commercialising their technology has also been a reason for the lack of transparency in process and technical specifications.

An earlier economic analysis of torrefied pellet making by ECN gave a favourable impression to potential investors [7]. Their analysis used a set of specifications of torrefied pellets which over the years have been widely cited in subsequent studies [109-115]. ECN’s economic conclusions were very clear: there is a clear economic benefit of producing torrefied pellets over that of conventional pellets for the scenario considered. They found that the production of torrefied pellets generated greater cash flow over the lifetime of the investment resulting in a payback time half that of conventional production with a significant difference in the internal rate of return.

From the findings described in Chapter 4, however, it is clear that in achieving a torrefied fuel in pellet form, compromises need to be made with regards to key properties. In other words, ideal properties in all aspects cannot be realised simultaneously. Instead, it is better to speak of an optimised pellet whose documented properties have begun to appear in literature. It is a good time, therefore, to revisit the economics with the state-of-the-art pellet specifications.

An economic evaluation is beneficial to determine whether a candidate technology is commercially viable. In the case of torrefied pellet production, the minimum of information required for such an evaluation includes: the amount of capital invested and the lifetime of a production plant, the quantity of consumable goods used in production, the thermal balance of the production process and the market value of produced pellets – which depends on their fuel properties.

As with the determination of environmental impact, economic evaluation of torrefied compared to conventional wood pellet production is worthwhile because the latter production process is an
established industry. Moreover, the costs associated with pellet production from lignocellulosic feedstock are reasonably well established [32, 33, 116, 117].

The economics of producing torrefied pellets can be compared to conventional pellets. By using published data on fuel properties of both pellet types and data on consumable inputs used in their production, the production costs and market value of the pellets is estimated. Despite not knowing the investment cost of a given torrefaction process, the maximum investment of such a process can be determined using the market value of fuel.

The following question can be answered through this economic analysis: Based on state of the art in torrefied pellet making today, what would be the maximum capital investment acceptable so that their production is economically viable?

The scenario used for the analysis is not the same as that used for the environmental impact study. It is simpler and compares two production processes: torrefied pellets and conventional wood pellets. Both processes use the same amount of saw dust as a feedstock but annual production amounts from each plant is not the same due to mass loss through torrefaction and the greater energy density of torrefied pellets. The annual production is 64,000 and 80,000 tonnes for torrefied and conventional pellets respectively. The two fuels are produced at the same location and shipped the same distance to their end-use location where they are sold. The analysis estimates the net present value of the investment, the pay-back period and describes the sensitivity of the results to production parameters and pellet properties.

5.6.1 Capital Investment and Depreciation

Capital investment is the total cash invested by the shareholders in a commercial enterprise. It includes working capital, the cash on hand that investors own, and the financed capital, the amount taken on the loan which must be paid back at a financing rate \( i \) after a certain time period \( n \) measured in years. The capital recovery factor (CRF) is found from the relation [33]:

\[
CRF = \frac{i(1 + i)^n}{(1 + i)^n - 1}
\]
(10) \[ CRF = \frac{i(1+i)^n}{(1+i)^n - 1} \]

The product of the CRF and the total financed capital is the annual financing cost of the loan.

The value of a tangible investment depreciates with the passing of time. The depreciation \( C_D \) is an annual cost which is often assumed to be of a linear form.

(11) \[ C_D = \frac{C_i - C_S}{t_D} \]

The amount of annual depreciation is calculated from the difference between the capital investment \( C_i \) and the end-of-life value of the investment \( C_S \) divided by the lifetime \( t_D \) of the investment.

5.6.2 Production and Logistics Costs

Pellet production costs consist of feedstock cost, wages for personnel and the cost of utilities and other consumables during production.

Logistics Costs

Logistics and transport costs can be broken down into individual physical operations with regard to packaging, storing, loading and shipping of the product [7]. In the scenario considered, pellets are shipped from a production site to their end use location, a port-to-port distance of some 11 450 km. To put this into perspective, this is the shipping distance from the Finnish port of Kotka to Rio De Jan Eiro, Brazil \(^6\), and is much greater than any intra-European shipping distance within the present day EU.

The cost of most logistics operations depends only on the volume of product. Product volume depends on annual tonnage of produced pellets and on bulk density. The annually produced amount of torrefied pellets has less volume than produced conventional pellets

\(^6\) International port-to-port shipping distances:
http://www.searates.com/reference/portdistance/
because their annual tonnage is less and their bulk density is greater. Logistical costs for torrefied pellets are consequently lower.

The ratio of product volume ($V_{tor}/V_{con}$) was used by ECN to scale the logistical costs of torrefied pellets in relation to conventional pellets [7]. This ratio had a value of 0.57 in the ECN analysis. Using this same method, the handling and transport costs were calculated herein based on the conventional pellet costs used by ECN. However, in this study the ratio has a value of 0.74 (Table 9). The itemisation of handling and transport costs for both processes is presented in Table 8.

Table 8. Itemisation of handling and transport operations and their cost, adapted from [7] (Paper VI).

<table>
<thead>
<tr>
<th>Operation</th>
<th>TOR (€ t(^{-1}))</th>
<th>CON (€ t(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Road transport to harbour</td>
<td>4,40</td>
<td>4,40</td>
</tr>
<tr>
<td>Harbour storage</td>
<td>1,33</td>
<td>1,80</td>
</tr>
<tr>
<td>Transfer handling</td>
<td>2,96</td>
<td>4,00</td>
</tr>
<tr>
<td>Sea transportation</td>
<td>25,63</td>
<td>34,62</td>
</tr>
<tr>
<td>Transfer handling</td>
<td>2,96</td>
<td>4,00</td>
</tr>
<tr>
<td>Harbour storage</td>
<td>1,55</td>
<td>2,10</td>
</tr>
<tr>
<td>Water transport to user</td>
<td>1,48</td>
<td>2,00</td>
</tr>
<tr>
<td>total costs (€ t(^{-1}))</td>
<td>40,33</td>
<td>52,92</td>
</tr>
</tbody>
</table>

5.6.3 Profitability Analysis

The profitability analysis uses an assumed market price for the produced pellets which is based on their (as received) heating value. Annual revenue from the sale of the annual production amount of pellets is then calculated. After subtracting production costs and annual depreciation the gross profit is found. A tax rate is assumed and the net income is calculated. Finally, annual cash flow generated can be found with which the pay-back period and internal rate of return of the investments can be obtained.
5.6.4 Pay-Back Period and Return on Investment

The payback period of an investment is the amount of time required to regain the value of the original investment. If comparing two or more investments, a shorter payback period is desirable from the perspective of the investor. The payback period $t_p$ is calculated from the following equation in which $C_0$ is the capital investment and $R_c$ is the annual cash flow generated [118].

\[
(12) \quad t_p = \frac{C_0}{R_c}
\]

In its simplest form, used herein, the time value of money, taxes and depreciation are not taken into account in calculation of the payback period.

The return on investment (ROI) is a simple measure of performance used in economic evaluation of projects and is defined as [118]

\[
(13) \quad ROI = \frac{\text{Net annual income}}{\text{total capital investment}} \times 100\%
\]

5.6.5 Net Present Value and Internal Rate of Return

The determination of the net present value (NPV) of an investment has more utility than payback period and ROI because it takes into account the time value of money. The NPV is the sum of the future cash flows ($C_t$) generated by the investment over a series of time periods (t), expressed as present values. The NPV is a function of the financing rate ($i$) and utilisation period (n) of the investment [118].

\[
(14) \quad NPV = \sum_{t=0}^{n} \frac{C_t}{(1+i)^t}
\]

There exists exactly one value of financing rate that will make the NPV equation equal to zero. This value of $i$ is known as the internal rate of return ($r$). It is interpreted as a measure of the highest possible financing rate that could be paid on an investment without loss [118]. Consequently, more profitable investments will have a higher internal...
rate of return than investments of low profitability. Therefore, \( r \) is an economic indicator of attractiveness to determine which of two or more capital investment projects is worthy of undertaking; the one with the largest \( r \) value is the better investment.

In practice, neither NPV nor \( r \) is known so that the solution to the above equation must be found graphically. This is done by estimating \( r \) and computing the sum of all the terms. For a project with a ten year lifetime and initial investment \((C_0)\), the equation to be solved has the following form.

\[
NPV = (-C_0) + \frac{C_1}{(1 + r)^1} + \frac{C_2}{(1 + r)^2} + \frac{C_3}{(1 + r)^3} \ldots + \frac{C_{10}}{(1 + r)^{10}} = 0
\]

5.6.6 Sensitivity Analysis

The base case of the economic evaluation uses the most probable input values for the analysis. Sensitivity analysis is a way of evaluating uncertainty in the base case results. Input values may represent a mean value in a valid range. In the simplest form, sensitivity analysis varies the value of a single input while keeping all other input values constant. The change in the results, due to the change of the single input describes the output’s sensitivity in relation to that input - in other words, how cash flow is affected. The change in the output can be plotted as a function of variation in each input. The purpose is to identify the influence of the input variables on the results.

5.6.7 Inputs Used in Economic Evaluation

Pellet production costs consist of feedstock cost, wages for personnel and the cost of utilities and other consumables during production. Feedstock is used both as the raw material for pellet making and as a combustion fuel for the drying operation.

The properties of torrefied pellets used in the evaluation are those that were determined experimentally to have the most beneficial fuel properties in Paper III (Sample 7). Moreover, data on utility use and thermal balance in torrefied pellet production are taken from Paper IV.
from the scaled-up plant using the same process which produced Sample 7. The corresponding data for conventional pellet production was obtained from [7]. Both production processes use the same annual feedstock amount. The input data for financing, production and pellet properties is given in Table 9.

Table 9. General input data used for economic evaluation (Paper VI).

<table>
<thead>
<tr>
<th>Financing</th>
<th>Torrefied</th>
<th>Conventional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fixed capital investment (M€)</td>
<td>5.60</td>
<td>3.90</td>
</tr>
<tr>
<td>Working capital (M€)</td>
<td>0.70</td>
<td>0.50</td>
</tr>
<tr>
<td>Financed principle (M€)</td>
<td>4.90</td>
<td>3.40</td>
</tr>
<tr>
<td>Rate of financing (unitless)</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Depreciation period (a)</td>
<td>10.0</td>
<td>10.0</td>
</tr>
<tr>
<td>Capital recovery factor (unitless)</td>
<td>0.13</td>
<td>0.13</td>
</tr>
</tbody>
</table>

| Production                          |           |              |
| Production capacity (t a⁻¹)         | 64 000    | 80 000       |
| Feedstock (t a⁻¹)                  | 170 000   | 170 000      |
| Feedstock moisture (%)             | 48.3      | 48.3         |
| Feedstock LHV, ar (MJ kg⁻¹)        | 8.6       | 8.6          |
| Mass yield, dry basis (%)           | 80        | 100          |
| Product volume (m³ a⁻¹)             | 91129     | 123077       |

| Pellet properties                   |           |              |
| LHV, ar (MJ kg⁻¹)                  | 18.28     | 15.80        |
| Moisture content, ar (%)            | 5.0       | 9.0          |
| Bulk density, ar (kg m⁻³)           | 702.3     | 650.0        |
| Energy density, ar (GJ m⁻³)         | 12.84     | 10.27        |
| Durability, as received (%)         | 97.10     | 98.00        |

The prices of utilities used in production correspond to the Dutch market rates in 2004. The unit prices of these items are given in Table 10 and have been obtained from [7] with the exception of nitrogen which has been estimated based on current industrial prices. The amounts of consumables used in production (Paper IV) are given in Table 11.
Table 10. Utilities prices used in the base case (Paper VI).

<table>
<thead>
<tr>
<th>Item</th>
<th>Price</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feedstock/drier fuel</td>
<td>5 € t⁻¹</td>
</tr>
<tr>
<td>Personnel, full time</td>
<td>50 000 € a⁻¹</td>
</tr>
<tr>
<td>Electricity</td>
<td>0.065 € kWh⁻¹</td>
</tr>
<tr>
<td>Natural gas</td>
<td>0.013 € kWh⁻¹</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>0.05 € kg⁻¹</td>
</tr>
<tr>
<td>Water</td>
<td>0.04 € m⁻³</td>
</tr>
</tbody>
</table>

Table 11. Utilities used in production (adapted from Paper IV).

<table>
<thead>
<tr>
<th>Utility</th>
<th>Torrefied</th>
<th>Conventional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electricity (kWh t⁻¹)</td>
<td>216,000</td>
<td>147,000</td>
</tr>
<tr>
<td>Natural gas (kWh t⁻¹)</td>
<td>30,000</td>
<td>0,000</td>
</tr>
<tr>
<td>Nitrogen (kg t⁻¹)</td>
<td>63,750</td>
<td>0,000</td>
</tr>
<tr>
<td>Water (m³ t⁻¹)</td>
<td>0,125</td>
<td>0,000</td>
</tr>
<tr>
<td>Drier fuel (kWh t⁻¹)</td>
<td>825</td>
<td>710</td>
</tr>
</tbody>
</table>

In financing the investments, it was assumed that the working capital available was 0.7 and 0.5 M€ for torrefied and conventional pellet production respectively. A financing rate of five per cent and a financing period of ten years were used. This resulted in the same CRF for both investments (Table 9). The end-of-life value of the investment is assumed to be zero. Labour costs consist of annual wages for five full-time employees. The selling price of energy for the co-firing market was assumed to be 7.30 EUR GJ⁻¹ and a taxation rate Cₜ of 35% was used. The results of the economic analysis are presented in Chapter 6.
5.7 Torrefaction in Pressurised Reactor

The influence of pressure on the torrefaction of wood was investigated in Paper V. Torrefaction was carried out using a custom-built pressurised thermogravimetric reactor (PTGR) which has been described elsewhere [119]. The device is designed for determination of weight changes at pressures up to 10 MPa and temperatures up to 1100°C. The two torrefaction temperatures used in this study were 240 and 280°C and four different pressure values: 0.1, 0.5, 1.0 and 2.1 MPa. Nitrogen was used as inert gas with flow rates being 2.0 L min$^{-1}$ at 0.1 and 0.5 MPa pressure and 3.0 L min$^{-1}$ at 1.0 and 2.1 MPa. For samples torrefied at 240°C, only runs at atmosphere and 2.1 MPa pressure were carried out.

Milled sample material was loaded into a sample holder and placed within the sample lock chamber located above the reactor at room temperature. The milled wood is held in the sample holder between a tube and a screen so that there is a thin layer of sample. The holder is attached to the filament of a microbalance-winch arrangement. A sample holder was not used for wood-cylinder samples which were suspended on a platinum wire of 0.4 mm diameter. The cooled sample lock chamber was continuously flushed with helium gas, keeping the sample cool while the reactor is brought up to temperature. Once the reactor temperature and pressure were stable, the sample carrier was lowered directly into the heated reactor. Reactor temperature was measured by a thermocouple located a short distance beneath the sample. Sample mass and temperature was recorded every two seconds.

Due to the lowering of the sample into reactor the time before the first weight measurement is obtained is approximately 28 s. The sample heating rate has not been estimated. Sample size was 50 ± 0.1 mg when using milled aspen and pine while the mass of single-particle cylinder samples were 90 ± 1 mg. Individual runs showed good reproducibility. The mass measurements using the PTGR are accurate to within at least two significant figures.

The inert gas stream within the PTGR flows upwards, producing a parallel drag force on the sample carrier. The magnitude of the drag

---

7 Manufactured by Deutsche Montan Technologie für Rohstoff, Energie, Umwelt e.V.
force is the difference in the sample carrier mass when weighed in the
gas flow (within the hot reactor) and at room temperature. To correct
data values this mass is added to that measured during PTGR runs.

The higher heating value of samples was calculated using an
empirical relation derived by Friedl et al. from a partial least squares
regression model [120] in which C, H and N are the per cent
composition of carbon, hydrogen and nitrogen in the dry sample. The
authors estimate that the standard error of prediction is 0.36 MJ kg⁻¹.
This relation was derived for different types of plant material.

\[(15)\quad \text{HHV}_{\text{PLS}} (\text{kJ kg}^{-1}) = 5.22C^2 - 319C - 1647H + 38.6CH + 133N + 21028\]

Additionally, the carbon yield of the samples used in the PTGR runs
are calculated using on the following relation using the CHN results.

\[(16)\quad Y_c (\%) = 100 \cdot \frac{\text{carbon in dry torrefied sample}}{\text{carbon in dry untreated sample}}\]

The results from the high-pressure torrefaction study are presented in
the next chapter.
6. Results and Discussion

In this chapter, the relevant results from the papers are presented and their importance with regard to feasibility is discussed. Firstly, the measured properties of produced torrefied pellets are described (Paper III). The discussion of the results draws on some of the key properties of torrefied wood from Chapter 4. Secondly, the environmental impact of torrefied pellet production is presented and compared to conventional pellets (Paper IV). Thirdly, results of the economic analysis are presented and compared to past results (Paper VI).

These first three topics deal with torrefaction of wood at normal atmospheric pressure. The final section in this chapter presents results of wood torrefaction at elevated pressure (Paper V) which, as will be seen, may enhance key properties and industrial production of torrefied fuels.

6.1 Torrefied Pellet Properties (Paper III)

Through research collaboration and international projects, semi-industrially produced pellets made of torrefied materials have become available to researchers only recently. Paper III describes the characterisation of six different torrefied pellet types, and conventional wood pellets for comparison. The torrefied pellets were produced at two European national energy institutes: The Energy Centre of the Netherlands (ECN) and the Spanish National Renewable Energy Centre (CENER). ECN pellets used Finnish feedstock (predominantly whole-tree pine chips and logging residues) while the CENER pellets used Spanish beech stem wood.

The measured and calculated pellet properties included: lower and higher heating value (LHV, HHV); equilibrium moisture content ($M_{emc}$); bulk density ($\rho$); energy density ($\sigma$); durability ($d$) and hardness ($h$). Additionally, these were evaluated on a dry and as-received basis. The summary of properties is presented in Table 12 in which the sample name describes feedstock and torrefaction temperature used - logging residues is abbreviated as Lres.
Table 12. Measured and calculated properties of the seven pellet samples. The sample column describes feedstock and torrefaction temperature and includes conventional wood pellets (Wood Pellet) and logging residue (Lres). Subscript ar and d indicate “as received” and “dry basis” Symbols used refer to (a) 22°C and RH 85% (b) not measured, average of reported values from [22], (c) ΔHHV is based on following untreated HHV values (MJ kg⁻¹): whole-tree pine (19.967), logging residues (20.206) and beech (19.674), (d) 3% wheat flour used as binding agent, (e) pellet properties (bold) used in economic evaluation (adapted from Paper III).

<table>
<thead>
<tr>
<th>Sample</th>
<th>HHV (MJ kg⁻¹)</th>
<th>ρ_ar (kg/m³)</th>
<th>M_ar (%)</th>
<th>M_emc (%)</th>
<th>d_ar (%)</th>
<th>d_a (%)</th>
<th>h_ar (kg)</th>
<th>h_a (kg)</th>
<th>M_2.5 (%)</th>
<th>M_15 (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wood pellet</td>
<td>20.48</td>
<td>678.5</td>
<td>6.67</td>
<td>11.66</td>
<td>98.0</td>
<td>98.2</td>
<td>20.9</td>
<td>19.9 (19)</td>
<td>31</td>
<td>77</td>
</tr>
<tr>
<td>Pine 235</td>
<td>20.80</td>
<td>556.6</td>
<td>7.89</td>
<td>10.60</td>
<td>80.0</td>
<td>69.2</td>
<td>15.1</td>
<td>18.3</td>
<td>33</td>
<td>66</td>
</tr>
<tr>
<td>Pine 245</td>
<td>21.77</td>
<td>633.1</td>
<td>5.49</td>
<td>9.50</td>
<td>92.0</td>
<td>86.6</td>
<td>20.7</td>
<td>19.4</td>
<td>32</td>
<td>53</td>
</tr>
<tr>
<td>Pine 255</td>
<td>21.91</td>
<td>633.8</td>
<td>5.65</td>
<td>9.37</td>
<td>88.2</td>
<td>81.6</td>
<td>18.8</td>
<td>20.3</td>
<td>32</td>
<td>51</td>
</tr>
<tr>
<td>Lres 240</td>
<td>21.59</td>
<td>681.3</td>
<td>7.09</td>
<td>9.41</td>
<td>89.1</td>
<td>84.3</td>
<td>17.8</td>
<td>18.6</td>
<td>32</td>
<td>46</td>
</tr>
<tr>
<td>Lres 250</td>
<td>21.70</td>
<td>643.2</td>
<td>6.99</td>
<td>9.84</td>
<td>86.8</td>
<td>79.1</td>
<td>9.8</td>
<td>10.6</td>
<td>33</td>
<td>50</td>
</tr>
<tr>
<td>Beech 270</td>
<td>21.60</td>
<td>702.3</td>
<td>4.99</td>
<td>8.93</td>
<td>97.1</td>
<td>95.5</td>
<td>19.7</td>
<td>20.4</td>
<td>32</td>
<td>39</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample</th>
<th>LHV_s (MJ kg⁻¹)</th>
<th>LHV_ar (MJ kg⁻¹)</th>
<th>ρ_m (kg/m³)</th>
<th>ρ_a (kg/m³)</th>
<th>σ_s (GJ/m³)</th>
<th>σ_a (GJ/m³)</th>
<th>ΔHHV (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wood pellet</td>
<td>19.17</td>
<td>16.61</td>
<td>633</td>
<td>717</td>
<td>12.97</td>
<td>11.91</td>
<td>0.00</td>
</tr>
<tr>
<td>Pine 235</td>
<td>19.51</td>
<td>17.13</td>
<td>513</td>
<td>573</td>
<td>10.66</td>
<td>9.82</td>
<td>4.17</td>
</tr>
<tr>
<td>Pine 245</td>
<td>20.51</td>
<td>18.28</td>
<td>598</td>
<td>661</td>
<td>13.03</td>
<td>12.09</td>
<td>9.03</td>
</tr>
<tr>
<td>Pine 255</td>
<td>20.65</td>
<td>18.43</td>
<td>598</td>
<td>660</td>
<td>13.10</td>
<td>12.16</td>
<td>9.73</td>
</tr>
<tr>
<td>Lres 250</td>
<td>20.44</td>
<td>18.12</td>
<td>598</td>
<td>664</td>
<td>12.98</td>
<td>12.02</td>
<td>7.39</td>
</tr>
<tr>
<td>Beech 270</td>
<td>20.38</td>
<td>18.28</td>
<td>667</td>
<td>733</td>
<td>14.41</td>
<td>13.39</td>
<td>9.79</td>
</tr>
</tbody>
</table>

The general trend exhibited in the pellet properties is that a higher torrefaction temperature results in a higher heating value, higher energy density and lower equilibrium moisture content. Of the properties determined, the beech sample was observed to have the most favourable properties of a torrefied pellet (as received basis); lowest moisture content (M_ar = 5%), bulk density (ρ_ar = 702 kg m⁻³), lower heating value (LHV_ar = 18.3 MJ kg⁻¹), relative heating value increase (ΔHHV = 9.8%), greatest energy density (σ_ar = 12.8 GJ m⁻³) and highest durability (d_ar = 97.1%). In fact, these were the only tested torrefied pellets which met the minimum level of durability as defined by the standard. These specifications are used in the economic evaluation of torrefied pellet production (Paper VI).
The findings from rainfall and immersion tests were especially revealing with regard to the hydrophobicity of torrefied wood. After an equivalent rainfall of 2.5 mm h\(^{-1}\) (for one hour duration), it was found that all the pellet samples showed the same level of moisture (31% to 33%). The pellets formed an approximate monolayer on the sieves used in this experiment. This configuration corresponds to those pellets lying on the surface of a heap. These results say nothing about the moisture content of pellets within a heap itself. Durability and hardness tests were attempted after rain exposure but (with the exception of one pellet type) this was not possible because the pellets disintegrated. The exceptional pellet was the beech pellet.

After 15-minute immersion in water, a clear correlation between torrefaction temperature and amount of water absorbed by the pellets was observed; the level of moisture retained after immersion was observed to depend inversely on torrefaction temperature. Feedstock type also had an influence. Compared to the 77% moisture found with conventional pellets, torrefied pellets had a moisture content ranging from 39% to 66%. None of the torrefied pellets exhibited hydrophobic behaviour. Even the lowest observed moisture content (39%) would not be satisfactory for outdoor storage of this fuel because of the disintegration behaviour described above.

By considering the measured durability of the three samples of pine pellets, it can be surmised that a torrefaction temperature of 245°C is the optimal one to maximise durability (87%). Consequently, if the heating value increase of the pine pellets could be further improved at a higher torrefaction temperature, durability would likely decrease. This shows how durability is limited by the degree of torrefaction. If a valid trend for all sample types, this suggests that the durability of beech torrefied at higher temperature would be lower.

The Lres 250 sample (logging residue pellets) contained 3% wheat flour as a binding agent. This sample showed markedly low hardness and higher moisture retention compared to Lres 240 which was torrefied at lower temperature. Both these traits may be a result of the binder agent which was itself untreated.

Pellet properties described above showed good agreement with the findings reported by Larsson et al. [81], although in Paper III a wider
range of feedstock types was studied and the pellets were produced from two different reactor designs. Less agreement is seen, however, with earlier work by ECN [7]. The findings from these two studies are compared to those of Paper III in the table below, which also includes the properties of conventional wood pellets.

Table 13. Comparison of pellet properties: conventional wood pellets and torrefied pellets as reported by Larsson [81] and ECN [7]. Symbols refer to (a) from [22] (b) Dry ash-free (c) Lower heating value (LHV) reported (d) calculated using LHV (e) After 15-hour water immersion (f) Based on improvement values reported as decimal fractions (na) not available (adapted from Paper III).

<table>
<thead>
<tr>
<th>Property</th>
<th>Wood pellets</th>
<th>Larsson et al.</th>
<th>ECN</th>
<th>Paper III</th>
</tr>
</thead>
<tbody>
<tr>
<td>HHV (MJ kg⁻¹)</td>
<td>18.8-19.0</td>
<td>22.5ᵇ</td>
<td>20.4-22.7ᶜ</td>
<td>20.5-21.9</td>
</tr>
<tr>
<td>LHV (MJ kg⁻¹)</td>
<td>16.7-17.9</td>
<td>21.0ᵈ</td>
<td>19.9-21.6</td>
<td>17.8-19.3</td>
</tr>
<tr>
<td>ΔHHV (%)</td>
<td>0.0</td>
<td>10.8</td>
<td>15-28ᵇ</td>
<td>4.2-9.8</td>
</tr>
<tr>
<td>Mᵦ (%)</td>
<td>7-12</td>
<td>1-5</td>
<td>1-5</td>
<td>5-8</td>
</tr>
<tr>
<td>ρₑ (kg m⁻³)</td>
<td>560-690</td>
<td>683-713</td>
<td>750-850</td>
<td>557-702</td>
</tr>
<tr>
<td>σₑ (GJ m⁻³)</td>
<td>10.5-13.1</td>
<td>12.3-14.7</td>
<td>14.9-18.4</td>
<td>10.7-14.4</td>
</tr>
<tr>
<td>M₁₅ (%)</td>
<td>na</td>
<td>na</td>
<td>7-20ᵉ</td>
<td>39-66</td>
</tr>
<tr>
<td>h (kg)</td>
<td>17-21</td>
<td>na</td>
<td>28.5-38.0</td>
<td>9.8-20.7</td>
</tr>
<tr>
<td>d (%)</td>
<td>&gt;97.5</td>
<td>82.8-90.3</td>
<td>&gt;97.5</td>
<td>80-97</td>
</tr>
</tbody>
</table>

There are significant differences in observed pellet properties between studies as evident from the above table. The greatest ΔHHV was reported by ECN and its range has a maximum value more than twice that of other studies. The bulk density range of these pellets is also significantly higher than in other studies. Exceptional values are also seen for energy density $\sigmaₑ$ and moisture content after 15-hour (compared to the 15-minute immersion in Paper III) water immersion which is reported to have a range of 7 to 20%. Pellet hardness and durability are qualitatively reported to be very high.
6.2 Environmental Impact (Paper IV)

The main result from the environmental impact study (Paper IV) is the total CO$_2$-equivalent emissions from production and use of torrefied pellets in co-firing. This is compared to conventional pellet production using the same framework and scenario. The main result is shown in Figure 13. The total emissions from the considered production scenario for torrefied and conventional pellets are within 43-45 g MJ$^{-1}$ for electricity production ($\pm$ 4 g MJ$^{-1}$). Additionally, the net energy ratio was found to be 4.7 for conventional and 4.4 for torrefied pellets indicating slightly greater energy payback for the conventional pellets.

In other words, both fuel types produced similar emissions from their production and use in co-firing. The interpretation of this result is that, in environmental terms, no barrier exists in torrefied pellet production and use in co-firing. The use of torrefied pellets in this scenario will generate an emission savings of 77% over fossil coal use in electricity generation. This is slightly less than the 78% savings using conventional pellets. These results also extend to a scenario in which production takes place in western Canada (Figure 13). Despite the much greater distance, North American production has only slight increases in emissions because maritime shipping and rail have much lower emission footprints than the lorry transport used in the Finnish scenario.

Based on the aforementioned facts in Sections 3.5 and 4.3.5 regarding the friability of wood, it follows that co-firing rates using torrefied pellets could be significantly greater than with conventional pellets. This, in itself, enables greater amounts of wood to be combusted and therefore greater emission reductions.
Figure 13. The total emissions from the final energy commodity from the considered production and supply scenario of conventional and torrefied pellets in case of production in Finland (FI) and Canada (CA). The fossil fuel comparator value for electricity from coal is 198 g MJel⁻¹ (Paper IV).

The influence of the fuel’s energy density on emissions can be understood from Figure 14 which shows the total emissions of the two pellet types as a function of lorry transport distance to end user (i.e. no other modes of transport). At a distance greater than 401 km – the point of intersection of the two lines in the figure – torrefied pellets have lower emissions. However, when the transport distance is less than 401 km, conventional pellets have lower emissions. Similar equations can be found for rail transport and maritime shipping, whose corresponding equal-emission distances are 1850 km and 25 700 km respectively.
Figure 14. Total generated CO₂-equivalent emissions from final energy commodity \( (EC_e) \) from conventional and torrefied pellet production as a function of lorry transport distance of product. Assuming only lorry transport, total emissions for torrefied pellet production and use are less at transport distances greater than 401 km (Paper IV).
6.3 Economic Analysis (Paper VI)

Using the input data and equations Chapter 5, the annual costs associated with pellet production were calculated. Financing and depreciation costs were found using Equations 10 and 11 respectively with data from Table 9. Logistics and transport cost are taken from Table 8. Utility costs were calculated using the values in Tables 10 and 11.

The contributions to the total annual production costs and the costs per tonne of produced pellets are presented in Table 14. The annual cost of torrefied pellet production is less (6.11 M€) than that of conventional pellets (7.05 M€). The thermal energy content of produced pellets can be found from multiplying the annual production amount by the lower heating value of pellets (Table 9). This value is 1170 and 1264 TJ for torrefied and conventional pellets respectively. Using the above annual production costs, the specific costs per unit of thermal fuel energy for both pellet types are 5.22 and 5.58 € GJ\(^{-1}\) for torrefied and conventional pellets respectively.

However, if logistics and transports costs are excluded, then the total cost of torrefied production is more (3.53 M€) than conventional (2.82 M€). Without transportation of the pellets, the specific costs of thermal fuel energy for torrefied production is then 3.02 € GJ\(^{-1}\) compared to 2.23 € GJ\(^{-1}\) for conventional. In other words, the shipping distance used in this analysis is required to see the economic benefit of torrefaction (based on energy cost) because at the production site, the cost of the produced fuel energy is lower for conventional pellets.

The specific production costs are more for torrefied pellets; 95.54 € t\(^{-1}\) compared to 88.09 € t\(^{-1}\) for conventional pellets. The latter cost agrees well with the range of other studies for wood pellets in 2004 [32], indicating that the basis of comparison used here is valid.
Table 14. Components of annual production costs of torrefied and conventional pellets (Paper VI).

<table>
<thead>
<tr>
<th>Annual production costs (M€)</th>
<th>Torrefied</th>
<th>Conventional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feedstock cost</td>
<td>0.85</td>
<td>0.85</td>
</tr>
<tr>
<td>Financing</td>
<td>0.63</td>
<td>0.44</td>
</tr>
<tr>
<td>Depreciation</td>
<td>0.56</td>
<td>0.39</td>
</tr>
<tr>
<td>Labour</td>
<td>0.25</td>
<td>0.25</td>
</tr>
<tr>
<td>Utilities</td>
<td>1.24</td>
<td>0.88</td>
</tr>
<tr>
<td>Logistics and transport</td>
<td>2.58</td>
<td>4.23</td>
</tr>
<tr>
<td>Total</td>
<td>6.11</td>
<td>7.05</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Annual production costs (€ t⁻¹)</th>
<th>Torrefied</th>
<th>Conventional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feedstock cost</td>
<td>13.28</td>
<td>10.63</td>
</tr>
<tr>
<td>Financing cost</td>
<td>9.92</td>
<td>5.50</td>
</tr>
<tr>
<td>Depreciation</td>
<td>8.75</td>
<td>4.88</td>
</tr>
<tr>
<td>Labour cost</td>
<td>3.91</td>
<td>3.13</td>
</tr>
<tr>
<td>Utility cost</td>
<td>19.36</td>
<td>11.04</td>
</tr>
<tr>
<td>Logistics and transport</td>
<td>40.33</td>
<td>52.92</td>
</tr>
<tr>
<td>Total</td>
<td>95.54</td>
<td>88.09</td>
</tr>
</tbody>
</table>

Using the selling price of energy for the co-firing market, the resulting market prices (h) of produced pellets are 133.44 € t⁻¹ and 115.34 € t⁻¹ for torrefied and conventional respectively. Using the market prices, the annual sales revenue can be found by assuming all the produced pellets are sold. The revenue, along with other calculations in the profitability analysis is presented in Table 15.
Table 15. Results of profitability analysis for torrefied and conventional pellet production. Adapted from [7] (Paper VI).

<table>
<thead>
<tr>
<th>Description</th>
<th>Operation</th>
<th>Torrefied Pellets (€)</th>
<th>Conventional Pellets (€)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A Sales revenue</td>
<td>$P\cdot h$</td>
<td>8 540 416</td>
<td>9 227 200</td>
</tr>
<tr>
<td>B Production cost</td>
<td>$C_a$</td>
<td>6 114 289</td>
<td>7 047 116</td>
</tr>
<tr>
<td>C Operational Income</td>
<td>A-B</td>
<td>2 426 127</td>
<td>2 180 084</td>
</tr>
<tr>
<td>D Depreciation</td>
<td>$C_0/n$</td>
<td>560 000</td>
<td>390 000</td>
</tr>
<tr>
<td>E Profit before tax</td>
<td>C-D</td>
<td>1 866 127</td>
<td>1 790 084</td>
</tr>
<tr>
<td>F Taxes</td>
<td>$E\cdot \text{tax rate}$</td>
<td>653 145</td>
<td>626 530</td>
</tr>
<tr>
<td>G Net Income</td>
<td>E-F</td>
<td>1 212 983</td>
<td>1 163 555</td>
</tr>
<tr>
<td>H Cash flow</td>
<td>G+D</td>
<td>1 772 983</td>
<td>1 553 555</td>
</tr>
</tbody>
</table>

The revenue from pellet sales generates annual cash flows of some 1.77 M€ and 1.55 M€ for torrefied and conventional pellets respectively. Using these amounts, the pay-back period (Equation 12) of torrefied pellet production is found to be 3.2 years, while for conventional pellets it is 2.5 years. The return on investment (Equation 13) for torrefied pellets is found to be 21.7% compared to 29.8% for conventional pellets. These two economic indicators favour conventional pellet production.

Figure 15 presents the graphical solution to the internal rate of return ($r$) for torrefied and conventional production; the two $r$ values are equal to the x-axis intercept values. The rate for torrefied production is 0.292 (29%) and the rate for conventional production is 0.382 (38%). The value of $r$ also indicates that, for the scenario considered, conventional wood pellet production is a more attractive investment option than torrefied pellets. This is despite the fact that torrefied pellets generate some 30% more cash flow annually.
Comparison of the results here with those of earlier studies, the greatest differences between the costs of the TOP process [7] and the CENER process (Paper IV) are the use of utilities. Firstly, in the TOP process, the amount of recoverable energy (for drying) from the torrefaction gases is greater. The TOP process uses half the amount of natural gas in drying as in conventional production. The TOP process also has a reduced electrical load in the grinding of torrefied sawdust. The CENER process, however, uses more natural gas presumably because the full heating energy of the torgas is not retrievable. The TOP process, therefore, assumes that more energy is technically retrievable from the torrefaction gases.

Another significant factor is the as-received moisture content of the pellets. In their study, ECN uses a value of 3% for their pellets whereas CENER pellets from Paper III were measured to have 5% moisture content. As seen from Equation 5, lower moisture content increased \( \text{LHV}_{\text{ar}} \). The influence of LHV on economic feasibility is discussed in the following section.
6.3.1 Sensitivity Analysis

The sensitivity of the internal rate of return for torrefied pellet production is shown in Figure 16. The intersection of the four lines in the figure corresponds to the value of \( r \) (y-axis) found for torrefied pellet production in the previous section (29%).

![Figure 16. The sensitivity of the internal rate of return (r) for torrefied pellet production to variation of selected base-case variables (Paper VI).](image)

To understand how a change in one of the four presented variables will affect the \( r \) value, consider the second variable in the figure, the LHV of pellets. If the LHV of torrefied pellets could be increased by 10%, the \( r \) value would increase to some 40% (i.e. the y-axis height at which the LHV curve crosses the +10% vertical line in the figure). Since 40% is greater than 38% found for conventional pellets, torrefied production would be the better investment.

The value of \( r \) is most sensitive to capital investment and the LHV of torrefied pellets. A reduction of some 17% in capital investment for torrefied pellet production results in the same \( r \) value for both processes. It follows that further reduction in capital investment
would make torrefied production more viable. The internal rate of return is less sensitive to feedstock price and the bulk density of torrefied pellets. Although \( r \) increases with a decrease in feedstock price, the same is true for conventional pellet production whose \( r \) increases more rapidly. Consequently, conventional production is more favourable even when feedstock cost is zero.

The bulk density of torrefied pellets would need to increase more than 50% (a value of more than 1050 kg \( \cdot t^{-1} \)) to reach an \( r \) value of 38%. Moreover, the product of LHV and bulk density is the energy density of pellets (GJ m\(^{-3}\)). Large independent changes in these pellet properties are not possible because the LHV and bulk density of pellets are interrelated through production, as described in Chapter 4. For example, a greater torrefaction degree increases the LHV but also makes pelletisation more difficult. As a result, the bulk density of the pellets is reduced. Additionally, the shipping costs here depend on fuel volume but there is also some upper limit on mass of goods in a ship. Therefore, even if bulk density could be improved there is some maximum value above which transport savings from torrefied pellets would not apply.

The internal rate of return is highly dependent on the LHV of torrefied pellets and LHV depends on the moisture content (as received) of pellets. Therefore, the importance of a dry fuel is demonstrated in Figure 16. In view of this dependence, the difference in the moisture content here and in the ECN study can be better appreciated.

Summarising the sensitivity results for state-of-the-art pellets, the cost of torrefaction is the major barrier to economic feasibility. If the investment costs can be lowered from the initial estimate by at least 17%, torrefied production can compete with conventional pellets; this corresponds to fixed capital investment cost of 4.6 M€ compared to 3.9 M€ for conventional pellets (Table 9).

### 6.3.2 The Cost of Cutting CO\(_2\)

Despite the differences in the production scenarios of Paper IV and Paper VI, the results from both studies can be combined to estimate the cost of cutting CO\(_2\) emissions through the co-firing of pellets. This
is done by using the market price of pellets, found in the economic evaluation, and the emissions savings of the considered production scenario, obtained from the RED framework. The cost $C_{CO2} (\text{€ t}^{-1})$ is simply the price of replacing coal with pellets $\Delta h (\text{€})$ divided by the mass of CO$_2$ emission reduction $m_{CO2} (\text{t})$.

\begin{equation}
C_{CO2} = \frac{\Delta h}{m_{CO2}}
\end{equation}

The fuel replacement cost depends on the market price of coal, pellets and on their heating values. The mass of CO$_2$ reduction is found from Equation 18.

\begin{equation}
m_{CO2} = S_{el} \cdot EC_{F(el)} \cdot E_{el}
\end{equation}

Here, $S_{el}$ (unitless) is the emission savings, $EC_{F(el)}$ (g MJ$^{-1}$) is the fossil fuel comparator from RED and $E_{el}$ (MJ) is electricity produced from co-firing which can be found from Equation 19. It depends on the mass of pellets required to replace a mass of coal $m_{pellets} (\text{t})$, their lower heating value $LHV_{pellets}$ (MJ kg$^{-1}$) and the efficiency of electrical generation $\eta_{el}$ (unitless) at the power plant.

\begin{equation}
E_{el} = m_{pellets} \cdot LHV_{pellets} \cdot \eta_{el}
\end{equation}

Equation 20 is found by combining Equation 18 and 19 with a kilogram-gram unit conversion factor.

\begin{equation}
m_{CO2} = S_{el} \cdot EC_{F(el)} \cdot m_{pellets} \cdot LHV_{pellets} \cdot \eta_{el} \cdot \frac{1 \text{ kg}}{1000 \text{ g}}
\end{equation}

If a maritime shipping distance of 11450 km is used in the GHG emission balance model of Paper IV, the emission savings $S_{el}$ is found (Equation 8) to be 0.784 (78%) and 0.797 (80%) for torrefied and conventional pellets respectively. Here, the difference between these values is slightly greater than in the results presented in Section 6.2 (Paper IV) because lorry and rail transport are not used - maritime shipping has a smaller emission footprint.

The energy equivalent to one tonne of coal is 1.37 t torrefied pellets and 1.58 t conventional pellets. The price of replacing one tonne of coal with pellets $\Delta h$ is found to be 122.81 € for torrefied pellets and
122.47 € for conventional pellets. The cost of cutting CO$_2$ (Equation 17) is 105 € t$^{-1}$ for both pellet types. This estimate assumes a coal price of 60 € t$^{-1}$ with LHV of 25 MJ kg$^{-1}$ and a 30% efficiency in electricity generation. For comparison, the market price of carbon within the EU Emission Trading System (ETS) has been below 30 € t$^{-1}$ since 2008 and much lower in recent years.

An additional cost factor of co-firing at the power plant is the cost of milling pellets. Considerably more energy is needed to mill wood than coal but the grindability of wood is improved markedly through torrefaction (Section 4.3.5). Energy requirements for milling torrefied and conventional pellets were estimated to be 15 and 50 kWh t$^{-1}$ respectively based on laboratory-scale studies (Paper IV). Therefore, the difference in energy represents a savings in electricity when using torrefied pellets instead of conventional pellets. For the electricity price in Table 10, this savings is 3.81 € per tonne of coal replaced. For torrefied pellets this results in a lower $\Delta h$ price of 119.00 € and a CO$_2$ cutting cost of 104 € t$^{-1}$.

Returning to the differences in the production scenarios, it is noted that the economic study considers saw dust as a feedstock, whereas the GHG emission balance considers logging residues. Therefore the value of the emission savings $S_{el}$ is not necessarily the same for both scenarios. In the case of the latter, however, emissions from the Supply stage of production (Figure 13) are similar for both pellet types so that if this stage were subtracted from total emissions, the two scenarios may be more comparable.

Based on the estimates above, there could be some savings at the power plant when choosing torrefied pellets over conventional ones. The fuels must first be produced, however, and the production costs do not depend on the co-firing rate nor do they take into account emission-based operational savings. These, if they were based on reduction amounts, would be the same for both pellet types (due to similar $S_{el}$). However, if torrefied pellets enable higher co-firing rates than conventional pellets, then larger net emissions (and savings) could be realised.

If the only goal is to cut CO$_2$ emissions, then the extra cost of torrefied pellets may not be an issue. However, there are many other renewable
energy technologies which can cut CO₂ emissions despite not being economically feasible.

6.4 Torrefaction at Elevated Pressure (Paper V)

The studies and findings presented thus far have been exclusively on torrefaction at normal pressure. Paper V is one of the first studies – and possibly the first done with gas flow in the reactor – to determine the influence of pressure on the torrefaction of wood. In this section, the mass, energy and carbon yields of high-pressure torrefaction are presented.

The results of CHN analysis, heating value and the yield calculations on torrefied wood samples using Equations 1, 2, 3, 15 and 16 are presented in Table 16.
Table 16. CHN analysis, heating values and yields of torrefied wood samples (Paper V). Symbols refer to: \( \alpha \) Reactor flow rates were 2.0 L min\(^{-1}\) (0.1 and 0.5 MPa) and 3.0 L min\(^{-1}\) (1.0 and 2.1 MPa), \( \beta \) Calculated by difference from 100%, \( \Omega \) Torrefaction time 14 min, single run only, \( \chi \) Mass yield is for whole cylinder.

| Sample          | T (°C) | \( P^a \) (MPa) | C   | H   | N   | \( Q^b \) | HHV\(_{net}\) (MJ/kg) | \( \Delta \text{HHV}_{net}\) (%) | YC (%) | YM (%) | YE (%) |
|-----------------|--------|-----------------|-----|-----|-----|----------|------------------------|--------------------------|_______|_______|_______|
| Untreated aspen |        | 48.43           | 5.96| 0.11| 45.56| 19.15    | 0.0                    | 100.0                    | 100.0 | 100.0 | 100.0 |
| Untreated pine  |        | 51.23           | 6.36| 0.09| 42.53| 20.43    | 0.0                    | 100.0                    | 100.0 | 100.0 | 100.0 |
| Untreated beech |        | 48.14           | 5.66| 0.04| 46.17| 18.97    | 0.0                    | 100.0                    | 100.0 | 100.0 | 100.0 |
| Aspen           | 280    | 0.1             | 50.33| 5.98| 0.14| 19.89    | 3.9                    | 79.08| 76.8 | 79.08 |
|                 |        | 0.5             | 52.18| 5.72| 0.14| 19.99    | 8.1                    | 79.4 | 73.7 | 79.7 |
|                 |        | 1.0             | 52.87| 5.71| 0.12| 21.02    | 9.8                    | 78.5 | 71.9 | 78.9 |
|                 |        | 2.1             | 53.18| 5.52| 0.19| 21.08    | 10.1                   | 75.5 | 68.7 | 75.7 |
|                 | 2.1\(\Omega\) | 53.37| 5.66| 0.11| 21.24    | 10.9                   | 82.3 | 74.6 | 82.8 |
| Aspen           | 240    | 0.1             | 50.16| 5.98| 0.12| 19.80    | 3.9                    | 95.1 | 92.8 | 95.4 |
|                 |        | 2.1             | 51.18| 5.96| 0.12| 20.33    | 6.2                    | 90.3 | 87.3 | 90.7 |
| Pine            | 280    | 0.1             | 53.39| 5.77| 0.12| 20.73    | 4.2                    | 81.8 | 78.4 | 81.7 |
|                 |        | 2.1             | 56.59| 5.62| 0.12| 22.73    | 11.2                   | 78.3 | 70.9 | 78.9 |
| Beech           | 280    | 0.1             | 52.58| 5.47| 0.18| 20.86    | 9.6                    | 84.2 | 77.1 | 84.5 |
|                 |        | 0.5             | 53.68| 5.74| 0.09| 20.52    | 12.8                   | 80.4 | 72.2 | 81.4 |
|                 |        | 1.0             | 54.23| 5.36| 0.12| 21.56    | 13.3                   | 80.8 | 71.6 | 81.1 |
|                 |        | 2.1             | 56.38| 5.36| 0.11| 22.46    | 18.4                   | 79.0 | 67.5 | 79.9 |
| Beech cylinder, inner | 280 | 0.1             | 51.78| 5.41| 0.00| 20.41    | 7.6                    | 87.7 | 81.7 | 87.9 |
|                 |        | 0.5             | 52.81| 5.37| 0.04| 20.84    | 9.9                    | 88.1 | 80.3 | 88.2 |
|                 |        | 1.0             | 54.12| 5.26| 0.10| 20.53    | 12.8                   | 87.0 | 77.4 | 87.3 |
|                 |        | 2.1             | 56.71| 5.34| 0.05| 22.51    | 18.7                   | 81.7 | 71.1 | 84.4 |
| Beech cylinder, outer | 280 | 0.1             | 51.74| 5.38| 0.04| 20.82    | 7.5                    | 87.9 | 81.7 | 87.9 |
|                 |        | 0.5             | 52.65| 5.37| 0.09| 20.79    | 9.6                    | 87.8 | 80.3 | 88.0 |
|                 |        | 1.0             | 53.85| 5.31| 0.14| 21.36    | 12.3                   | 86.6 | 77.4 | 86.9 |
|                 |        | 2.1             | 55.79| 5.06| 0.17| 22.06    | 16.3                   | 82.4 | 71.1 | 82.7 |
The Influence of Pressure on Torrefaction of Milled Aspen at 280°C

Mass-loss curves of milled aspen, torrefied at 280°C for 30 minutes, at four different pressures are shown in Figure 17. The mass yield of torrefaction varied inversely with reactor pressure. Over the pressure range of 0.1 to 2.1 MPa, the corresponding mass yields ranged from 77 to 69% (Table 16). The carbon content of the torrefied aspen was increased with pressure from 50.3 to 53.2% compared to the original aspen whose C content was 48.4%. The corresponding heating values had a range of 19.9-21.1 MJ kg\textsuperscript{-1}. This represents a $\Delta$HHV range of 3.9 to 10.1% over the range of used pressures.

![Figure 17. Mass fraction of milled aspen versus torrefaction time at 280°C for 30 minutes at four reactor pressures (0.1, 0.5, 1.0, and 2.1 MPa). The mass yield at point A (0.1 MPa) is the same as that at point B (2.1 MPa) but achieved with approximately 50% reduction in torrefaction time (Paper V).](image)

Mass Yield of Milled Aspen Torrefied at 240 and 280°C

Using a torrefaction temperature of 240°C, the mass yields of milled aspen were 92 and 87% at atmospheric pressure and 2.1 MPa pressure respectively (Figure 18). Carbon content was 50.2 and 51.2% compared to the untreated value of 48.4% (Table 16). The HHV after torrefaction was calculated to be 19.9 and 20.3 MJ kg\textsuperscript{-1} at atmosphere...
and 2.1 MPa pressure respectively. The HHV of untreated aspen was 19.2 MJ kg⁻¹. At atmosphere, ΔHHV was found to be 3.9% and increased to 6.2% at 2.1 MPa.

Figure 18. Mass yield of torrefaction using milled aspen as a function of pressure at torrefaction temperatures of 240°C and 280°C (Paper V).

Milled Wood Versus Single-Particle Cylinder

The mass yields of beech, both in milled form and as a single-particle cylinder, are presented in Figure 19. The single-particle cylinders had a mass-yield range of 71-82% while the corresponding range for milled beech was 68-77%.
Figure 18. Mass yields of milled beech and single-particle cylinders as a function of torrefaction pressure at 280°C (Paper V).

Effect of Pressure on Mass-Loss Rate

The first derivatives of the aspen mass-loss curves are shown in Figure 19 with units of mass fraction per second. For all runs, both the initial and peak rate of mass loss depend strongly on pressure. After this initial dependence, which lasts approximately six minutes, the rates are constant and roughly equal being independent of pressure.
Figure 19. Mass-loss rates (with units of mass fraction per second) versus torrefaction time of aspen at 280°C using four reactor pressures (Paper V).

Carbon Content

After torrefaction at 280°C and 2.1 MPa pressure, the carbon content of the samples increased from the untreated state by 4.8% (aspen), 5.4% (pine), 8.2% (milled beech), 8.6% (inner cylinder) and 7.7% (outer cylinder). For aspen at 240°C the increase was 2.8%. In Figure 20, the carbon content as a function of four torrefaction pressures for torrefaction at 280°C is plotted for milled beech, the inner and outer sections of the beech cylinders, and for aspen. The increase in carbon content for beech, both milled and cylinder sections, is linear as seen by fitted linear equations. The difference in C between the inner and outer sections of the cylinders increases with torrefaction pressure. The relation for aspen, however, is nonlinear and can be approximated by two straight lines. In the pressure range of 0.1-0.5 MPa, pressure improves C markedly in aspen. However, at pressures above this range, C increases only slightly.
Figure 20. The relationship between carbon content and torrefaction pressure for the milled beech (×), inner (□) and the outer (△) beech cylinder, aspen (○) samples. The torrefaction temperature was 280°C (Paper V).

Figure 20 has two implications for an industrial torrefaction process. Firstly, if the particle size is large, carbon content does not increase uniformly throughout the sample. This means that HHV of the sample is not uniform throughout. The C of the inner section of a particle improves to a greater extent linearly up to 2.1 MPa – possibly a result of prolonged residence time of vapour-phase products within the bulk of the solid sample. Conceptually, the inner mass of the cylinder can be thought of as being surrounded with a wooden sheath – namely the outer cylinder. Vapour-phase products from within must diffuse through the wood medium which requires time. Consequently, residence time of vapours within the bulk of the sample is extended and is a function of distance from the outer surface. Longer residence time allows more so-called secondary (char-forming) reactions to take place within the solid. The result is a greater carbon yield.

It is not known if such non-uniform torrefaction is commonly seen at atmospheric pressure, although those developing commercial
technologies have indicated it does occur [31]. Torrefaction at 2.1 MPa pressure improved the HHV of the inner wood cylinder by 18.7%, aspen by 10.1%, milled beech by 18.4% and pine by 11.2% (Table 16). The cylinder results are expected to be more representative of torrefaction of wood chips at higher pressure than that observed from milled wood results.

Secondly, the data for aspen shows that moderate pressure (0.5 MPa) is sufficient for most of the improvement in HHV which is a strong function of C. Moreover, only slight additional gains in HHV are achieved above 0.5 MPa. This is seen as the difference in the slopes of these two line segments (Figure 20). The observation that most of the pressure-dependent effect on C and HHV occurs up to 0.5 MPa (aspen) is consistent with those of high temperature cellulose pyrolysis on char yield as described by Blackadder et al. [121] and also seen in the pyrolysis data of Mok et al. [122]. The fact that this is not seen in the beech-wood data may be attributed to the lower cellulose (higher hemicellulose) content of beech.

Atmospheric torrefaction of milled aspen at both 240 and 280°C produced approximately the same ΔHHV (3.9%) despite the temperature difference. However, the mass, energy and carbon yields for the 240°C runs are much higher (92, 95 and 95%) than the latter (77, 80 and 80%). This result indicates that for this wood type, torrefaction at higher temperature is not warranted, since lower temperature produces the same improvement in HHV while retaining a much greater portion of the original feedstock.

Interestingly, Blackadder et al. found that char formation in wood pyrolysis was enhanced by pressure above temperatures of approximately 320°C [121]. This is in fact opposite of what is observed in wood torrefaction where mass yields decrease with reactor pressure. The large amount of volatile matter still present in wood below this temperature, however, is probably the reason behind the difference in behaviour. From Figure 20, it is clear that C is increasing but a large fraction of volatile matter is still present at torrefaction temperatures. This could be the reason behind the difference in behaviour. As temperature is increased, there must be a temperature and pressure at which the behaviour approaches that observed by Blackadder et al. Determining this cross-over point and how it depends on wood species is an interesting topic of future studies.
Based on the results presented, the following question can be asked: Can torrefaction using elevated pressure be beneficial for industrial production of torrefied fuels? The following statements can be made:

- The rates of decomposition reactions in the torrefaction of wood are a strong function of reactor pressure. As a consequence, elevated pressure reduces the torrefaction time required to achieve a given mass yield. A reactor pressure of 2.1 MPa reduced the torrefaction time of milled aspen by 50%.

- Torrefaction at elevated pressure improves the carbon yield of wood compared to atmospheric torrefaction. This was observed from torrefaction at 240°C and 280°C. Consequently, the heating value improvement observed with atmospheric torrefaction is significantly enhanced at high pressure. In the case of aspen, pine and beech samples torrefied at 280°C, ΔHHV values were more than doubled using a reactor pressure of 2.1 MPa. For aspen torrefied at 240°C, ΔHHV was 6.2% at 2.1 MPa compared to 3.9% at atmospheric pressure.

- Higher reactor pressure allows for a shorter residence time to obtain a desired mass yield. Moreover, torrefaction at elevated pressure (with shorter torrefaction time) is preferable. Beech torrefied at 2.1 MPa had carbon content of 56.4% and ΔHHV of 18.4% compared to respective values of 52.6% and 9.6% of beech torrefied at atmosphere.

- After approximately six minutes, the mass-loss rate for all wood samples was independent of pressure, roughly equal and constant. This shows that to minimise the torrefaction time, it is sufficient that the early stages of torrefaction be carried out at elevated pressure.

- Torrefaction at low temperature and high pressure results in significantly greater mass, energy and carbon yields than torrefaction at high temperature and low pressure. For aspen at 240°C and 2.1 MPa, the carbon yield was 92.3% and ΔHHV was 6.2% compared to values of 79.8% and 3.9% from torrefaction at 280°C and 0.1 MPa pressure.
Pressure increases the amount of vapour-phase products formed in the initial stages of torrefaction and therefore shorter residence times can be used. The carbon content of produced chars increases with torrefaction pressure, but the extent to which this occurs depends on particle size and at least to some extent the wood species. Consequently, elevated pressures can be used to tune C content and heating value of different feedstock. However, the reason for the observed difference between the two wood species studied is not known.

From the results of Paper V, it seems likely that the torrefaction process can benefit through the use of elevated pressure and that these benefits will influence feasibility. However, a high-pressure process also brings drawbacks and these should be fully investigated.
7. Conclusions

It is now time to return to the stated objective of this thesis: Is production of pellets from torrefied wood for co-firing in existing pulverised-coal power plants a venture worth pursuing?

Torrefaction of Wood

The relevant chemical and physical changes occurring in wood through torrefaction have been described. Although itself a complex material, wood has a composition which is bound by certain natural limits. Modification of chemical structure is, therefore, limited by the initial constituents of wood. Consequently, key properties of wood with regards to its potential as a co-firing fuel have a finite range. For many of these properties, their range and derived benefits as a fuel in co-firing have been adequately described by laboratory-scale investigations. The most ideal benefits gained from wood torrefaction cannot all be realised simultaneously in a single product. High-energy yields of the process are achieved at the expense of other key properties such as heating value increase, friability, EMC and the ability to pelletise the material.

A good example of this is Loblolly pine whose heating value can be improved by 21% but only under the most extreme conditions of torrefaction (300°C for 80 min.) and with a mass and energy yield of 61% and 73% respectively. Additionally, moderate torrefaction reduces the grinding energy of wood (beech, pine, spruce) by 78-89% in laboratory-scale milling. This reduction seems significant for mill capacity at pulverised-fuel plants. Moreover, the as-received heating values of torrefied wood can be higher due to greater HHV and lower EMC. Through product packaging and proper storage, already used with conventional pellets, torrefied pellets have been shown to have an EMC at least a few per cent lower than conventional wood pellets.

Pellets

Without densification, the energy density of torrefied wood cannot be improved above untreated wood. A moderate level of torrefaction is required in order to observe substantial improvements in fuel properties. However, pelletisation of even moderately torrefied fuels
is challenging and achieving a standard level of pellet durability is not trivial. Typical heating value improvements of torrefied pellets are often half the magnitude of those obtained from extreme torrefaction described above. Furthermore, wood species has an influence on pelletisation success. Low EMC in the feedstock material leads to difficulties in feedstock flow, increases frictional forces in pellet presses and generally makes densification more demanding. Consequently, the energy density of torrefied pellets is limited because the greater the degree of torrefaction, the more difficult pelletisation becomes.

The reduced EMC of pellets made of torrefied wood does not make this fuel hydrophobic. Brief exposure to water from rainfall or emersion results in a high level of moisture retention. There is no evidence that torrefaction, extreme or otherwise, imparts true hydrophobicity to wood. Therefore, torrefied pellets are best handled and stored in the same manner as conventional wood pellets. Finally, that industrial-scale torrefaction could achieve more commercially advantageous results than those observed in the laboratory cannot be assumed.

Environmental Feasibility

Comparison of torrefied and conventional pellet production has shown that there is no environmental barrier, based on energy and emission balance, to their production and use in co-firing. In fact, scenarios with long product transport distances can generate fewer overall emissions than conventional pellets. It is noted, however, that emissions from construction of pellet plants have not been taken into account in this analysis. For an intra-European production scenario, production in Finland and co-firing in Spain, the total CO\textsubscript{2}-equivalent emissions from torrefied pellets was found to be 45 g MJ\textsuperscript{-1} of electricity. A reduction of 77% in emissions could be realised, if this production and end-use scenario would be implemented. Additionally, substantial CO\textsubscript{2} emission reductions appear possible using torrefied pellets if laboratory milling results carry over to industrial scales for direct co-firing.
Economic Feasibility

The economic analysis assumed that the produced pellets had the fuel properties of a demonstrated sample (Paper III) using a demonstrated pilot-scale process and a moderate product shipping distance. Furthermore, it assumed that the production process consumed the inputs described in a scaled-up version of an existing torrefaction pilot-plant (Paper IV).

Three economic indicators (payback period, return on investment and the internal rate of return) show that torrefied pellet production cannot compete economically with conventional production based on the used scenario and properties of state-of-the-art pellets in the analysis. This is true even for transatlantic shipping distances and suggests that any economically viable production scenario will not fulfil the EU energy policy objective of increasing inland sources of energy. Sensitivity analysis of the results has shown that economic feasibility is strongly dependent on the required capital investment of production and the energy density of torrefied pellets. A reduction of about one fifth in capital investment costs would make production economically comparable to conventional pellets. Alternatively, the same could be achieved with an improvement of 10% in lower heating value or 50% improvement in bulk density of pellets. The sensitivity of the internal rate of return to the lower heating value of the pellets highlights the importance of a dry fuel. Therefore, proper packaging, handling and storage of the fuel were implicitly assumed in the economic analysis.

High-Pressure Torrefaction

The use of elevated pressure in the torrefaction of wood has been shown to bring significant benefits to the torrefaction process. The carbon content of torrefied wood increases with torrefaction pressure, enhancing heating value improvements, but the extent to which this occurs depends on particle size and at least to some extent the wood species. Results have also shown that pressure increases the amount of vapour-phase products formed in the initial stages of torrefaction and therefore shorter residence times and/or lower torrefaction temperatures can be used. This benefit may reduce reactor sizes and/or thermal inputs to a torrefaction process.
Future Investigations

The results obtained in this thesis can be used to direct future research in this field. There are clearly five areas where more understanding is required. Firstly, pelletisation of torrefied wood is of great importance. Industrial-scale pelletisation research, in particular, is lacking and unlikely to emerge without relevant large-scale research projects because trials require large amounts of feedstock and are costly. Furthermore, better quantitative methods are needed in pellet making and new equipment designs may be beneficial to have better control over process parameters. Additionally, pellet binding mechanisms need to be better understood and possibly new additives investigated to increase pellet durability. Moreover, such studies must be systematic, starting with raw materials and ending with combustion. As torrefied pellets are an optimised product, research must focus on every stage of the product accordingly.

A second area of required research is on the effective use of vapour-phase products (torgas) in torrefaction systems. As seen from earlier work, assumption about the use of torgas can play a pivotal role in feasibility. Can the trace amounts of fuel present in these gases be utilised effectively? To the author’s knowledge, such studies have yet to be documented.

Thirdly, industrial-scale data on milling torrefied wood and pellets are lacking. As with pelletisation research, this will require adequate funding. Related studies on conveying milled material in a direct-firing system are also of interest.

Research on specific capital investment costs for different torrefaction technologies is an area that would require significant collaboration with technologies developers. Admittedly, this appears challenging but in this way, the most optimal production processes and reactor technologies could be identified.

Lastly, the influence of pressurised torrefaction on the above areas creates another dimension to the research. In particular, research on the influence of pressure on other key properties of torrefied wood should continue at laboratory scales.
Closing Words

From the information presented in this thesis, it should be clear that the overall assessment of feasibility of torrefaction in the context considered is complex. Demonstrated pellet properties and economics suggest that the state of the art does not warrant investment at this time. The absence of environmental barriers in torrefied fuel production combined with a societal concern over greenhouse gas emissions may encourage many to find the technology feasible regardless of the costs incurred. However, other mechanisms for emission reductions may be better suited at lower cost.
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David Agar

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Doctoral Thesis

Laboratory of Inorganic Chemistry

ÅBO AKADEMI

FAKULTETEN FÖR NATURVETENSKAPER OCH TEKNIK

Johan Gadolin

Process Chemistry Centre

15-01

15-02
Mikko Ylä-Sari

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