



Value through Intensive and Efficient Fibre Supply Programme Report 2010–2013





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Programme Report 2010–2013

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FOREWORD

The forest industry has been the central pillar of the Finnish economy for more than a century. Today, structural changes in the pulp, paper and board markets coupled with increased cost pressure throughout the value chain have created a need for radical renewal of the industry. Our northern location gives us a climate handicap in terms of the modest growth rate of our national lifeblood – wood. This and the structural changes in the market can be compensated or minimized only by finding new forestry solutions and by employing the best minds in the industry to develop more efficient pulping processes.

To address these urgent challenges, Finland's first strategic programme for forest sector renewal – Intelligent and Resource-Efficient Production Technologies (EffTech) – was launched in 2008. After this two-year programme, the three-year continuation programme Value Through Intensive and Efficient Fibre Supply (EffFibre) was set in motion.

EffFibre focused on two key areas: wood raw material maximization through increased forest growth and intelligent forest operations, and novel chemistry towards increased pulping yield and reduced process costs. These ambitious targets provided a unique opportunity to show what can be achieved when partners from across the entire national value chain are brought together with an extensive test environment, novel technologies, and top-class national and international research competences.

The programme's forest research results have exciting potential. Although it will take decades to see these results in the field, it is crucial to push this work towards implementation to ensure vital payback in the future. In mechanized forest operations, research into intelligent supportive solutions and decision making systems is opening up new paths to improved efficiency and generating competitiveness and employment already in the short term. The highly promising results achieved in the pulping studies offer exceptional opportunities to improve our position in pulp production and boost the future of this sustainable raw material. This, too, is clear evidence of what can be achieved when there is a clear goal, the right resources, and the right competences and motivation. We now face an urgent need to put results into practice and, economics permitting, to have the courage to make it happen.

Mika Hyrylä UPM-Kymmene Oyj Chairperson of the Programme Management Group

BETTER COMPETITIVENESS THROUGH HIGH-QUALITY FIBRE SUPPLY AND EFFICIENT COOKING

The availability of high-quality feedstock, as well as new resource-efficient chemical pulping technologies, are the driving forces for the future success of the Finnish pulp industry.

The EffFibre programme has focused its research projects on the areas that are most relevant to the forest industry of the future. These include maximizing sustainable raw material reserves and developing cost-effective solutions for pulping processes.

Half of the EffFibre programme was targeted at developing new measures to improve tree growth, at identifying desirable wood characteristics, and at finding new ways to intensify forest management. The practices developed can increase annual cutting volumes by over 40 percent. Moreover, the technological and logistical solutions that have been promoted ensure a more cost-efficient wood supply value chain.

The other half of the programme centred on developing high-yield pulping concepts, as well as deeper understanding of the phenomena involved in pulping, mainly by modelling and simulation.

A broad range of Finnish forest cluster companies have actively participated in the EffFibre programme. The companies involved highlight the results and business relevance in the following chapters.

Leap in plant breeding expertise

The EffFibre programme promoted a major advance in forest plant breeding expertise and methods. Better understanding of genetic mapping and its influence on growth, quality and preservation of wood open up new opportunities for industrial applications, especially in short rotation forestry. In Finland, genetic know-how can also be utilized when selecting better seed and cultivation materials.

Pekka T. Rajala, Stora Enso:

"The results are good, and some of them exceed expectations; Stora Enso will be utilizing the results in its short-fibre wood plantations."

More detailed data on forest resources

The results of the EffFibre programme show that Finland has a sound basis for the sustainable management of forest resources, and a high potential harvesting volume. The scenario work performed in the programme shows that there is significant potential for increasing harvesting volumes by applying intensive forest management. A new method for assessing potentials was developed from data on the development of the forest industry, harvesting volumes and forest resources. Detailed data on forest resources will serve as a basis for forest policy decisions, as well as for recommendations for forest management. The prerequisite for the development of existing production plants and investment in new products is the certainty of a long-term sustainable supply of feedstock from Finnish forests.

Advances in mechanizing forest regeneration and harvesting

The field tests of forest machines carried out in the EffFibre programme were promising. Also various operator training and decisionsupport systems were studied with positive and interesting results and outcomes.

Kalle Einola, Ponsse Plc: "These results are very relevant and contain implementation value."

The pilot projects for mechanizing forest management had encouraging results, especially for improving cost-efficiency. The development of technologies and operational models in harvesting are essential to improve productivity, which will influence the profitability of forestry and the competiveness of the forest industry. Potential annual savings from improved productivity and cost-effectiveness of mechanized timber harvesting are estimated at 40–70 million euros.

New perspective for environmental evaluation

The EffFibre programme scientifically studied environmental indicators in order to determine dynamic forest biomass carbon emissions and sequestration, and the related climate impacts in product life-cycle assessment.

The programme produced dynamic quantification of forest biomasses based on carbon balances and new information about their climate potential. The results give a new perspective when evaluating the environmental benefits of forest products, in comparison to carbon footprint calculations limited to fossil carbon flows.

More economical pulping with new cooking concepts

The EffFibre programme demonstrated that major or even radical improvements in chemical pulping are still possible to achieve, when delignification is better understood. In the research into pulping, the issues of high yield were extended to oxygen delignification concept development. The processes were modelled, showing that novel two-stage kraft oxygen-alkali pulping [KrOxy] can bring significant reductions in wood consumption.

The programme improved understanding of selective delignification, both in softwood cooking and oxygen delignification. The phenomena were described in forms of workable models, allowing the development of future industrial applications. Modelling can be completed in future research and will act as a joint platform for Finnish scientific competence in chemical pulping and in future wood-based biorefineries. The scientific knowledge created will form a basis for future development projects and commercialization of the concept.

Ismo Reilama, Metsä Fibre:

"The results have confirmed that carbohydrate yield in delignification can be remarkably higher than in conventional applications. Metsä Fibre has taken the first step in this field by introducing polysulphide cooking at the Joutseno pulp mill in 2013."

Better understanding of reaction chemistry by simulations

Wood is a very heterogeneous material. The reaction chemistry in pulping is a combination of mass transfer, inorganic and organic reactions. In the EffFibre programme it has been possible to model and simulate the complex reaction matrix. The work will have great significance when new processes and applications based on feedstock wood are developed.

Great value in networking

EffFibre promoted R&D cooperation in forestry and pulping processes between companies and research organizations. The programme combined their different competences in a new way. Although the resources were limited, the quality of the results was good – thanks to the availability of the best scientific and development resources. The whole value chain from research organizations to technology suppliers and forest companies gained great benefit from networking in EffFibre, and the understanding between operators in different phases of the value chain was increased.

The atmosphere of open innovation in networking and cooperation gave a good opportunity for the forest industry companies to evaluate their competences and skills.

Hannu Råmark, Andritz:

"From the technology supplier's point of view, the EffFibre programme served as a platform to find the right partners for development consortiums in the future."

Basis for forest sector renewal

The knowledge created in the EffFibre programme has scientific importance and significant implementation value. The companies are starting consortium development projects in order to refine the results into industrially applicable forms.

The networks created in the EffFibre programme will also be utilized in the development consortiums of the future.

The companies have used the programme to develop methods and technologies that will improve profitability and competiveness, and help in the renewal of the whole sector.

A future challenge is to promote more international and cross-disciplinary networking in joint research and development.

INTRODUCTION

1. Background

The National Research Strategy of the Finnish forest-based sector was published in 2006. To help implement the strategy, the publicprivate partnership Forestcluster Ltd was established in 2007 with the main goal of taking forward the research priorities outlined in the strategy. In 2012 Forestcluster Ltd, then started to expand its activities from research focused on the forest industry towards the bio-based economy. As a result of the metamorphosis the company name was changed to Finnish Bioeconomy Cluster FIBIC Oy. Today FIBIC has activities in three strategic focus areas: Intelligent, Resource-Efficient Production Technologies, Future Biorefinery and Sustainable Bioenergy Solutions.

Research programmes are the core of FIBIC's operations. Their aim is to foster collaboration between end-users, companies and researchers in creating opportunities for research and new business through open innovation and new ways of networking, and to speed the transition from research results to commercial products.

Intelligent and Resource Efficient Production Technologies (EffTech) was the first research programme launched by Forestcluster in 2008. In the second phase (2010-2013), the EffTech programme was divided into two interlinked programmes in order to sharpen the research focus and to diversify the number of research participants. The three-year research programmes, Value through Intensive and Efficient Fibre Supply (EffFibre) and Efficient Networking towards Novel Products and Processes (EffNet) together cover the whole value chain from forest to printing press. The EffFibre programme focused on improving the availability and supply of high-quality raw material from Finnish forests and developing new production technologies for chemical pulping.

The overall objective of EffFibre was to improve the competitiveness of the whole forest cluster by developing novel energy- and resource-efficient forest exploitation and fibre production technologies. The three-year research programme had a total budget of 11 million euros. The Finnish Funding Agency for Technology and Innovation (Tekes) provided 60% of the financing, with the remainder sourced from the participating companies and research institutes.

2. Programme portfolio and goals

The specific goals of the EffFibre programme were to sustainably and cost-efficiently increase the availability and supply of high guality raw material from Finnish forests, and to improve competitiveness throughout the forest cluster by developing new energy- and resource-efficient production technologies and finding ways to reduce the capital intensiveness of the cluster. The key targets were to improve the availability of raw material, to improve the cost-efficiency of the wood supply value chain, and to increase the utility value of domestic raw material. For chemical pulping, the key targets were to utilize the special characteristics of northern wood species to generate a new value chain and to improve efficiency by increasing pulping yield and reducing energy consumption and capital intensiveness. The programme portfolio for the three years included six work packages (Figure. 1).



Figure 1. EffFibre programme portfolio.

3. Management of the programme

The EffFibre programme was administered by a Management Group (MG) comprising representatives from industry and academia. Implementation was headed by a programme manager together with industrial and scientific coordinators. Daily management tasks were performed in each work package (WP) under the leadership of the WP manager.

The main tasks of the Management Group were to supervise the progress of the programme with respect to the objectives of the national forest cluster research strategy and the EffFibre programme plan, and to assess the scientific progress and techno-economic feasibility of the results. In 2011, the MG's main tasks included mid-term evaluation of the programme and organization of discussions with the shareholder companies of Forestcluster Ltd in order to harmonize the EffFibre programme with the companies' research strategies and to define the key focus areas for the second programme period. In 2013, the MG focussed on evaluation of new business opportunities and identifying commercialization and development project ideas.

The Management Group had the following members:

- Mika Hyrylä, UPM-Kymmene, Chairman
- Kalle Einola, Ponsse
- Kalle Ekman, Stora Enso
- · Jari Hynynen, Metla, Scientific Coordinator
- Tore Högnäs, Metsähallitus
- Kari Kovasin, Metsä Fibre, Industrial Coordinator
- Olli Laitinen, Metsä Forest
- Markku Leskelä, FIBIC (Lars Gädda until April 2013)
- Erkki Peltonen, Myllykoski (until December 2011)
- Ismo Reilama, Metsä Fibre
- Leif Robertsén, Kemira
- Hannu Råmark, Andritz
- Pauliina Tukiainen, VTT, Programme Manager
- Päivi Uusitalo, Metso (until October 2013)
- Lauri Verkasalo, Metsä Board (Ari Kiviranta until September 2011)
- Mikko Ylhäisi, Tekes

Detailed project reports and publications are available via the FIBIC portal. Programme seminars have also been held annually bringing together experts from academic and industrial fields and providing a comprehensive overview of the programme's research activities and results.

4. Participants and international cooperation

The EffFibre research programme brought together the leading forest cluster companies and public research groups related to chemical pulping technology, material science, modelling and simulation and forestry research in Finland. Eleven companies and five Finnish universities and research institutes participated in the programme. In addition, research was also subcontracted from external partners.

Industrial partners:

- Andritz
- Kemira
- Metso
- Metsä Board
- Metsähallitus
- Metsä Fibre
- Metsä Forest
- Myllykoski
- Ponsse
- Stora Enso
- UPM

Research organizations:

- Aalto University
- The Finnish Forest Research Institute [Metla]
- University of Helsinki
- University of Jyväskylä
- VTT Technical Research Centre of Finland

International cooperation was built into the EffFibre programme and plays an important role in the development of novel resource-efficient production technologies. Research organizations were encouraged to pursue international collaboration for this purpose with the aim of strengthening the position of Finnish research groups in international communities and opening up new cooperation opportunities. The programme participated in cooperation with six countries: Argentina, Austria, Canada, France, Sweden, and the USA. Close links forged with the international scientific community have been maintained, particularly in the areas of biological science, sustainability research, chemical pulping and modelling research. The international cooperation initiated during EffTech was further broadened in EffFibre, and programme participants have been active in presenting the programme results at several international conferences.

The EffFibre programme was designed to minimize research overlap with related projects and to maximize synergy between other research activities. Many of the programme's researchers were also involved in other related projects, which ensured active information exchange and rapid application of results. EffFibre research groups participated, for example, in the European Community's 7th Framework Programme projects and several COST actions.

The EffFibre programme's core research also supports several industry-driven projects aimed at developing industrial applications. While many of these projects are confidential, active participation of industrial partners within the programme has ensured active information flow, in turn speeding the development process.

FUNCTIONAL GENOMICS OF WOOD FORMATION

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PARTNERS

University of Helsinki

Metla

EFFFIBRE PROGRAMME REPORT

ABSTRACT

The future supply of woody raw material for industrial use will rely increasingly on highefficiency forest production systems using superior trees with defined fibre properties and other quality traits, as well as enhanced growth rate. The main objective of this study was to find and establish ways to improve wood growth and quality in poplar (Populus spp.) and to provide a generic biotechnology tool applicable also to other tree species. The plant hormones cytokinin and ethylene are essential in mediating wood formation. We have identified several genes that regulate various hormone-responsive wood properties and, by enhancing the signalling components of the cytokinin and ethylene pathways, we have been able to significantly increase stem growth rate and modify wood chemistry. We are conducting long-term field trials with the most promising transgenic hybrid aspen lines based on greenhouse experiments. We have shown the benefits of selective seed harvesting from Scots pine seed orchards, improved the chemical analysis methods for heartwood extractive analysis, investigated extractive biosynthesis gene expression using RNA sequencing, and isolated plasma membranes to study lignin monomer transport for eventual use in lignin amount and quality breeding.

The identification of cytokinin and ethylene signalling genes – which regulate various hormone-responsive wood properties – can be used as a novel tool in tree breeding to improve the availability and quality of wood material and thus enhance the competitiveness of the Finnish forest industry. This knowledge can also be transferred from poplar to more widely used tree species for the enhancement of (multi)varietal forestry.

Our results open new opportunities for tree breeding. For example, the lignin component of Norway spruce xylem is an obstacle to the use of spruce wood fibre in pulp production, yet it has high calorific value as a biofuel. Depending on the target application, the information provided by our research can be used for genetic modification of trees and in classical tree breeding to create better wood for specific purposes.

A fast and non-destructive means of measuring stilbene concentration would enable the selection of good quality heartwood material for end users (long-term goal via genetic improvement, short-term goal via grading of existing timber). Information gained from extractive analyses can be used to select for pine seedlings with the genetic potential to produce high quality heartwood as mature trees.

Keywords:

poplar, pine, spruce, cytokinin, ethylene, signalling, growth, monolignol transport, monolignol toxicity, peroxidases, lignin polymerization, heartwood, stilbenes, pinosylvin, heritability, selective harvesting

1. Background

Helariutta's group has shown (in collaboration with Prof. Rishikesh Bhalerao, UPSC, Sweden) that the plant hormone cytokinin is required for normal wood development and that trees with reduced cytokinin signalling are thinner and shorter than regular trees (Nieminen et al. 2008, PNAS 105:20032-20037]. To determine whether enhanced cytokinin signalling increases plant biomass production, Helariutta's team has constructed 25 cytokinin signalling overexpressor lines. The lines overexpress an Arabidopsis CKI1 gene, which constitutively activates the cytokinin signalling pathway. The team has also constructed 12 cytokinin hormone overproducer lines that overexpress an Arabidopsis gene that encodes the cytokinin biosynthetic enzyme.

Kangasjärvi's research group has earlier shown, together with our Swedish partner (in collaboration with Prof. Björn Sundberg's research group, UPSC], that ethylene regulates tension wood formation and that it is a stimulator of cambial growth when directly applied to hybrid aspen stem (Love et al. 2009, PNAS 106:5984-5989). Ethylene responses are mediated through a large family of Ethylene Response Factors ERFs (170 members in Populus trichocarpa genome assembly v3.0] characterized by their conserved ERF domain. To modify wood formation, we selected twenty candidate ERF genes based on their expression profiles in response to ethylene treatments and overexpressed them in hybrid aspen. Under greenhouse conditions, overexpression of several poplar ERFs resulted in enhanced growth rate. Additionally, overexpression of five ERFs also showed clear modifications in wood chemistry, including modified lignin composition. Four ERFs (with two to three individual lines each) were selected for further experiments.

A major research and development goal of EffFibre programme was to gain more precise understanding of the genetic regulation of wood formation to enable the generation of trees with enhanced growth and improved quality. This includes the amount and quality of lignin in trees - an area in which considerable effort has been invested globally, as lignin is an important compound in pulp and paper and sawn timber. Lignin has a high combustion value and hence is also a valuable compound for bioenergy uses. One of the few remaining unknowns in lignin biosynthesis is the transport of monolignols into the cell wall space this important mechanism is the focal point of the Fagerstedt research group.

Among the extractives of Scots pine, the stilbene pinosylvin (PS) and its monomethyl ether (PSM) provide the decay resistance for heartwood timber as well as the defence for living trees against pathogens and pests. The longterm aim is to enhance the supply of high-quality and sustainable materials for the mechanical wood industry. Naturally durable heartwood timber is a realistic alternative to impregnated timber. To achieve this goal, there is need to develop fast optical screening methods to quantify stilbenes, to evaluate the possibilities of selective seed harvesting from seed orchards to obtain durable forest regeneration material, and to develop early testing methods for seedlings.

2. Objectives

The main objective was to find and establish ways to improve wood growth and quality and to provide a generic biotechnology tool that is also applicable to other tree species. The plant hormones cytokinin and ethylene are essential in mediating wood formation.

Key objectives of the Identification of cytokinin and ethylene signalling genes task:

- Generate and screen for transgenic hybrid aspen lines with modified wood chemistry phenotypes with enhanced cytokinin signalling or its downstream genes (Helariutta) and by overexpressing various ERF genes (Kangasjärvi).
- Obtain trees overexpressing cytokinin or ERF signalling genes with enhanced wood formation.
- Investigate the detailed function of various cytokinin signalling or its downstream genes and ERF genes in wood formation.
- Establish long-term field trial in order to evaluate the performance of overexpressed cytokinin and ERF genes under natural growth conditions.
- Determine the hormonal interactions crucial for wood formation.

Key objectives of the Genetic components of lignin formation in wood task:

- Optimize plasma membrane purification methods for Norway spruce xylem tissue. This is the first step towards identification of monolignol transporters or transport mechanism(s) in Norway spruce tissues.
- Obtain new knowledge on the toxicity of free coniferyl alcohol to plant cells.
- As a long-term goal, genetic transformation of Norway spruce with peroxidase genes affecting the monolignol pathway and lignin polymerization. The first Norway

spruce transformants are now being grown in Fagerstedt's laboratory and in the greenhouse.

Key objectives of the Decay resistance and heartwood extractives task:

- Identify co-regulated gene sets in stilbene biosynthesis.
- Identify second-level candidate genes for stilbene biosynthesis.
- Explore the natural variation in pine stilbene synthase STS genes.
- Determine whether one (PST-1) or more (PST-1 to PST-5) stilbene synthase genes are active in pine.
- Participate in developing high-throughput non-destructive analysis methods for heartwood extractives (stilbenes) (together with FuBio programme).
- Investigate the natural genetic variation in heartwood extractives in tree breeding populations and phenotypic variation in a natural stand.
- Evaluate possibilities for selective seed harvesting from Scots pine seed orchards.

3. Partners and their contributions

Research was carried out by University of Helsinki and Finnish Forest Research Institute. Table 1 presents research partners and their roles.

4. Research approach

In order to be able to modify the properties of wood, a full understanding of the factors regulating wood formation is essential. The Helariutta and Kangasjärvi research groups have earlier shown that both cytokinin and ethylene are important hormonal mediators of xylogenesis and that there is a major opportunity to stimulate growth through tree biotechnology. Research on the interaction between cytokinin and ethylene is of great importance, as these two hormones may co-regulate wood formation. We were able to significantly increase the growth rate of hybrid aspen by overexpressing various cytokinin and ERF signalling enhancer genes. Additionally, overexpression of five ERFs (ERF18, ERF21, ERF30, ERF85 and ERF139) resulted in modified wood chemistry.

The objective in studying lignin biosynthesis and its regulation is to gain fundamental knowledge on how the amount and guality of lignin in wood could be altered. We have focussed on the transport of monolignols into the apoplastic space in Norway spruce xylem tissue by isolating plasma membrane vesicles and identifying the putative transport proteins. We have also studied the effects of class III peroxidase genes in Norway spruce transformation experiments using genes that we have cloned previously. If this polymerization step can be modified, we could also alter the lignin content and composition of the xylem by applying the knowledge gained from the Norway spruce breeding programme.

Table 1. Partner organizations and their research roles.

Work package partners	Role of participating organization
University of Helsinki, Department of	Populus trichocarpa genome mining, gene model
Biological and Environmental Scien-	analysis, molecular biology, gene expression analysis,
ces, Plant Biology (Ykä Helariutta,	vector construction and plant transformations. Gene
Jaakko Kangasjärvi, Kurt Fagerstedt,	expression analysis of the transgenic overexpression
Juha Immanen, Kaisa Nieminen, Airi	lines. Growing and phenotyping the transgenic lines.
Lamminmäki, Jorma Vahala)	
	Work with purified plasma membranes and radiola-
	belled monolignols, toxicity tests for monolignols in
	tobacco cell cultures.
University of Helsinki, Department of	Detection of natural variation in the pine PST-1 gene,
Agricultural Sciences (Teemu Teeri,	correlations between phenotypic variation in decay
Tanja Paasela, Kean-Jin Lim]	resistance and molecular variation in PST-1 gene (to-
	gether with Metla), second-level candidate genes for
	stilbene biosynthesis using RNA sequencing.
Finnish Forest Research Institute (Met-	High-throughput screening of stilbenes, natural gene-
la), Northern Finland Regional Unit,	tic and phenotypic variation in heartwood extractives,
Punkaharju Research Unit (Katri Kärk-	selective seed harvesting from orchards.
käinen, Anni Harju, Martti Venäläinen)	

High-throughput screening of stilbenes from a wide range of wood samples is needed to describe the existing variation and to make selection decisions for tree breeding purposes and for the grading of heartwood material. The aim in stilbene quantification is to replace laborious wet chemistry with optical analyses. The concentration of stilbenes measured by UV Raman analysis was compared to the measurements by GC-MS analysis in collaboration with Aalto University School of Science. Increment core samples for the model validation were collected from one progeny trial of Scots pine.

Natural variation in pine stilbene biosynthesis and regulatory genes are the causative reasons for inherited differences in stilbene content and decay resistance of pine heartwood. In this part of the work package, analyses of genetic variation in the induction of stilbene biosynthesis during laboratory-managed abiotic stress were conducted. The goal was to find markers for pine heartwood quality that could be used early in selection. The wide genetic variation in stilbene concentration found among mechanically injured seedlings offers a possibility for early selection of naturally late-expressing trait.

Trees with a genetic capability for high stilbene production could be valuable for practical forestry. The stilbene production of genotypes used to produce commercial seed for forest tree nurseries were therefore screened and, together with seed orchard owners, possibilities for selective seed harvesting were examined. The results indicate that, to obtain a sufficient supply of certified pedigree seed for orchards, a new larger sampling area covering several seed orchards is needed.

5. Results

5.1 Growth rate of wood

Verification and characterization of cytokinin stimulated radial growth

Previously, we have genetically reduced cambial cytokinin levels by engineering transgenic hybrid aspen trees to express a cytokinin catabolic gene, Arabidopsis CYTOKININ OXIDASE 2, under the promoter of the birch CYTOKININ RECEPTOR 1 gene. The resulting transgenic trees showed reduced radial growth, demonstrating the essential role of cytokinins in cambial activity (Nieminen et al. 2008). In contrast, overexpression (by 35S overall overexpression promoter and LMX5 cambial overexpression promoter) of the Arabidopsis CYTOKININ IN-DEPENDENT 1 (CKI1) gene codes the molecule; able to activate cytokinin signalling regardless of cytokinin) resulted in about a 50% increase in stem radial growth, measured as amount of biomass and stem volume, based on results from 14-week-old trees grown under greenhouse conditions. We also observed even better results by overexpressing overexpressing IPT7 in Arabidopsis under LMX5 promoter: the stem diameter increased and height remained the same as the wild type. Based on preliminary results, the cytokinin storage form activator Lonely guy 7 [LOG7] was not shown to have an important role in the regulation of radial stem growth. The most promising CKI1 overexpressor lines were studied under seminatural growth conditions outdoors (sheltered outdoor greenhouse compartment at Viikki campus, with light and temperature close to ambient conditions) during summer 2012.

Verification of enhanced growth of ERF lines under semi-natural field conditions

After initial screening of the ERF overexpressor lines (during autumn 2009 and winter 2010), we selected ERF lines overexpressing four ERFs (ERF18, ERF21, ERF30 and ERF33) for

further study; overexpression of these ERFs enhanced stem radial growth under greenhouse conditions, three of which also showed clear modification, e.g., in lignin composition. Additionally, we have observed that overexpression of ERFs does not disturb normal dormancy cycling in poplar. However, results obtained from greenhouse experiments are not always conclusive and may differ significantly from experimental results obtained in outdoor conditions due to various environmental factors. Thus, a selected population of ERF lines (overexpressing four ERFs) was grown during summer 2011 and 2012 under semi-natural outdoor conditions. Based on the results obtained from the summer 2012 growth trial, we can confirm that overexpression of ERF30 and ERF33 results in an enhanced stem biomass growth rate of up to 25% (measured as dry weight]. In particular, overexpression of ERF30 also increases wood density up to 5%. However, we observed inconsistent results for ERF lines overexpressing ERF18 and ERF21, as the growth rate did not differ from the wild type as in our previous experiments (under both greenhouse and semi-natural outdoors conditions). This highlights the major impact of environmental factors on growth.

Generation of transgenic poplars overexpressing selected genes in Finnish hybrid aspen clone

The origin of the hybrid poplar background clone for the current ERF lines is the Czech Republic, and the clone is not able to fully acclimatize to Finnish winter conditions. We therefore developed and established a transformation procedure for Finnish hybrid aspen clone 51. Currently, we have succeeded to optimize the transformation procedure and have observed several transgenic overexpression lines (both for enhanced cytokinin signalling and ERFs) that are currently maintained and in vitro propagated. For ERF30 and ERF33, 12 and 4 new ERF lines, respectively, have undergone initial greenhouse growth trials. The best lines will be selected for larger scale field experiment.

5.2 Genetic components of lignin formation in wood

Plasma membranes and monolignol transport

Optimization of the isolation methods of Norway spruce and poplar plasma membranes has been finalized, and plasma membranes have been purified from developing xylem of Norway spruce clonal material. Purification of the plasma membrane vesicles has been verified with several enzymatic means and with transmission electron microscopy of thin sections of the isolated vesicles. Analysis of monolignol transporters in the isolated plasma membrane vesicles has been commenced with radioactively labelled coniferyl alcohol. A similar set of experiments has been started with coniferyl alcohol containing a fluorescent label that can be observed with a confocal microscope.

The method for monolignol transport with plasma membrane vesicles has been tested, and separation of the vesicles and the surrounding solution with Sephadex columns has been shown to work. Experiments are in progress.

Experiments and their replicates on the toxicity of coniferyl alcohol to plant cells and the effect of added potassium iodide to remove hydrogen peroxide have been finalized, and the writing of a manuscript has been started. This work has been conducted in collaboration with Adjunct Professor Anna Kärkönen at the Department of Agricultural Sciences of Helsinki University and Professor Stefan Willför at Åbo Academi University in Turku.

Norway spruce tissue culture lines transformed at the Swedish University of Agricultural Sciences with the peroxidase genes previously cloned at Helsinki University have been grown further at our Helsinki laboratory and plantlet regeneration has been achieved. The first plantlets are growing well in unsterile conditions in soil, and after a cold period of five weeks the dormant buds have started to grow. A proteomics study to find any ABC or MATE type transporters in the isolated plasma membranes has been started in collaboration with the Umeå Plant Science Center proteomics unit and Professor Gunnar Wingsle. The results are yet to be published.

The results of the study of membrane transporters with monolignols containing a fluorescent label (kindly provided by Prof. John Ralph, Madison, USA) were inconclusive. This is probably due to the relatively large size of the fluorescent label in comparison to the monolignol molecule. The research focus has therefore been shifted to proteomics instead of the confocal study.

5.3 Decay resistance and heartwood extractives

High-throughput screening of stilbenes: from wet chemistry to optical analyses

During this project, the stilbene chemical analysis (gas chromatography mass spectrometry, GC-MS) technique was improved at the Finnish Forest Research Institute (Metla) laboratory. It was shown that in stilbene quantification this chemical method works better than the tested optical method, UV resonance Raman spectroscopy (UVRRS). Heartwood material for the study was collected from a Scots pine progeny trial. Identical samples were used both in chemical GC-MS analysis (Metla) and in optical UVRRS measurements (Aalto University). There was a clear positive correlation between the stilbene concentrations measured by these two methods, although the correlation was not yet high enough to be reliably applied in large-scale measurements. The estimated genetic parameter, heritability, was twice as large when using the chemical data as when using the data originating from the optical measurements. Thus, the chemical method gives more reliable measurement results. Comparison of these two stilbene quantifying methods emphasizes that it is essential to choose relevant techniques to obtain reliable phenotypic values for genetic analyses. A fast and reliable stilbene quantification method is needed to survey Scots pine breeding material and to grade existing timber.

Natural genetic variation in heartwood extractives

Stilbenes pinosylvin (PS) and pinosylvin monomethyl ether (PMS) are chemical markers for heartwood durability in Scots pine. Phenotypic variation in the concentration of stilbenes and in total phenolics of heartwood was wide in two mature natural stands and in two 40-year-old progeny trials. Growth trait variation was also wide, but none of the measured phenotypic traits of the trees, including heartwood amount and density, correlated with stilbene concentration.

Samples from a mature natural stand (Patasalo] were collected to analyse within-stem variation in heartwood stilbene concentration. Two sampling heights (1.3 m and 5 m) and two sampling positions within each height (outer and inner heartwood) were used. Chemical analyses showed that outer heartwood had a higher concentration of stilbenes than inner heartwood, and that the lower stem had higher concentrations than the upper stem. Moreover, the concentration of stilbenes measured from different parts of the heartwood correlated strongly with each other. For example, if a tree had high concentration of stilbenes in outer heartwood at a height of 1.3 m, it also had a high concentration in outer heartwood at 5 m compared with other trees. In vitro decay testing of the outer heartwood samples from the two measured heights (1.3 m and 5 m) showed that samples with high density and rich in stilbenes had lower mass loss due to the cellar fungus Coniophora puteana compared to lighter and less extractive-rich samples.

Estimated genetic parameters predict remarkably higher genetic gains for stilbene content in heartwood than for tree growth traits. The genetic parameters were assessed for material originating from the two 40-year-old Scots pine progeny trials. Although the genetic correlation between chemical characteristics and growth traits was slightly negative indicating opposite directions of selection, strategic tools can be applied in tree breeding to address traits with negative genetic correlations. On the whole, the results are very positive for the purposes of traditional forest tree breeding. However, the present tree breeding programme does not include chemical quality traits of heartwood as target traits for quality breeding.

Selective seed harvesting from Scots pine seed orchards

According to our study, selective seed harvesting from seed orchards would be a realistic way to obtain seedlings with high genetic potential to produce durable heartwood as mature trees. We analysed the stilbene content of the heartwood of clones growing in two seed orchards managed by SiemenForelia. In addition, their 40-year-old progenies growing in a field trial were evaluated. Remarkable clonal repeatability in stilbene concentration and total phenolics was found among the seed orchard clones. When the heartwood of the seed orchard clones and their progenies were compared, a highly significant correlation in the concentration of stilbenes pinosylvin (r=0.83) and pinosylvin monomethyl ether [r=0.60] as well as in total phenolics (r=0.50) was found (Figure 1). The results were published in the Canadian Journal of Forest Research in October 2011. A fast and reliable stilbene screening method would enable seed orchard clones to be analysed for heartwood chemical quality, thus enabling more effective estimation of possible selective breeding gains.

Stilbene biosynthesis

Induction of stilbene synthesis was analysed in 6-week-old pine seedlings under ultraviolet induction. The stilbenes pinosylvin and pinosylvin monomethyl ether could be extracted from plantlet needles (analysed on TLC plates) 24 hours after induction. Induction of STS stilbene synthase gene transcription appears to be very rapid, taking place within a few hours. The induction is independent of translation, indicating that the factors required for STS transcription are preformed in pine cells. Moreover,



Figure 1. Heartwood pinosylvin (PS) content and heartwood density correlation between mother trees grafted in seed orchards and their progenies growing in a field trial (published in Partanen et al. 2011).

inhibition of translation (with cycloheximide) itself caused induction of the STS gene. This may be a sign of a labile repressive regulator acting on the pathway. Increasing phosphorylation level of proteins in the cells with a phosphatase inhibitor cocktail also strongly induced STS expression, suggesting that an activator of STS transcription requires phosphorylation, or that an inhibitor requires dephosphorylation. The plant hormones ethylene and jasmonic acid together (but not separately) induced STS. The promoter for the STS gene PST-1 was analysed in tobacco cells and seems to contain an element that slows gene expression - an observation that is in good concordance with the effect of cycloheximide on STS transcription.

In order to find a suitable window for investigating genes induced during heartwood formation, sampling of increment cores throughout the growing season was conducted during summer 2010. The dry zone (heartwood) and wet zone [sapwood] boundary was marked on the increment cores, and laboratory inspection under UV showed blue fluorescing pinosylvin to be present throughout the dry zone with the exception of the outermost annual growth ring. RNA was extracted from annual growth rings throughout the increment cores and STS transcription was observed nearly exclusively in the outermost ring of the dry zone (with no signal in the inner annual rings of the dry zone and a low signal in the sapwood growth rings). This suggests that the main site of stilbene biosynthesis is just inside the dry/wet boundary, which can be defined as the transition zone.

The transition zone showed preferred STS expression throughout the growing season (May to September), with the sharpest pattern in the June and July samples. Sampling for transcriptome analysis with RNA sequencing was carried out from low and high extractive trees in June 2011. Monthly increment core samples were collected throughout the year, from March 2011 to February 2012. These samples are stored at -80°C for possible later analysis.

To identify genes that are transcribed differentially (and correlated with STS transcription) in seedlings with different genetic capability of stilbene biosynthesis under induction and heartwood formation, we prepared samples for transcriptome analysis. Stems of 5year-old pine seedlings were wounded by drilling, and tissue samples were collected at time points 0, 1, 24 and 96 hours. Needles of 6week-old seedlings were exposed to UV-C and samples were collected at time points 0, 2, 6 and 24 hours, or to cycloheximide and sampled at 0, 2, 8 and 24 hours. 40-year-old trees were sampled for their sapwood and transition zone to heartwood. For each time point or developmental stage, four biological replicates were analysed, representing trees or half-sib families with both low and high inherited capacity for stilbene formation in the heartwood [except for cycloheximide treatment, where only two replicates were analysed). Sequencing yielded 5-50 million 50-75 bp SOLiD sequence reads per sample. The read depth is sufficient for all samples.

Before the transcriptome reads were mapped on published pine gene lists, an analysis of gene expression across all pine genes was carried out by individually mapping the four known genes involved in stilbene biosynthesis in the libraries (PAL, 4CL, STS and PMT). Of these, we monitored the strong STS induction during sample preparation, finding it to have very similar high induction also in the digital transcriptome analysis. Interestingly, all four genes show induction (albeit at different levels] only during heartwood formation, while after wounding or UV-C, the genes PAL, 4CL and PMT are either constitutively active or practically silent. This may be indicative of as yet uncharacterized gene families with different members having different roles. By searching for methyl transferases that correlate with STS expression, we found one gene candidate (TC197639) that is induced in the different stress or developmental contexts very similarly to STS. We are currently isolating this gene in full length to test whether the enzyme it encodes catalyses pinosylvin methylation.

Likewise, we have investigated sequences similar to 4CL, the enzymatic step before STS. The published 4CL seems to have a role in lignin rather than stilbene biosynthesis. Pine stilbenes (pinosylvin and its monomethyl ether) lack the hydroxyl group at position 4 of the aromatic ring. Accordingly, the 4CL related enzyme should have a slightly modified active site. We have good candidates for this type of gene, which are induced similarly to STS. In addition, methyl transferase genes are being isolated in full length and tested for the enzymatic activity they encode. In summary, we have found that two stilbene biosynthesis genes are misannotated in literature, and we have identified better candidates.

Transcriptome reads have all been mapped to published pine gene sequences [http:// compbio.dfci.harvard.edu/cgi-bin/tgi/gimain. pl?gudb=pine]. There are indications that high and low stilbene half-sib families have differences, although statistical proof is still lacking. Identification of genes whose expression is correlated in all induction conditions will be published during 2014.

6. Exploitation and impact of results

Conventional tree breeding is an effective yet extremely lengthy means of gaining more productive genotypes. We have established generic tools to significantly speed up this process through tree biotechnology. Cytokinin and ethylene are plant hormones that mediate wood formation. We have identified several genes that regulate various hormone-responsive wood properties and, by enhancing the performance of these genes that control hormonal levels and/or the regulation of target genes (e.g. by ERFs), we are able to stimulate the growth rate and chemistry (ERFs) of wood. To our knowledge, cytokinin and ethylene biotechnology have not been applied to any plant species so far. Thus, our research may provide novel tools to enhance the growth rate of trees and to modify wood properties. This opens up unique opportunities for improving the competitiveness of the Finnish forest industry. Additionally, these biotechnologies could be used to achieve maximal wood productivity per hectare, potentially further reducing the need to exploit natural forest resources.

The deliverables include fundamental research results on the transport mechanism of monolignols. This information can be used in the breeding of Norway spruce to produce trees with desired lignin quality or quantity. Combined with previous knowledge on lignin biosynthesis in deciduous forest trees, targeted modification of lignin amount (and quality) can also be extended to rapidly-growing bioenergy tree species.

Trees with a genetic capability for high stilbene production (i.e. high extractives content) could be valuable for practical forestry with the aim of producing improved wood material for end users – in the long term via genetic improvement and in the short term via grading of existing timber. This study lays the groundwork for improved grading and allocation of timber to appropriate and profitable applications through the description of phenotypic and genetic variation in heartwood characteristics. Genotypes with a capability for high stilbene production would be more resistant to pests and pathogens as living trees and at mature age would contain high quality heartwood for the production of durable timber. It is therefore highly beneficial to screen the stilbene production of genotypes used to produce commercial seed for forest tree nurseries.

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POTENTIAL AND FEASIBILITY OF INTENSIVE WOOD AND BIOMASS PRODUCTION

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ABSTRACT

This comprehensive scenario study identified significant potential to increase raw material supply from Finnish forests through the use of more intensive management practices, increasing annual wood and biomass removals by ca. 50% compared with current removals in a sustainable manner and with no marked decrease in forest carbon sequestration. Additionally, the target supply [13 mill. m³] of energy wood can be met by recovering only small-size stemwood, logging residues and stumps. Intensification of forest management requires significant investments in silviculture, including a doubling of the forest area treated annually with precommercial thinnings. On the other hand, intensive management would provide annually over 12,000 jobs in the wood supply chain compared to the current ca. 8,000 jobs supported. Intensive forest management was shown to be 50% more profitable than businessas-usual management. However, there is large regional variation in profitability due to differences in climate, production potential of forest sites, and existing forest structure. The potential export value of forest industry products in the intensive scenario were estimated to be over 10 billion euros, an increase of more than 2 billion euros compared to the current value. The importance of tree growth rate on wood fibre properties, the thermomechanical pulping (TMP) process and paper properties was evaluated, and environmental indicators were developed for assessing the environmental performance of forest products. Intensive forest management practices were also demonstrated and tested in practice by Metsähallitus. Research results were disseminated by establishing demonstration sites and providing decision support tools for assessing forest management alternatives.

Scenario analysis showed the long-term potential of Finland's forests as a source of renewable raw material for the forest industry by providing research-based knowledge and decision support regarding feasible measures to cost-efficiently and sustainably intensify biomass supply. Tools for environmental communication in forestry and forest-based industries were improved by developing new research-based methods for assessing the environmental performance of biomass supply and forest products. The decision support tools and demonstration material, which support decision making in forest management planning, are of considerable benefit to the forest industry in promoting management practices that secure the availability and sustainability of wood at competitive cost.

Keywords:

forest management, forest growth and yield, profitability, scenario analysis, decision support, wood properties, environment, indicators, carbon, water, land use, biodiversity

1. Background

Productive domestic forest resources and a competitive wood supply are crucial to the vitality of the Finnish forest cluster. Sustainable availability and cost-efficient supply of domestic raw material are prerequisites for the forest industry maintaining production in Finland. Although domestic wood and biomass resources are currently abundant, the challenge is to improve the availability of raw material and the cost-efficiency of the wood supply value chain, and to increase the utility value of domestic raw material. The core challenge is to ensure that Finland's domestic wood and biomass supply is competitive with fast-growing plantation forests in tropical and subtropical countries.

The operating environment for industrial wood supply in Finland will become increasingly challenging in the near future. Due to the structure of Finland's forest resources, an increasing proportion of domestic wood and biomass will be harvested in the future from stands that are expensive to harvest and that offer lower removals per unit area. The proportion of more cost-efficiently harvestable final felling stands will decrease from ca. 70% to 50%, while the proportion of removals in less cost-efficiently harvestable forest areas, namely thinning stands and peatland forests, will significantly increase. Furthermore, the ongoing trend in the structure of private forest ownership is likely to lead to increasing numbers of forest owners and decreasing average size of forest holdings. In addition, many forest owners base their forest management on values other than wood production, leading to lower yields. The abovementioned trends in the industrial wood supply operating environment require new ways of thinking and novel solutions. One solution is to focus intensive, cost-efficient and sustainable management and harvesting in forest areas where the conditions for commercial wood and biomass production are most favourable.

The potential and methods of intensive forest management to increase domestic wood and biomass production sustainably are relatively well known due to extensive Finnish forest research, which has long traditions and is internationally recognized to be of high scientific quality. The first stage of the EffTech programme tackled the issue of raw material availability. In the "New value chains" project, a scenario analysis addressing spruce and pine wood supply chains in Southern and Central Finland indicated that wood and biomass production can be markedly increased in a cost-efficient and sustainable manner by applying intensive forest management practices. The crucial factors for successful intensive forest management are artificial regeneration by planting or seeding using high quality planting stock, intensive silvicultural practices throughout the rotation, and an efficient wood procurement chain.

Increasing the use of domestic wood by 25% is one of the strategic goals of the Finnish Forest Cluster. This target can only be achieved by cost-efficiently and sustainably intensifying the supply of forest-based raw material.

2. Objectives

The overall objective is to identify and develop sustainable and feasible measures to improve the competitiveness of the Finnish wood supply chain. The focus is on assessing and developing operations models for intensified raw material supply.

Specific objectives are to:

- Identify the most feasible production and raw material supply strategies for utilization of Finnish forest resources by:
- a. Assessing the potential of intensive management of Finnish forest resources

to provide high quality raw material for the forest industry in a sustainable, costefficient and environmental-friendly manner.

- b. Comparing the impacts of intensive wood supply chains with less intensive management and procurement alternatives.
- 2. Increase the environmental competitiveness of forest products:
- a. Improve and develop environmental communication.
- b. Develop scientifically-grounded methods of evaluating key environmental indicators such as biogenic carbon balance, water footprint, land use change and biodiversity.
- 3. Demonstrate large-scale implementation of intensified forestry in practice and develop practical operations models.

3. Partners and their contributions

The research was carried out jointly by research organizations and Finnish forest cluster companies. Table 1 presents research partners and their roles.

 Table 1. Partner organizations and their research roles.

Work package partners	Role of participating organization
Finnish Forest Research	Management of work package. Coordination and participation
Institute (Metla)	in Tasks 1 (Scenario analysis), Task 2 (Consequences of intensive
	management on forest resources and on the availability of forest-
Vantaa, Rovaniemi and	based biomaterial), Task 5 (Dissemination).
Joensuu Units	
VTT Technical Research	Coordination and participation in Tasks 3 (Controlling raw material
Centre of Finland	quality throughout the value chain) and Task 4 (Environmental
	performance of forest products).
Metsähallitus	Participation in Task 1 (Scenario analysis) and Task 5 (Dissemination).
Metsäliitto Cooperative	Participation in Task 1 (Scenario analysis) and Task 5 (Dissemination).
Metsä Board Ltd.	Participation in Task 4 (Environmental performance of forest
	products]
Myllykoski Ltd.	Participation in Task 4 (Environmental performance of forest
	products]
UPM-Kymmene Ltd.	Participation in Task 4 (Environmental performance of forest
	products)

4. Research approach

Scenario analysis

A national-scale scenario analysis was conducted to assess future alternatives for the utilization of forest resources. The scenarios included business-as-usual, two scenarios of intensified wood and biomass supply, and a scenario of decreased raw material use by forest industries. For each, a specific set of forest management schedules was developed. Calculations of the future development of Finnish forests over a 100-year period were then made based on national forest inventory sample plot data. The calculations were performed using the MOTTI stand simulator.

Consequences of intensive management on forest resources and on the availability of forest-based biomaterial

Each scenario was analysed from different viewpoints. Raw material production was estimated by analysing the growth and yield of wood and biomass and the development of forest resources. The profitability of the forest management alternatives from the point of view of different agents was also analysed.

Controlling raw material quality throughout the value chain

The impacts of the alternative scenarios on raw material properties were addressed with a focus on pulping and papermaking processes. The behaviour of fast-grown wood in the TMP process was studied in a pilot-scale refiner. The effect of raw material/TMP quality on the behaviour of furnish in the papermaking process was also examined in realistic conditions. The behaviour of fast-grown wood in the new alkaline delignification process being developed in KrOXy project was also studied.

Environmental performance of forest products

In addition to studying carbon footprint indica-

tors, an evaluation of different approaches for including the forest carbon cycle in forest product life cycle assessments was carried out. The impacts of the different forest management options on forest carbon sequestration and emissions were then studied based on the simulation results obtained in scenario analysis. Finally, based on the conclusions of the evaluation, the most appropriate method for including forest carbon stock impacts in product LCA and in multi-product systems was tested.

The forest sector is a vast water consumer, both directly in its industrial operations and indirectly in tree growth for wood production. Water footprint (WF) may therefore become a critical indicator for the forest sector as a measure of sustainability and comparative environmental performance, as well as a tool for assessing water-associated business risks and communicating with customers and other stakeholders. The objectives of the water footprint study were to gain better understanding of whether current WF methods accurately reflect water use in Nordic forestry, and, if necessary, to promote more robust methods for measuring the water sustainability of the forest sector.

Life cycle assessment (LCA) is widely accepted as a holistic methodology for the environmental impact assessment of products and services. A framework for the inclusion of landuse impact assessment and a set of land use impact indicators has been recently proposed for product LCA. In Task 4, the proposed methodology and a set of relevant indicators was tested for Scandinavian managed forestry. A comparative case study was conducted for bioenergy from wood, agro-biomass and peat, and sensitivity to forest management options was analysed.

Potential biodiversity indicators were sought from the literature, and a set of indicators (Biodiversity.fi) was chosen to be discussed and evaluated with stakeholders and experts. The indicators were evaluated using scientifically grounded criteria. The list of indicators selected by this process was complemented with LCA biodiversity indicators.

LCA is a well-established methodology that captures the environmental impacts of a product or process from raw material procurement to production, use and end-of-life. LCA assesses the potential environmental impacts of product systems in accordance with the stated goal and scope. The method used in the present study is compliant with life cycle assessment ISO standard 14040-14044. To calculate climate impacts, indicators with a 100year timeframe (GWP(bio)-100) that represent Finnish forest biomass were used. These indicators depend upon the studied time interval and the lifetime of a product. Biomass carbon stored in the products and the timing of emissions are taken into account in the calculations in addition to traditional fossil carbon footprint calculation. For freshwater eutrophication and marine eutrophication impacts the ReCiPe methodology for life cycle impact assessment [LCIA] was used. Water emission data for nitrogen (N), phosphorus (P) and suspended solids [SS] emissions from forest management operations in the alternative scenarios was provided by the Finnish Forest Research Institute [Metla]. The framework for the case studies was the whole forest product value chain from forest to end use.

Dissemination

The dissemination task involved activities aimed at sharing information on intensive forest management among different stakeholders. In order to demonstrate intensive forest management implementation in practice, demonstration sites in a large intensively managed forest area were established in state forests in cooperation with Metsähallitus. The objective was to put into practice up-to-date research-based knowledge and methods to increase wood and biomass production and improve raw material availability in a cost-efficient and sustainable manner on a large-scale. As part of this effort, a network of demonstration stands showcasing different intensive management practices was established. Information related to the demonstration sites was prepared (treatment description, measurements and predictions on posters and websites). Decision support tools for forest management as well as forest owner's guides for intensive management were also prepared.

5. Results

5.1 Impacts of intensive management on forest resources and on the availability of forest-based biomaterial

5.1.1 Methodological development

First, the following forest utilization scenarios were jointly agreed and defined by the work package partners.

1. Business as usual (BAU)

- Current situation and ongoing trends in the forest sector are assumed to occur as expected.
- b. Prevailing forest management and utilization will be applied in the future:1. volumes of silvicultural operations remain at current levels.
 - 2. utilization rate of commercial forests remains at current level:
 - annual cuttings equal to 78% of sustainable allowable cut

2. Active forest sector and intensive biomass production (INT)

 a. Vital forest and bioenergy industry – increasing demand for domestic raw material b. Intensive forest management measures are widely applied:

1. intensive regeneration and young stand management practices

- 2. early and intensive commercial thinnings
- & short rotations
- 3. fertilization

4. biomass recovery in final fellings (logging residues and stumps)

3. High quality raw material production for forest industry and bioenergy (QLTY)

- a. Increasing demand for high quality raw material for the forest industry (especially wood products)
- b. Forest management for combined energy wood and timber production
- c. Wood quality aspects emphasized in forest management (higher density of young stands, quality thinnings and longer rotations)

4. Decreasing forest industry operations – increasing non-material services (EXT)

- a. Decreasing pulp industry volumes decreasing demand for industrial wood
- b. More emphasis on forest externalities and carbon sequestration
- c. Extensive forest management is applied:1. 25% of forests treated according current silvicultural practices

2. 25% of forests left completely unmanaged

3. 25% of forests: only regeneration and final felling

4. 25% of forests: regeneration, one thinning (energy wood) and final felling

In addition, the WP partners also agreed on key calculation parameters to be applied, such as prices of timber assortments, applied commercial wood and bioenergy wood prices, forest management unit costs and interest rates. For future predictions of forest resources under each scenario, forest management schedules specific to each scenario were constructed and entered into the MOTTI stand simulator, which was used for simulations of forest development over a 100-year period. National forest inventory data served as the initial data for the simulations. The current forest resources were determined based on data from a network of over 46,000 inventory sample plots across Finland.

5.1.2 Main results

The scenario calculations clearly showed that Finland's forest resources have significant production potential. However, in order to intensify their utilization and increase wood and biomass production, more intensive thinning programs and shorter rotations should be applied. Accordingly, there is a need for increased investment in silviculture, particularly young stand management.

The scenario analysis shows the output if the given management strategies are adopted. The scenarios as such are not management plans or future predictions.

The main results, classified according to each viewpoint applied in the analysis, are given below.

Wood and biomass removals

Intensive management would enable a sustainable increase in annual removals of ca. 50% (INT & QLTY) compared with current levels. Annual removals could be increased to 84–88 million m³, of which pulpwood and logs would account for ca. 75 million m³ and energy wood 10–13 million m³, thus also meeting the target production level for energy wood. The removals for business as usual scenario (BAU) are close to those of extensive management scenario (EXT) (Figure 1). The results reveal that current level of harvested removals represents rather extensive use of forest resources.



Figure 1. Mean annual removals per forest management scenario between 2010–2110 compared with realized annual removals between 2001–2010.

Development of growing stock

Intensive management ensures sustainable long-term wood and biomass production despite increased removals. However, intensive management leads to smaller standing volumes of growing stock due to shorter rotations when compared with current forest management.

Carbon sequestration

Intensive management aiming at high quality raw material production (QLTY) allows an increase in removals of 50% without a marked decrease in carbon sequestration to forests compared to the current situation. The extensive management scenario (EXT) resulted in a ca. 40% increase in carbon sequestration compared to intensive management. However, in the long run, intensively managed forests provide more efficient carbon sequestration than extensively managed forests. The climate impacts depend on the use of the removed carbon.

Management practices

Intensification of forest management requires significant investments in silviculture. In the intensive management scenarios, the annual area of precommercial thinnings is 2–3 times larger compared to the current level (Figure 2). Furthermore, shorter rotations in intensive forestry increase the annual area of forest regen-



Figure 2. Mean annual area per silvicultural treatment 2010-2110.

eration and site preparation. In terms of work contribution, intensive forestry would represent an increase in annual person years from the current the level of 1,500 to over 2,000.

In order to implement the required increase in thinning frequency and regeneration cutting area, around 30% more forestry machines would be needed. Accordingly, labour demand would increase from the current 5,500 to ca. 8,000 person years.

Profitability

The net present value of future incomes over the next 100 years at 3% and 4% interest rates was applied as a measure of profitability. Applying current wood prices and management unit costs, intensive forest management proved to be 50% more profitable than business-as-usual management. At the national level, scenarios with intensive forest management (INT, QLTY) result in a 50% increase in mean annual gross stumpage earnings compared to current levels, i.e. from EUR 1800 million to EUR 2800 million.

There was, however, a large regional variation in profitability due to differences in climate, production potential of forest sites, and current structure of forests between provinces (Figure 3).

5.2 Controlling raw material quality throughout the value chain

Pilot-scale refining trials were performed to determine how the growth rate of wood affects the energy-quality relationship of mechanical pulping both in normal TMP conditions and in energy-saving RTS conditions (high temperature, high rpm). The mechanisms were studied by measuring temperature profiles along the plate gap radius, and by taking pulp samples from different radii of the plate gap.

Unexpectedly, there was no clear difference in energy consumption in refining (TMP and RTS) slow- and fast-growth wood in the pilot refining trials. This was due to only minimal differences in wood properties (fibre length, basic density) of the samples used, despite a distinct difference in growth rate. There was no difference in tensile strength, but light scattering and brightness were somewhat better for pulps made from fast-growth wood. The temperature along the refiner plate gap radius was only slightly higher for slow-growth than for fast-growth wood chips, and no conclusions on, e.g., the



Figure 3. Present value of net income by forestry centre 2010-2110 (interest rate 3%).

packing of pulp in the plate gap can be drawn.

The effects of different wood raw materials and furnishes on drainability, runnability and paper quality were studied in realistic papermaking conditions with the KISU dynamic short circulation device (VTT Jyväskylä). TMP pulps were produced with a 12" disc refiner and 20% and 30% kraft contents were used in the furnish.

In the KISU tests clear differences in wood properties between fast and slow-growth samples were evident, including annual ring width 2.7 vs. 1.4 mm, density 390 vs. 440 kg/m³, and length-weighted fibre length 2.4 vs.

3.1 mm, respectively. These were, however, not the only factors affecting the wood properties. The wood samples were sourced from trees of different provenance, size and age (slow-growth being thicker and older). Refining of fast-growth wood had significantly higher energy consumption than refining of slow-growth wood. As regards paper properties, fast-growth wood gave higher light scattering than slow-growth wood, as well as better formation (Figure 4 and Figure 5). This is probably due to smaller fibre dimensions and lower bonding ability of TMP fibres and fines. Fast-growth wood also gave better wet tensile and residual tension (Figure 5), indicating low-



Figure 4. Specific energy consumption of TMP refining (with 12" Sprout Waldron) of fast and slow-growth Norway spruce (left). Light scattering coefficient of paper made with the KISU dynamic short circulation device using 80% and 70% fast- or slow-growth spruce TMP in the furnish (right).



Figure 5. Formation of paper made with the KISU dynamic short circulation device using 80% and 70% fast- or slow-growth spruce TMP in the furnish (left). Residual tension after 0,475 s at 1% strain as a function of dry solids content (right).

er web-break tendency in the open draws of an actual papermaking line. These differences in runnability are mainly due to better formation, and thus derive indirectly from differences in fibre properties.

It can be concluded that whereas clearly reduced fibre length and wood density have a clear impact on the papermaking process, small differences in wood and fibre properties due to intensified forest management do not have a significant effect on the TMP process or paper properties. By comparing pure mechanical pulp laboratory sheets, the typical effects of changed particle size distribution (smaller size) and fibre and fines quality [less bonding, more scattering] were revealed: lower strength and better optical properties. However, the studies with realistic furnishes in realistic papermaking process conditions showed notably better formation for furnishes with TMP made of fast-growth spruce, which led, for example, to better wet and dry tensile strength and runnability potential.

5.3 Environmental performance of forest products

A literature review of carbon footprint assessment (peer-reviewed approach developed) and water footprint assessment (review of existing methodologies and local characterization factors) was conducted and reported. These approaches were tested in case studies. Key environmental aspects related to forestry land use (i.e. not limited to biodiversity, water and carbon issues) were identified and quantified in LCA cases.

5.3.1 Carbon

Based on the literature review, a peer-reviewed approach was developed for the inclusion of forest biomass carbon in product LCA. The key conclusions of the literature review were:

- Timing of sinks and emissions must be considered (including carbon in products) in the climate impact indicator.
- A reference situation, i.e. evolution of the forest in the absence of the activity, is required.
- Future-oriented assessment with multiple timeframes is required to support decision making.
- If substitution credits through product displacement are considered, they should be differentiated transparently in the results.

A case study to include forest carbon stock emissions and sequestration into product carbon footprints was carried out using the MOTTI model for the Pirkanmaa Forest Centre [MK5]. Forest stands were assumed to be harvested within the first 5-year period in accordance with 'silviculture as usual' and then left to develop without human disturbance. The carbon impacts of harvesting were compared to the no harvesting scenario. The key conclusions of the case study were the following:

- The carbon stock of harvested forest is lowered for around 100 years compared to zero harvesting.
- The warming impact of the overall carbon stock change is higher for the first 85 years than that of a corresponding amount of (extracted) fossil carbon. The warming impact reduces in line with increased quantity and length of time that product carbon is stored.
- Carbon neutrality of forest carbon stocks is achieved within roughly 90 years, but the system does not reach climate neutrality within the studied 100 year period.
- Uncertainties are high, especially in the long-term, in the no harvesting scenario.
 Risks of unforeseeable events such as storm damage were not considered. These risks should be better included in further analyses together with soil carbon stocks.
In addition to the case study, the carbon stocks and warming impacts of the various forest management scenarios in the Pirkanmaa region were studied using the MOTTI model. According to the results, the forest carbon stock is significantly higher in the no-use baseline than in any other forest management scenario (BAU, INT, EXT, QLTY) over the 100 year period studied. In all scenarios, the reduction in forest carbon stock from the "no use" baseline exceeds, for a period of decades, the carbon stock removed from forests as logging outturns. Consequently, the cumulative warming impact of reduced forest carbon stock compared to the "no use" baseline is higher than that of carbon released from logging, which is used to obtain substitution credits. Substitution credits were not studied in this case study but are very likely to be lower than the warming impact caused by harvesting. This is because one tonne of biomass carbon typically replaces less than one tonne of fossil carbon. Thus, in the short term (decades), the highest climate benefits are achieved by letting the forest sequester carbon.

The climate impacts of the continuous use of land for forest biomass harvesting are evident in the decade-to-century timeframe. However, in the long-term (centuries), the climate benefits of forest biomass utilization may exceed (even significantly) the benefits of shortterm carbon sequestration. It is worth recognizing that no-use of forests, which appears to be the optimal solution for climate change mitigation in the short term (decades), is also unsustainable in the long run if non-renewable resources are used for material and energy production instead of wood. Furthermore, an understanding on the evolution (increase) of carbon stocks in Nordic forests is essential in discussing what is a "reasonable". "equitable" or "sufficient" level of climate change mitigation for each country or sector of society

5.3.2 Water footprint

Water footprint assessment is an ambitious tool for measuring human appropriation of water resources and promoting sustainable use of water. Based on recent case studies and examples from water-abundant Fennoscandia, it was shown, however, that volumetric-based water footprint methods, such as the Water Footprint Network, are not appropriate tools for evaluating the water use sustainability of forestry and forest-based products. It was found that current WF methodologies have severe limitations in the way they understand water as a cycling renewable resource. Because the global water cycle is closed, locally consumed water will be rapidly available for other uses either locally or in more distant locations. Therefore, the results of volumetric water consumption inventories are always dependent on the geographical boundaries selected for the analysis. Additionally, aggregating catchment level water consumption over a product life cycle does not consider fresh water as a renewable resource and is inconsistent with the principles of a closed hydrologic cycle, where, e.g., locally evapotranspired water rapidly becomes available for re-use in the form of precipitation. The conclusion is that volumetric water use inventories are useless and potentially misleading if used to compare the water use efficiency of a product or a company. Exceptions to this "rule" are well-defined and accurately monitored blue water use efficiency or water consumption in direct pulp mill operations. A detailed analysis of the fundamentals of the hydrologic cycle, the pros and cons of the WF concept, and opinions on its future development can be found in Launiainen et al. (2013).

If WF is to be used as a tool for water use sustainability analysis at the product or company level, water consumption must be contextualized (at least) against local conditions. This approach shares common ground with life cycle impact assessment (LCIA) methods and also forms the basis for the current ISO-WF standard proposal. It is important to recognize that the sustainability of fresh water use can be defined in terms of relative fresh water availability (quantity), suggesting that sustainable water use should not exceed locally available, renewable supply. It can also be defined in terms of potential water quality degradation or negative impacts on ecosystem service delivery. This means that the potential impacts on water availability, water quality or water-dependent ecosystem processes have to be considered when evaluating forest sector water use sustainability. In the forest sector as a whole, sustainable water use means applying water and energy efficient processes and technologies, efficient wastewater purification methods, and limiting water consumption to levels supported by local water resources. In forestry, water sustainability means minimizing negative impacts due to changes in quantity and/or quality of surface and ground waters.

The current ISO WF standard 14046 proposal defines WF as the potential impact on local water resources, including impacts on water availability (volume), water quality and water-dependent habitats. Water footprint is always a measure of an impact, and several characterization factors are used to define it in terms of either local water scarcity or local water quality change. WF methodology development is currently ongoing with collection of primary data and characterization factors. Compared to earlier WF methodologies the ISO standard draft is significantly better suited to humid regions, especially Finland where water is abundantly available and efficient water purification methods are applied in industry, accompanied with strict guidelines on minimizing the water quality impacts of forestry. As with previous water footprint methods, the ISO WF standard draft also includes green water use, consisting mainly of evapotranspiration during biomass production, as part of the water use inventory. This inclusion may, however, be in fact positive for the Finnish forest sector - in particular when domestic biomass sources are considered. This is both because it can be indirectly shown that the green water use of managed 'semi-natural' boreal forests does not significantly differ from that of unmanaged forests, and because water is abundantly available in Finland (e.g. Launiainen et al., 2013). Together, these two issues ensure that the "waterstress weighted" ISO 14046 water footprint will be negligible for Finnish pulpwood, which may form a competitive advantage against wood sourced from semi-arid regions where water availability is limited.

As a part of work package, a pilot study was carried out, in which total nutrient and suspended solid emissions in a paper life cycle were estimated. The results indicated that forestry's share of total emissions is small. The results suggest that water protection methods applied in forestry are efficient (and constantly under development) and greater potential for emission reduction may occur elsewhere in the production chain.

To conclude, water footprint indicator(s) are under constant development and no established method is yet in place. The direction of current indicator development, such as ISO standard 14046, is positive and the resulting methods are likely to be better applicable to forestry than the methods available at the start of EffFibre program. The major result of the 'Water' task has been the critical scientific evaluation of existing indicators that formed the basis for steering the development of the ISO 14046 WF method. As more established WF reporting tools such as the ISO standard become available, future projects could deliver primary data, calculation tools and concrete answers for specific reporting requirements.

5.3.3 Land use

Significant differences were found between different solid biofuels originating from different land-use classes (arable, managed forest or mineral excavation area). In all management scenarios, forest biomass occupied a larger land area per unit energy than agro-biomass, while peat occupied the smallest area. The land use impact indicator results differed significantly from mere calculation of hectares of land occupied because the impact indicators differentiate the impacts of these three land types. Although forest bioenergy has higher land occupation needs than agro-bioenergy, the land use pressure and impact indicator results are of a similar magnitude or even lower for forest bioenergy. The case study reveals that no conclusions nor comparisons should be made regarding the environmental impacts of land use based only on the land use inventory results (m²a and m² converted per functional unit). This would treat forest biomass production especially unevenly, as one hectare of managed forest has a lower environmental impact than one hectare of more intensively managed land. Naturally, the overall impacts can be higher if much more land is occupied in the forest biomass system than other competing systems that provide same function. Land use impact assessment is needed. As deforestation is relevant to Scandinavian forestry, impacts from land use change were found to be relevant only to agro-biomass in a sensitivity analysis.

The results suggest that intensive forestry with higher yields and levels of harvesting would place less pressure on productive land availability, but existing LCA methods are still too coarse to identify the impacts of changed forest management, especially with respect to ecosystem service and biodiversity impact indicators. Although previous literature indicates that the actual environmental impacts through land use are known to be significant, especially for agricultural and artificial 'sealed' use of land, it still remains questionable whether these are captured with satisfactory reliability with the applied LCA method and LCIA indicators, especially for managed forest land use.

5.3.4 Biodiversity

Biodiversity indicator set developed based on stakeholder workshop and expert interviews:

- Soil preparation [% share on average of clear-cut areas]: a) soil harrowing of clear-cut sites, b) soil mounding on clear-cut sites.
- Deadwood [amount of 10+ cm diameter deadwood]
- Share of protected/natural forests, or old-growth forest [100, 120 & 140 y] with more than 20 m³/ha deadwood, within the management area Tree species composition: a]
- Share of domestic tree species of all trees or of cut trees, b) share of deciduous trees
- Preservation of valuable habitats in loggings [%]
- Retention trees per regeneration area [m³, at regional level]

These indicators are globally usable when measuring the effects of forest management and forestry operations on forest biodiversity. However, in order to take into account the changes in biodiversity throughout the whole supply chain, different kinds of indicators are needed. Several proposals have been made for general biodiversity indicators for LCA. However, biodiversity is not commonly taken into account in LCA studies.

Promising indicators for LCA assessment of forest products:

 Habitat-based BD indicator. This indicator measures biodiversity aspects related to land use in forestry operations. The intrinsic quality of an area is assessed on the basis of Ecosystem Scarcity and Ecosystem Vulnerability and an index on Conditions for Maintained Biodiversity. The indicator can utilize similar data to traditional forest biodiversity indicators, such as amount of decaying wood (regional estimates).

- BDP (biodiversity damage potential). This indicator quantifies biodiversity impacts in LCIA in different world regions. It is based on empirical data on several species groups. Relative species richness is used to calculate characterization factors for occupation impacts of terrestrial ecosystems expressed as a biodiversity damage potential (BDP).
- Human appropriation of net primary production (HANPP) is an indicator that measures the disturbance of energy storage in ecosystems and relates to ecosystem diversity and stability. HANPP describes the difference in the free NPP left for ecosystems between the current land use and a reference natural state (HANPP = natural state NPP – reduction in NPP – harvested).

In order to obtain reliable results from biodiversity studies in the LCA context, the BD indicators in LCA should be developed further.

5.3.5 Case studies

Climate impact

The climate impact from the loss of additional carbon sequestration was shown to be higher than the equivalent amount of fossil carbon over a period of several decades. In the studied value chain, the absolute climate impact from the use of biomass exceeded the fossil impact for 100 years. The unfavourable results for biomass were due to the short life time of a typical paper product, large share of biofuels used in mill integrates, and slow growth of boreal forests. These results differ markedly from the traditional approach of considering sustainably grown biomass as carbon neutral in LCA. Even though the renewability of biomass is already considered in the modelling of GWPbio-100 factors, the climate indicators do not take into account that the depletion of non-renewable (fossil) resources can be avoided. Although the developed bio-GWP factors are scientifically sound and technically applicable in LCA, the background forest models used in this approach contain considerable uncertainties in estimating the development of carbon stocks, especially for longer time horizons. These uncertainties should be considered when interpreting and reporting the results of this specific product case study.

Water impact

The results showed that the production of pulp and paper is the main source of P, N and suspended solids emissions in the value chain. The contribution of forest management (fibre production) to total emissions is most significant for suspended solids (SS), causing up to 12% of the total load, whereas for P (and the freshwater eutrophication potential impact category] the share is 1-2% and for N (and the marine eutrophication potential impact category] 5-6%, respectively. The alternative forest management scenarios have a small influence on total emissions in the value chain of supercalendered (SC) paper. This is because the share of forest management related emissions is small in the value chain, and also because the loads normalized by the amount of pulpwood produced in different management scenarios are not very different. The largest difference is in SS emissions between EXT and QLTY, the load in the latter scenario being 12% higher, due to more intensive forest management operations such as fertilization and ditch drainage. It must be noted that even though forest management practices increase the nutrient and suspended solids load on watercourses compared to natural background loads, the increase is small expect for SS. The share of forestry impact on paper product life cycle suspended solids load is greatest in forestry scenarios involving intensive forest management (INT) and high quality timber production (QLTY), since more ditch network maintenance operations are carried out in these scenarios. The data sources and calculation methods presented here can be used as part of water footprint assessment of forest products which, according to the preliminary ISO water footprint standard, should also include analysis of water quality impacts and impacts on ecosystem service delivery.

Allocation

Allocation factors were determined for forestry products (pulpwood, sawlogs and energy wood, as well as thinnings and final fellings), sawmill products (sawn timber and chips) and pulp mill products (chemical pulp and energy). The factors vary depending on the allocation method, for example the environmental burden of forestry is attributed for the most part to pulpwood in the volume allocation (favours sawlogs) and to sawlogs in the economic allocation (favours pulpwood). In addition to the chosen allocation method, also the selected route of the fibre to the pulp mill affects the result. For example, the carbon footprint for chipped pulpwood was 70-85% higher for fibre that comes directly from forest to mill compared to fibre coming via sawmills (where the biggest environmental burden from chipping is allocated to sawn timber). For pulp mill products, the results show that the economic allocation of mill products favours energy and the energy allocation favours pulp. A key weakness of the economic allocation is its dependence on fluctuating prices. The carbon footprint of electricity sold by a pulp mill varies significantly depending on the allocation method chosen, whether fibre balance, energy balance or economic value. In conclusion, the allocation method can significantly influence and even determine the result of assessments.

5.4 Dissemination

The results were disseminated in a number of ways. The main partners involved in the dissemination task were Metsähallitus, Metla and Metsä Group.

First, the feasibility of intensive forest management practices were tested and demonstrated in an operational environment of a large forest owner. The demonstrations were coordinated by Metsähallitus and were conducted in a designated demonstration area in Nurmes, Finland.

Demonstrations of silvicultural practices

The quality of mechanized tending of young stands was studied at the Nurmes demonstration area by the Lieksa forestry team. The level of work quality [site impact] was strongly sitedependent unusually wet conditions in 2012 had a negative effect on the measured work quality In dry conditions the measured damage frequency of remaining stems was lower than in earlier studies.

The quality of mechanized forest planting was studied at both mineral soil and peatland sites with a shallow peat layer on top of the soil in the Nurmes demonstration area. A Bracke planter was used for mechanized planting, while manually planted areas were mounded and planted using planting tubes. The results did not markedly differ between the two soil types, and there were no marked differences in the quality of the manually and mechanically planted stands.

In addition, the following sub-demos were carried out:

- Combining forwarding of logging residues and soil preparation
- Mechanized pre-clearing of undergrowth integrated in cutting a tree harvester in first thinnings

- Repeated cleaning during the same growing season
- Repeated cleaning of forest roadsides during the same growing season

Demonstrations of harvesting operations

Integrated harvesting

Several harvesting methods were demonstrated for first commercial thinnings. In addition to traditional pulpwood harvesting, integrated harvesting of pulpwood and energy wood, and harvesting of pulpwood using a multi-tree harvesting head were demonstrated and compared in field conditions. New forwarder grapple types were also field tested.

Novel load space solutions for forwarders

Metsähallitus, Metsä Group and Ponsse jointly tested and demonstrated new load space solutions in forwarders for energy wood and pulpwood extraction. Extendable and compressible load space solutions were tested in practical harvesting operations. A forwarder equipped with a laterally extendible load space was found to be effective in forwarding logging residues. Many improvements were suggested for further development. The compressible load space was found to be useful in forwarding whole trees, but its market potential was considered quite low in Finnish conditions.

Summer harvesting of peatland forests

Harvesting of peatland forests in summer conditions was studied together with Metla. The study was divided into two focus areas: 1) research and demonstration of different harvesting methods in thinning of peatland forest and 2) research on use of airborne laser scanning (ALS) data for planning of peatland forest harvesting. In conventional pulpwood thinning operations in challenging weather conditions, with logging residues laid on the strip road, it was found that after one harvester pass and 2–3 three passes by a loaded forwarder, rut formation was usually under 10 cm. When energy wood was harvested in combination with pulpwood, as much as 28–48% of logging residues were left in the forest. When predicting the depth of ruts after forwarding, the rut depth after harvester and the total mass passed over the measurement point, were good predictors.

ALS data was found to be useful in planning peatland forest harvesting and estimating soil bearing capacity. However, the bearing capacity of peatland stands is highly variable, making accurate local prediction difficult.

Demonstration of laser scanning

To obtain relevant forest data the Nurmes demo area was measured using airborne laser scanning. Several new uses for ALS information were tested and demonstrated. ALS with increased pulse density was also used on small areas for research purposes. The following were demonstrated or studied using the ALS data:

- Need for young stand tending
- Need for clearing undergrowth before first thinning
- Improved analysis of stand characteristics in peatland forests
- Classification of poorly productive forest areas
- Decision support for planning of remedial ditching in peatland forests
- Decision support for planning fertilization of peatland forests

Three-dimensional laser scanning of sample plots was also tested at Nurmes. The equipment performed well in varying weather conditions, and in favourable conditions volume estimates of timber assortments were accurate. In contrast, the results from a small-scale test revealed a number of practical areas for development, such as cost efficiency of measurement and usability in diverse stands, which need to be solved prior to wider adoption of the method.

Model-based calculations for demonstrating the impacts of intensive forest management

Intensive management of Metsähallitus forests

Metsähallitus and the Finnish Forest Research Institute (Metla) conducted three joint projects to investigate the feasibility of intensive management practices in the state-owned forest area administrated by Metsähallitus. Firstly, alternative forest management regimes for young Scots pine stands (Metla) were evaluated in order to develop silvicultural guidelines for young stand management for Metsähallitus. Secondly, a scenario analysis for the Nurmes demonstration area was carried out to assess the economic potential of intensive silviculture at the regional scale. Thirdly, a scenario analysis for Metsähallitus's Eastern Finland forestry area was completed. The aim was to determine the optimal allocation of alternative silvicultural practices within a given budget.

Profitability of silvicultural practices

Metsä Group and Metla implemented a study to demonstrate the impacts of alternative silvicultural practices on profitability at the forest stand level. The focus was on a) assessing regional variation in profitability throughout Finland, b) determining the impact of young stand management practices on profitability, c) assessing the profitability of energy wood thinnings, and d) studying the impact of wood prices on profitability. A simulation study was completed with the MOTTI stand simulator of Metla.

The results revealed that young stand management plays a key role in ensuring profitable forest management. In first commercial thinnings, pulpwood recovery outperforms energy wood recovery with current wood prices and management costs. In later commercial thinnings, thinning from above proved to be more profitable than thinning from below in well-managed coniferous stands. In southern Finland, intensive management ensures good profitability, while in northern Finland good results can be achieved with less intensive management programmes.

Demonstration sites in experimental stands

Metla coordinated the establishment of demonstration sites to illustrate the impacts of different forest management alternatives.

A set of permanent field experiments in the Vesijako research forest area was selected for demonstration purposes. The purpose was to illustrate the impacts of management alternatives in different types of forest stands. Long-term experiments are well-suited for demonstration purposes as the treatment effects can be clearly seen. Experimental stands were equipped with informative signposts and posters. In addition, comprehensive information on the demonstration sites and their management practices were made available online [www.metla.fi].

Decision support tool for assessing forest management alternatives at forest stand-level

Metla's MOTTI program is a stand-level decision support system for assessing the impact of alternative forest management practices on wood and biomass production and profitability. Metla has recently developed and published an updated version of the Motti program.

6. Exploitation plan and impact of results

The results of work package may have a major impact on the Finnish forest and energy sectors. We have shown the potential of Finnish forest as a resource of renewable raw material. We have provided research-based knowledge on the most feasible, cost-efficient and sustainable measures to intensify biomass supply. Moreover, we have demonstrated the implementation of these measures in practice both in large-scale forestry (demonstration forest area of Metsähallitus) and at the individual stand level (demonstration plots of Metla). The results and tools are therefore directly applicable in forestry planning at different levels of decision making by different kinds of forest owners (private forest owners to large forest companies and state forests]. The results are also serve as valuable background information for forest policy making.

The results regarding forest management serve to confirm and summarize the existing scientific knowledge on the impacts of forest management practices on the availability and cost of wood and biomass and thus assist forest owners in their decision making. In addition, the demonstration sites promote the application of best practices. In the long-term, the final impacts on the availability and cost of wood and biomass will depend on the preferences of the forest owners. It is to the benefit of the forest industries to promote management practices that secure the availability and sustainability of wood at a competitive cost.

Sustainability and environmental-friendliness are basic principles of forest resource use. From this viewpoint, novel results on environmental performance of biomass supply and forest products were presented. Environmental indicators were comprehensively assessed and related new measures and methodologies were developed. These results strengthen the scientific basis of environmental indicators and their application in analyses of the environmental performance of forest products. Finally, the project provides improved means for environmental communication in the forest sector.

7. Publications and reports

Peer-reviewed scientific publications :

Helin, T., Holma, A. and Soimakallio, S. 2014. Is land-use impact assessment in LCA applicable for forest biomass value chains? Findings from comparison of use of Scandinavian wood, agro-biomass and peat for energy. Int J Life Cycle Assess (in press), doi: 10.1007/ s11367-014-0706-5.

Helin, T., Sokka, L., Soimakallio, S., Pingoud, K. and Pajula, T. 2012. Approaches for inclusion of forest carbon cycle in life cycle assessment – a review. GCB Bioenergy [2012], doi: 10.1111/gcbb.12016.

Launiainen, S., Futter, M., Ellison, D., Clarke, N., Finér, L., Högblom, L., Laurén, A., and Ring, E. 2013. Is the Water Footprint an appropriate tool for forestry and forest products: the Fennoscandian case. Ambio. doi: 10.1007/s13280-013-0380-z.

Mäkinen, H. and **Hynynen, J.** 2012. Predicting wood and tracheid properties of Scots pine. Forest Ecology and Management, 279, 11-20.

Mäkinen, H. and Hynynen, J. 2012. Wood density and tracheid properties of Scots pine: Responses to the timing of the first commercial thinning and fertilization. Submitted to be published in Canadian Journal of Forest Research.

Posters and presentations:

Ahtikoski, A. 2012. Realizing monetary gains of tree breeding: economic impacts of breeding for growth and quality. Joint workshop of EffFibre and NovelTree: Realized gains and future prospects in tree breeding and seed deployment Workshop, 18 October 2012, Helsinki, Finland, House of Science and Letters, Kirkkokatu 6. Helin, T., Mattila, T., and Antikainen, R. 2012. Applicability of land use impact assessment methods in LCIA - findings from practitioners' perspective. Poster. 6th SETAC World Congress 2012, Berlin, 20.-24.5.2012.

Holma, A. 2012. Biodiversity indicators for forest industry. Presentation in IUFRO 2012, Second International Conference on Biodiversity in Forest Ecosystems and Landscapes, University College Cork, Ireland.

Holma, A. and Antikainen, R. 2011. Determining biodiversity indicators for forest based industry. 2011. Poster. ALTER-Net Summer School 7.-16.9.2011, Peyresq, France. [Poster]

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OPERATIONAL EFFICIENCY OF INTENSIFIED WOOD PRODUCTION AND SUPPLY

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ABSTRACT

The objective of the Operational efficiency of intensified wood production and supply work package was to improve the productivity and cost effectiveness of mechanized timber harvesting and forest regeneration through advanced information technology and automation.

The studies provide test results on the functionality and accuracy of a tree mapping system used in harvesting, and evaluation of its utilization possibilities. The tested prototype system is applicable for automatic measuring of stand parameters during cutting. Furthermore, the effect of different forwarding practices on productivity was assessed, a Logging Map pilot system for visualization of terrain properties for harvester operator was developed, and a CAN-bus based terrain mobility mapping system for harvesters was studied. These methods have remarkable potential for improving productivity and quality of wood harvesting, and their commercial development is therefore a key point of focus.

Field tests of adjustable forwarder load space and load compaction technologies in energy wood hauling showed that both alternatives increased load size and improved productivity by 5–30% compared to ordinary load space constructions depending on the energy wood assortment and compression technique.

A survey and pilot test of novel spatial data sets supporting wood procurement planning and operations were carried out. In particular, a new digital elevation model and soil wetness index were found to be effective in forecasting soil conditions. A preliminary study on the potential of remote sensing to measure timber quantity and quality parameters gave promising results, and provides a good basis for further development of inventory methods targeting more detailed tree parameters.

An automatic planting spot detection system and practices for cost-effective seedling logistics for a continuously operating planting machine resulted in the preliminary description of a machine concept. The results provide the basis for the construction of a prototype machine.

Comparison of various tending practices from the point of view of forest growth and profitability of the wood production chain showed mechanization with selective methods to be reasonable compared to motor-manual cleaning. Test results of the excavator-based Risumoto cleaning machine proved that the concept reduced substantially the costs of mechanized cleaning. Pilot tests of an operator-supporting camera system in the cleaning machine indicated that the camera system helps improve work quality.

The results improve the cost efficiency of the wood supply chain through better harvesting productivity and new information sources enabling more conditions-specific planning and execution of wood procurement. The results also enhance the mechanization of silviculture, thus decreasing forest regeneration costs and manpower. The tested equipment, machine and system concepts enable new business opportunities for forest machine manufacturers, system providers and forest machine contractors.

Key words:

wood supply, wood harvesting, forest regeneration, productivity, cost-effectiveness, operator tutoring, tree mapping, mechanization

1. Background

Timber harvesting has become fully mechanized in Finland in recent 30 years. Today's cutting and hauling machines and methods are the result of a long and complex development process. It is becoming increasingly difficult to achieve significant further improvements in the basic construction and functions of harvesting machines. However, emerging new technologies are offering interesting potential for increasing productivity and work quality and for improving the work conditions of machine operators. A system for automated positioning of harvesting machines with simultaneous tree mapping and measurement, and an operator tutoring system based on artificial intelligence are examples of new possibilities. Basic research on the potential of high-level technology has been carried out during the last few years with very promising results.

The expected lack of labour and decreasing trend in profitability in forestry require improvements in the productivity of silvicultural operations. Tree planting and pre-commercial thinning are the most important and resourceintensive tasks in forest regeneration. Mechanization of this work is taking its first steps. Today, the mechanization rate in tree planting is about 3% and in pre-commercial thinning even less. Totally new technologies are needed in order to achieve the rapid increase in mechanization that the industry requires. Robotics and automation offer two key routes towards the complete mechanization of silviculture.

2. Objectives

The overall objective was to advance technological and logistical solutions for intensified wood production and harvesting in order to improve the productivity and cost effectiveness of mechanized forest regeneration and timber harvesting. The focus was on:

- Clarifying the possibilities of automated tree mapping for more precise and efficient machine work in timber harvesting
- Improving the performance of the harvester-forwarder system with AI-based tutoring systems
- Mapping and testing new spatial information sources for wood procurement planning and operations
- Creating new knowledge and technological concepts for mechanized planting and cleaning

3. Partners and their contributions

The research was carried out jointly by research organizations and Finnish forest cluster companies. Table 1 presents research partners and their roles.
 Table 1. Partner organizations and their research roles.

Work package partners	Role of participating organization
Aalto University School of Science and Technology	Developing an autonomous system for planting spot detection and preliminary concepts for a planting device for a continuously operating planting machine. Testing the use of camera technology to support operator work in mechanized cleaning.
Argone Oy	Production of measurement data from different stand types using an on-board harvester laser scanning system. Calculation of tree-level parameters.
Ponsse Plc	Provision of forest machines with additional installations for the research purposes.
	Ponsse forwarder simulator adjustment for testing and validating the developed monitoring and assisting systems.
	Demo application for machine monitoring and operator assistance created jointly with Creanex Ltd.
	Data collection and evaluation of tree mapping data.
	Provision and participation in field testing of novel forwarder load space constructions for energy wood hauling.
Creanex Ltd. (subcontracted by Ponsse)	Participation in developing monitoring and operator assistance system test versions.
	Provision of machine installations needed for automated machine monitoring, and participation in their field testing.
	Participation in modification of the forest machine simulator for the study purposes.
Finnish Forest Research Institute (Metla) Vantaa and Joensuu unit	Research on the potential of operator tutoring/assisting systems in wood harvesting (feasibility study and questionnaire survey).
	Studying the potential of operator tutoring systems in a simulator environment and actual logging sites; benefit calculations.
	Studying and developing a forwarder routing system in a simulator environment and actual logging sites.
	Research on the use of mobility data recorded from harvester CAN- bus channels for developing prognosis systems for trafficability and soil disturbance mapping.

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Metla, Suonenjoki unit	Evaluation of the productivity and cost-effectiveness of machines developed for planting and pre-commercial thinning.
	Test data collection for the autonomous plant detection system and comparisons of seedling logistic alternatives for mechanized planting.
	Analysis of forest management principles supporting effective mechanization of cleaning.
Metsäteho Oy	Responsible for coordinating WP and for the following research tasks:
	Developing methods for utilizing automatically produced tree mapping data in harvesting work and quality control and for updating growing stock information after cutting.
	Clarifying the usefulness of novel spatial information sources for the prediction of harvesting conditions.
	Evaluation of the productivity and cost-effectiveness of machines developed for planting and pre-commercial thinning.
	Testing new machine concepts for mechanized cleaning.
Metsä Forest	Supporting field testing of the tree mapping system, novel spatial information sources, energy wood compressing technique and cleaning machine.
Metsähallitus	Supporting data collection and field tests for the development of harvester operator tutoring systems.
University of Helsinki	Preliminary study on the current potential of remote sensing for measuring timber quantity and quality parameters.

4. Research approach

The potential of new geographical information – particularly remote-sensing based soil and forest data – to support intensified wood production and harvesting was examined with a focus on wood supply planning and decision making. Information used for wood supply planning must be more precise and versatile than conventional data sets in order to describe spatial variation and anticipated changes in harvesting site and forest road conditions. The research was carried out as a preliminary study of the use potential of different data sets together with pilot field tests. A new precision digital elevation model was found to be a promising data source for use in decision support systems.

A on-board automatic tree mapping system was tested in harvesting operations and the potential of the system was analysed. The

main objective was to determine how effectively the laser scanner can detect trees in various forest conditions as well as the measurement accuracy of tree dimensions. The study was carried out as a field experiment in which sample plots in different types of thinning stands were measured to obtain reference data. A prototype tree mapping system was then installed on a harvester and test data collected in connection with cutting. The results were promising, indicating that the system can automatically detect trees and also produce estimations of the dimensions of remaining trees. The system has potential, e.g., for automatic control of thinning density and recording of stand parameters after cutting.

In the "Intelligent operator tutoring systems for wood harvesting" task the aim was to find and study new technologies and tutoring systems to improve the cost- and environmental competitiveness of timber harvesting. The task was divided into three subtopics: a) Multi-informative map-based assistance in harvesting, b) Using harvester CAN-bus data for mobility mapping and c) Principles and potential of forwarder load cycle guidance. As the main findings of task were as follows: 1. Forest machine operators are open to receiving online assistance in their work. Specifically, assistance should be aimed at providing more accurate information on terrain trafficability and offering more precise information and assistance during timber forwarding. 2. Multi-informative map-based assistance in harvesting approved helpful in planning strip-roads during harvesting. 3. Harvester CAN-bus data, including accurate ground speed information, can be used in creating mobility maps for forwarding. 4. The new forwarding techniques and advanced forwarder routing assistance studied in the task offer high potential for improving forwarding performance in terms of productivity, soil compaction, machine straining and fuel economy.

The cost-competitiveness of machines and devices used in mechanized planting and young stand tending and their manpower requirements were compared to motor-manual forest work. Cost calculations for the machines were made using Metsäteho's cost calculation models. Productivity data was collected from earlier studies in spite of new concepts, which were tested in-field with the methods of time and follow-up studies. One of the main conclusions was that mechanized planting and stand tending can essentially reduce the manpower requirements of silviculture. However, the cost-competitiveness of mechanized methods is still generally poor, although some costeffective practices - such as the uprooting method in cleaning - were identified.

The continuously operating planter study was based on the challenge of developing a machine vision system to enable the planting machine to find suitable planting spots. The system was specifically required to automatically find suitable planting spots for spruce seedlings after the mounder had prepared the soil. The principles of planting machine operation and planting logistics were examined as the basis for machine development. The project succeeding in building a machine vision system capable of detecting and classifying suitable planting spots. The project results provide the basis for the future design and commercial construction of a continuously operating planter.

5. Results

5.1 Mechanization of silvicultural operations

The aims of the mechanized planting subtask were to evaluate the costs and labour intensity of mechanized planting compared to manual work, to find new technical solutions and applications for the automation of mechanized planting work phases and to evaluate seedling packaging concepts for mechanical planting.

The comparative study covered the most commonly used planting devices, the Bracke and the M-Planter. The productivity per hour $[E_{15}]$ was 150 seedlings for the Bracke and 151 seedlings (E,,) for the M-Planter. Mechanized planting was compared with excavator spot mounding and manual planting. In the calculation assumptions, the planting density was 1,800 spruce seedlings per hectare, and the average planting area 2.5 hectares. Costs calculations were made using Metsäteho cost calculation models and were based on the prices of a new medium-sized excavator and a new planting device. The wages for operators and forest workers based on collective agreements. The working period for machines was 5 months for planting operations and 6 months for site preparation. The total working period included 3 weeks of holidays and stoppages. Operations were carried out in two shifts throughout the working period. Outside of this period the base machines were engaged in other work.

The costs of mechanized planting were 23% higher than the manual planting chain. The results suggest that in order compete with spot mounding and manual planting, mechanized planting must increase its current productivity $[E_{15}]$ by 25%. This is equivalent to a productivity of at least 190 seedlings per hour. Mechanized planting requires on average 20% less labour than mounding followed by manual planting.

For implementing a continuously operating planter, machine vision algorithms for detecting possible planting spots were studied as a technical solution. The implemented machine vision system uses stereo camera images to model the ground level. Planting spots are searched from mounds created by a continuously operating Bracke mounder. The machine vision system is able to recognize a mound from the modelled surface and to detect a suitable planting spot on the mound (Figure 1). The system was taught using reference planting spot locations provided by man-made reference measurements. Mounds and reference measurements from a different test plot were used to estimate the precision of the detector, which was 94.8%.

A concept-level continuously operating planting machine was designed based on a prototype mounder by Pentin Paja. In the mounder prototype, the rear wheels of the rear bogie of a forest tractor are replaced by mounder wheels. In the concept planter, a machine vision camera would be installed behind the mounder wheel to allow the fresh mound to be imaged (Figure 2). Behind the camera, a sliding boom would be installed to stabilize the planting tool during planting. The boom would be controlled to minimize forces on the planting tool. In addition, positional changes of the planting tool would be measured and actively compensated to avoid breaking the mound when planting and moving simultaneously.

Re-usable plastic cultivation trays were the most cost-efficient option for seedling logistics in the current operational environment when costs of seedling packaging and transportation were taken into account. Cardboard boxes were the most expensive concept due to due to being costly and non-reusable. Cultivation trays are good basic option for use with an automatic feeding system. When improving the competi-



Figure 1. Two successive pairs of stereo pictures presenting a mounding result (left) and examples of best planting spots detected by the automatic stereo-vision based system (right).

tiveness of mechanical planting, development of a reliable automatic feeding system takes precedence over seedling logistics – an automatic feeding system would bring about 5% cost-savings compared to currently used excavator mounted planting devices employing intermittent working methods. In addition, planting productivity would be 4–5 times higher and planting costs halved by the planned continuously advancing planting machine concept, as enabled by an automatic feeding system.

Mechanization of young stand tending

In the mechanization of young stand tending research area, the cost-competitiveness of current machines and devices and their manpower requirements compared to motor-manual forest work were calculated as the starting point for the research. more cost-competitive method when weighed against the combined cost of cleaning and precommercial thinning using a clearing saw. It was assumed that uprooting eliminates the need for subsequent pre-commercial thinning. In young stand tending as a whole, the manpower requirement of the Naarva uprooter was as much as 57–66% lower than manual tending.

The cost-competitiveness of mechanized methods in pre-commercial thinning is, in general terms, poor. However, the manpower requirements are lower than motor-manual work. The manpower requirement of the MenSe clearing head and the UW40 brushwood cutter were 16–37% and 1–26% lower than manual work, respectively.

The cost of the base machine has a major impact on the profitability of mechanized precommercial thinning. New machine concepts are therefore needed to improve the cost-



Cleaning with the Naarva uprooter was the

Figure 2. Working principle and instrumentation of the continuously operating planting machine.

competitiveness of mechanization. Our study of the excavator-based Risumoto machine [Figure 3] proved that the concept can reduce the cost of mechanized tending on average by 23% compared to harvester-based machines [Figure 4]. The costs were, however, higher than motor-manual work with a clearing saw.

Pilot testing of an operator-assisting camera system in the cleaning machine (Figure 5) indicated that the system assists the operator in improving work quality. A comparison of various cleaning practices with respect to forest growth and wood production chain profitability was carried out. The results showed mechanization with selective methods to be viable compared to motormanual tending (Figure 6).

In addition, a prototype cleaning head developed by Tekno-Tuote S. Mononen was tested. The working principle was trialled in two plantation types. According to the tests, the equipment proved non-competitive compared to existing cleaning heads.



Figure 3. Risumoto machine equipped with Marttiini extension crane and Mense Rp40-clearing head.



Figure 4. Cost of pre-commercial thinning with the Risumoto machine compared to motor-manual clearing saw work and harvester-based machine. Assumptions: harvester 76 \in /E₁₅-hour, Risumoto 58 \in /E₁₅-hour, forest worker 28 \in /hour.



Figure 5. The operator-assisting camera system, equipped with a wide field-of-view Full-HD camera system installed on an excavator instrumented for forest management operations with a Mense clearing tool.



Figure 6. Net present value of felling net cash flow with different pre-commercial thinning methods in spruce stands.

5.2 Advanced technologies for improved timber harvesting productivity

Potential of new geographical information sources

The potential of geographical information, especially remote-sensing based soil and forest data, was examined. The focus was on their

use in supporting the planning and decision making of wood supply operations.

The aim in wood procurement is a cost-efficient supply chain in which harvesting and transporting can be done flexibly and responsively to varying supply needs and spatially and temporally changing operating conditions. Information used for wood supply planning needs to be more precise and versatile than conventional data sets in order to describe spatial variation and anticipated changes in harvesting site and forest road conditions. The data should also be sufficiently up-to-date and, preferably, real time.

A key objective of the task was to study existing data sets and to develop methods and applications for providing new information on forest soil and terrain properties. This information is considered to be especially valuable in the appraisal of harvesting site trafficability and bearing capacity of forest soil, landing areas and roads. Changes in conditions due to weather should be able to be anticipated based on spatial and weather data and modelling. This information is valuable for the selection of harvesting sites and the timing of harvesting. The data is useful also for contractors and machine operators for planning site operations as well as for use in operator-assisting systems.

New public and free geographic information data sets and advanced remote sensing based methods for estimating stock and tree attributes have opened interesting opportunities for their use in information systems and wood procurement planning and guidance applications. In this study the usability of three data set types were examined:

- 2 x 2 metre resolution digital elevation model (DEM) based on airborne laser scanning data (National Land Survey of Finland)
- Surficial deposit map, 1:20 000 (GTK, Geological Survey of Finland)
- Airborne radiometric geophysical data (GTK)

Opportunities for developing tailored weather forecasting services for the needs of wood procurement were also preliminarily assessed. The data set with the most potential in terms of offering new terrain data for wood procurement is the new 2 x 2 m elevation model (Figure 7). It is already available from a large part of Finland and is readily applicable in most geographical information systems with minimal configuration. The visualized elevation model can be used in many ways to show variations in altitude, slope inclinations and other features of the terrain that should be considered in harvesting and machinery work.

Topography, and its variation, controls water flows and soil hydrological processes. The topographic wetness index (TWI) calculated from the digital elevation model is commonly used to describe the hydrological status of the terrain and to assess the moisture of the soil. The TWI value can be used to pinpoint areas where water is likely to accumulate, which can be critical to logging and forwarding in terms



Figure 7. The elevation model combined with a traditional base map accurately describes terrain conditions and elevation variation.

of soil bearing capacity. Several TWI calculation methods were studied. The most promising method was tested by comparing its values with field measurement data. There were clear correlations between TWI values and certain topographic and soil properties as well as with observed soil damage due to harvesting. However, based on these results alone it is not possible to provide a definitive TWI value for soil damage risk.

Reflected gamma radiation maps of a test area were produced from airborne radiometric geophysical data without the use of field control data (Figure 8). The ground resolution of the data seems to be too coarse for stand or site level planning. However, the data might be useful for distinguishing between thin and thick peat layers and in delineating peatland work sites according to soil bearing capacity.

The potential of surficial deposit maps is also quite restricted in wood procurement planning mainly due to its low resolution and the fact that mapping has been essentially focused on describing the dominating soil types at 1 metre from the soil surface. However, this soil type information can be useful at the area level in estimating the proportions of critical soil types in terms of bearing capacity.

Automatic tree mapping system for harvesters

The quality of automatic tree mapping data produced during harvesting in real-time was evaluated for accuracy, usability and scope. The tree mapping data evaluated was produced by the Optical Tree Measurement System (OTMS). OTMS is the product of a separate parallel development project conducted jointly by Argone Oy and Ponsse Oyj. The measurement system uses a low-cost bulk 2D laser with an inexpensive measuring platform to produce tree, stem and site map information in thinning stands.



Figure 8. Example of gamma radiation map illustrating potential wet areas of terrain (blue color).

The most significant initial uses of the system include forest stand measurement and operator guidance to achieve optimal thinning density. Other possibilities of the OTMS system include, for example: tree selection assistance during cutting, measurement of stem properties before cutting (bucking support), improved strip road and log pile location precision for efficient hauling, and, in the future, location information for semi-automated harvesting.

Three sets of research data (Kolho, Viitaniemi, Kurkisalo) were collected between June 2011 and June 2012 (53 sample plots) in different test stands and varying weather conditions. The system's accuracy, usability and scope of application were evaluated on the basis of the collected data. The results for the automatic tree mapping system were compared with manual measurements from sample plots (r=10 m). The test stands are located in the Mänttä-Vilppula area and the tests were conducted by Metsäteho Oy and assisted by Metsä Group. Measurement data is collected during cutting when the harvester is stationary, though the crane can move and fell during data collection. The OTMS system extracts individual trees from the collected forest point data and forms local tree maps from consecutive measurements made during harvesting (Figure 9 and 10). These local tree maps and tree locations can be transformed to a global coordinate system with the collected GPS position data. The OTMS calculates the diameter of individual trees from multiple locations and combines the values to obtain a more accurate diameter estimate (Figure 11), it can also find the start and end height of dead branches (canopy is too high for the current system), crooked-growth and bends can also be estimated up to the branch height. The OTMS calculates the thinning progress based on the number of trees left standing to help the operator to achieve optimal thinning density. The simplest information shown is the remaining standing trees versus the target count. It is also possible to indicate with mapping and colour information where further thinning is needed. The right-hand image in Figure 10 shows a completed test plot, with each remaining tree surrounded by a green border.



Figure 9. Left: forest measurement point cloud from one machine position. Right: extracted and calculated trees from the same position.



Figure 10. Left: Two consecutive measurement sub-maps combined. Middle: Larger maps can be formed when enough sub-maps have been measured. Right: OTMS example tree map showing tree and stump locations, strip road, measurement positions, thinning information, etc.

The tree maps formulated with the tree mapping system (OTMS), the laser scanner diameter measurements, and the taper curve measurements were compared with reference measurements and locations gathered during field measurements. Trees left standing after harvesting were located with the OTMS system with 99% accuracy (Table 2 and 3). In mature pine stands trees were located with most accuracy (100%), while in young spruce and birch-dominated forests locating the trees was more challeng-





Figure 11. Left: Combined multiple diameter measurements at different height of the tree (taper curve), including the evaluated start height of dead branches and fitted taper function for height estimate of a measured tree. Right: Example diameter calculation at a given measurement height.

Table 2. Felled and leftover tree identification with the OTMS system per tree species.

	Stem number, after felling		Cutted tr	ees	Stem number, b	Sample plots	
Tree species	N, kpl (reference)	N, % (laser)	N, kpl (reference)	N, % (laser)	N, kpl (referenssi)	N, % (laser)	N
Pine	485	99	448	88	933	94	26
Spruce	354	97	364	66	693	82	15/16
Birch	33	97	48	92	81	94	2
Mixed forest	214	96	298	74	512	83	9
	1086	98	1133	78	2219	88	52/53

 Table 3. Felled and leftover tree identification with the OTMS system in young [I thinning] and mature [II thinning] forests.

and the strength of	Stem number, a	fter felling	Cutted tr	rees	Stem number, be	Sample plots	
Development class	N, kpl (reference)	N, % (laser)	N, kpl (reference)	N, % (laser)	N, kpl (referenssi)	N, % (laser)	N
KL 3 (I thinning)	759	97	906	79	1665	87	33/34
KL 4 (I) thinning)	327	100	227	75	554	90	19
	1086	98	1133	1	2219	88	52/53

ing (97% accuracy). Before felling, around 88% of trees could be identified, with the basal area of the examined plots ranging between 15–49 m². Tree identification was most successful in mature pine and birch-dominated forests, whereas in spruce and young stands identification was more challenging due to stand and branch density. In addition, location accuracy is much higher on the side of the harvester to which the measurement device is fitted. Tree identification accuracy for the device-side half of the test plot was 100% for trees after felling,

whilst being about 91% on the opposite side of the harvester.

The accuracy of diameter measurements depends on the location of the trees and the measuring distance. The overall measurement accuracy of the device decreases as a function of the distance of measurement. The accuracy of the older device sufficient at a distance of up to 5–7.5 metres; at longer distances mean diameter values are reliable, but the standard deviation increases when meas-

Accuracy (mm)	N	Bias	Std. Dev.	N, %	Cumulative, %
-14		10			
10.00					
		1.			
		12.2		1.54	
-3530	2	-32,5	1,7	3,4	
-3025	2	-27,1	2,6	3,4	
-2520	5	-22,4	1,0	8,6	
-2015	1	-15,2		1,7	
-105	2	-7,1	0,1	3,4	
-5-0	2	-1,7	1,1	3,4	8,6
0-5	3	0,9	0,8	5,2	
5-10	3	6,7	1,6	5,2	17,2
20-25	1	20,3		1,7	29,3
25-30	1	26,0		1,7	34,5
30-35	3	32,7	1,4	5,2	43,1
35-40	2	37,3	1,5	3,4	53,4
40-05		HOUR		1.5	56,9
45-50	11	46,8		16.1	60,3
×58	6	1.67.5	6.69	10,5	100,0
	58	-17,1	65,3	100	

Table 4. Accuracy of diameter $[d_{1,3}]$ measurements in pine stands (automatic tree selection by OTMS) and cumulative proportion of observations in different accuracy classes (absolute values).

Accuracy (mm)	N	Bias	Std. Dev.	N, %	Cumulative, %
1.54				791	
-62-62		10.1		1.1	
-10-40					
- apa - 424		10.47	100.00	1.4	
-3530	2	-32,5	1,7	3,4	
-3025	2	-27,1	2,6	3,4	
-2520	5	-22,4	1,0	8,6	
-2015	1	-15,2		1,7	
-105	2	-7,1	0,1	3,4	
-5-0	2	-1,7	1,1	3,4	8,6
0-5	3	0,9	0,8	5,2	
5-10	3	6,7	1,6	5,2	17,2
20-25	1	20,3		1,7	29,3
25-30	1	26,0		1,7	34,5
30-35	3	32,7	1,4	5,2	43,1
35-40	2	37,3	1,5	3,4	53,4
00 45	A.	0.8		3,8	56,9
95-50	- 1	6.9		0.8	60,3
0.20		107.5	165,0	10.1	100,0
	58	-17,1	65,3	100	

Table 5. Accuracy of diameter $[d_{1,3}]$ measurements in spruce stands (automatic tree selection by OTMS) and cumulative proportion of observations in different accuracy classes (absolute values).

uring individual trees. The new device is capable of accurate tree diameter measurement at a maximum distance of 11 metres. The measurement accuracy depends on the number of laser measurements made, the type of undergrowth and the branch number and height. For this reason, the software monitors the measurement accuracy continually and automatically selects the best trees. At best, the diameter measurement accuracy of selected trees is equivalent to the measuring accuracy of a modern harvester. The standard deviation in diameter measurement of other (non-selected) trees is however bigger, and for this reason it is recommended to use the mean values and sampling method to estimate the mean diameter of the sample plot.

In pine stands (automatic tree selection by OTMS), the diameter (d1.3) bias of measurements was 0.9 mm and standard deviation 22.4 mm (Table 4). When the observations $(d_{1.3laser-d1.3reference}, [N=108])$ were classified into two groups based on the difference (Group 1 = $d_{1.3laser-d1.3reference} > \pm 30$ mm or Group 2 = $d_{1.3laser-d1.3reference}$

 $_{dl.3reference}$ < ±30 mm], 86.1% of observations fell into to Group 2 and the bias was 0.3 mm and standard deviation 13.0 mm.

In spruce stands (automatic tree selection by OTMS), the diameter (d1.3) bias of measurements was -17.1 mm and standard deviation 65.3 mm (Table 5). When the observations $[d_{1.3laser}-d_{1.3reference}, [N=58]]$ were classified into two groups based on the difference (Group 1 = $d_{1.3laser}-d_{1.3reference} > \pm 30$ mm or Group 2 = $d_{1.3laser}-d_{1.3reference} < \pm 30$ mm), 34.5% of observations fell into to Group 2 and the bias was -6.5 mm and standard deviation 15.8 mm.

Sample plots were located before and after felling using a Trimble Pathfinder ProXH receiver. The real location (x,y -coordinates) of the sample plot was calculated by using differential correction from three nearest virtual reference stations. The real locations were compared to uncorrected locations and differential corrected locations (VRS data from one base station, Orivesi). The mean difference (x,y- coordinates) were calculated. The influence of satellite geometry on the accuracy of location in different types of forest was studied (Table 6 and 7).

During felling, harvester strip roads were located with three different GPS receivers: Trimble Pathfinder ProXH, the harvester GPS, and the harvester's mapping system GPS. The GPS receivers were located on the roof of the harvester. Differential correction was done using data from Virtual Reference Stations. The locations were compared at the strip road and sample plot level. Comparing the accuracy of different GPS receivers, the mean difference in Viitaniemi was 3.91 m (std. dev = 1.35 m) and in Kurkisalo 2.07 m (std. dev = 1.04 m), when considering the differences between the Trimble GeoXH GPS receiver and the harvester's GPS receiver.

Current potential of remote sensing for measuring stand parameters

The pilot study was divided into four sections: a literature review, suggestions for an alter-

native forest inventory chain, a practical test, and discussion and conclusions. The following methods were included in the literature review: [1] Area-based approach using laser-scanning data and aerial photographs, conducted with existing and public remote sensing materials [e.g. airborne laser scanning, ALS, and aerial photograph materials from the National Land Survey of Finland or the Forest Centre]; [2] Laser-scanning-based individual tree detection, which can be done if the pulse density is higher than in the materials mentioned in [1]; and [3] Tree-level terrestrial laser scanning, which requires the scanner to be brought to the surveyed area (terrestrial laser scanning, TLS/ mobile laser scanning, MLS].

A vision for an alternative forest inventory chain in the future was produced based on the extensive literature review and on the idea that terrestrial laser scanning could be used to reinforce and/or improve the accuracy of forest attribute estimations from aerial applications (Figure 12). It is possible to utilize TLS/

Table 6. Accuracy of sample plot location using a Trimble Pathfinder ProXH receiver (n=33) before and after felling when comparing the differential corrected location (VRS data from three base stations) to the uncorrected location and to the differential corrected location from one base station (BT1).

Advantation of the barry	Sample	Mean difference, m				
weasurement time	plots	diff. corrected (1BT)	uncorreted			
Before felling	33	0,18	1,90			
Pine stands	25	0,13	1,70			
Spruce stands and mixed forest	8	0,34	2,53			
After felling	33	0,10	1,46			
Pine stands	25	0,10	1,48			
Spruce stands and mixed forest	8	0,09	1,40			

Table 7. Accuracy of sample plot location in metres using a Trimble Pathfinder ProXH receiver (n=33) before and after felling utilizing uncorrected and differential corrected data (VRS data from one and three base stations).

Advances of the state	Sample	Trimble PROXH, diff. corrected (BT3)		Trimble PROXH, diff. correted (BT1)			Trimble PROXH, uncorrected			
measurement time	plots	max. PDOP	DOP horz precision	st.dev.	max. PDOP	horz. precision	st.dev.	max. PDOP	horz. precision	st.dev.
Before felling	33	4,75	0,17	0,84	4,87	0,17	0,89	1,82	4,19	1,45
Pine stands	25	4,60	0,15	0,72	4.70	0,16	0.80	1,76	4,16	1,29
Spruce stands and mixed forest	8	5,21	0,23	1,21	5,41	0,20	1,17	2,00	4,28	1,95
After felling	33	3,37	0,12	0,32	3,46	0,12	0,35	1,62	3,23	1,02
Pine stands	25	2,78	0,12	0,30	2,94	0,12	0.34	1,48	3,15	0,94
Sonica stands and mixed forest	.8	5.74	0.11	0.20	5.10	0.11	0.41	2.03	95.5	1 26

MLS technologies also with area-based approaches by measuring the required field reference and to acquire more detailed information (e.g., stem distributions and quality features) from the forest management stands.

The vision of an alternative forest inventory chain was the basis of the practical test. The objective was to research the usability of different laser scanning methods, and the various methods in combination, to measure accurate tree-level stem distributions, timber quantities, and quality features. The results showed that TLS and a combination of TLS and high-density ALS can be used to measure timber quantities accurately, especially log volume (Figure 13, Table 8). Also, the accuracy of quality feature estimation was promising.



Figure 12. A vision for an alternative forest-inventory chain.



Figure 13. Number of stems classified by diameter at breast height estimated with the piloted methods.

 Table 8. Accuracy of timber assortment estimation at tree level after timber quality "outliers" were excluded (11 trees excluded). Reference: harvester measurement.

		Sawwood volume, dm ³					Pulpwood volume, dm ³		
	Bias	Bias%	RMSE	RMSE%	Bias	Bias%	RMSE	RMSE%	
Field	-8.9	-1.2	103.6	14.4	33.4	37.0	51.9	57.6	
TLS	21.0	2.9	125,8	17.5	38.3	42.4	54.2	60.1	
TALS	9.2	1.3	120,8	16.8	37.3	41.4	53.5	59.3	
ALShigh	110.8	15.4	250.1	34.7	32.9	36.5	51.5	57.1	
ALS	279.3	38.8	354.8	49.2	25.7	28.6	49.4	54.8	

Important future research areas include, e.g., using TLS and MLS to measure plot- or standlevel information; developing practical methodologies and detailed examination of cost efficiency; combining TLS/MLS/ALS and harvester measurements; improving tree location (GPS-IMU) accuracy; measuring the real quality of logs using laser scanning (sawn timber/x-ray measurements at the mill); determining optimal ALS pulse density; and use of new sensors (full-waveform and hyperspectral LiDAR) in stem-distribution estimation and tree-species detection. In addition, two main questions will be how to detect changes in the forest (forest management procedures, damages) and how to keep forest resource information up to date.

Load space constructions in energy wood forwarding

The objective of the study was to increase the productivity of forest energy extraction by increasing the load capacity of forwarders. Two methods were investigated. The first method involved widening the load space hydraulically (Figure 14). The second solution was based on load compression with hydraulically tiltable stakes (Figure 15). The productivity increase potential of variable and compressible



Figure 14. Extendible-width load space for stumpwood forwarding.

load spaces in forwarding was studied. The active partners in the project were Metsä Group, Metsähallitus and Ponsse Oyj.

The field studies were conducted in summer and autumn 2011 on harvesting sites in the Jyväskylä region. The study consisted of forwarding several different biomass materials with standard, adjustable and compressing load spaces. The materials forwarded were the following: logging waste / slash, tree stump chunks, cut-to-length industrial roundwood, bioenergy whole trees and delimbed energy wood. The study material comprised a total of 139 loads. The analyses were based on reported load scale tonnes.

Both load space alternatives increased the



Figure 15. Left: Compressible load space for forwarding slash. Right: Compressible load space equipped with straight stakes suitable for roundwood forwarding as well as biomass compression.

load size by 20-30% compared to a standard forwarder depending on the material assortment. With the adjustable load space, the increase in extraction productivity per effective hour was for logging residues 13% and for stumpwood 30%. With the compressing load space, productivity per effective hour of whole-tree forwarding increased 17%. For logging residues, the increase was 5% and for stumpwood 12%. Compression proved ineffective for stumpwood. For logging residues, the benefits of the compression method were attributable mainly to increased load space, rather than material compression. Load compression is therefore considered beneficial only for whole-tree extraction.

5.3 Intelligent tutoring systems for improved harvester-forwarder performance

The unit costs of timber harvesting have increased moderately during the last decade in Finland. Operator tutoring systems offer a means of increasing productivity, thereby offsetting higher unit costs and increased fuel prices. In the "Intelligent operator tutoring systems for wood harvesting" project the aim was to identify and examine new technologies and tutoring systems for improving the cost- and environmental competitiveness of timber harvesting. The project was divided into three subtopics: a) Multi-informative map-based assistance in harvesting, b) Using harvester CANbus data for mobility mapping, and c) Principles and potential of forwarder load cycle guidance.

Multi-informative map-based assistance in harvesting

Vast amounts of digital terrain and surface model data (DTM&DSM) are available for utilization in forest management and forest operations. Compared to base maps, the positioning accuracy and precision of the DTM&DSM data offer better information on terrain trafficability for harvesting. In Task 1, the main objective was to test and observe various map-based visualizations of DTM data for assisting the harvester operator in assessing terrain trafficability and in strip road planning. The DTM map visualization concept Logging Map was developed and tested in practical demonstrations at three harvesting sites (Figure 16). Feedback from two demo operators was collected.

Map-based visualizations of terrain inclination and elevation with 2-metre contour intervals and shaded 3D mapping as well as suggested timber removal zones were presented



Figure 16. Enhanced terrain data of the Logging Map concept (left) alongside a traditional base map view (Photo: Metla/Jari Ala-Ilomäki).

to the harvester operator during cutting work. Live comparison with the currently used base map visualization was accomplished to obtain immediate operator feedback. Generally, the operators felt that base maps do not help in planning strip roads, especially in challenging environments such as steep terrain and poor visibility. Unlike base maps, the accurate DTM visualization with elevation lines and inclination information supported planning of the strip road network. According to the operators, new map information on terrain trafficability would improve the productivity of forwarding as well as harvesting.

Using harvester CAN-bus data for mobility mapping

Presently, the trafficability of a harvesting site is assessed by estimation by the foreman or machine operator. This manual estimation can be challenging, and available assistive site measurement methods are currently too expensive. A comprehensive, continuous and low-cost means of site measurement to enable trafficability assessment is therefore needed. Using the harvester (preceding the heavier forwarder on the site) to create a mobility map of the extraction road network fulfils these requirements.

Harvester motion resistance can be measured using data from the harvester CAN-bus. At steady speed on level ground, engine power via transmission is expended on overcoming motion resistance, mainly wheel sinkage, which in turn is dependent on soil strength vs. loading. To measure expended power from the CAN-bus, data on the pressure and flow rate of the transmission hydraulics, ground speed and terrain inclination are needed. In addition, data on maximum available tractive force is necessary to calculate net tractive force.

The concept was tested on level, 180 m long test tracks, ranging from mineral soil to peat-

land with a peat depth of up to 3.35 m. Soil shear modulus was measured with a 200 mm diameter spiked shear vane (Ala-Ilomäki 2013) and, for maximum tractive force modelling, shear strength was measured with a 98 mm diameter spiked shear vane (Ala-Ilomäki 2013). An 8-wheeled Ponsse Fox harvester was instrumented for the measurements. To validate the map, the harvester was followed by an 8-wheeled Ponsse Wisent forwarder. Both machines were equipped with high flotation steel tracks front and rear.

According to the study results, harvester rut depth explained 74% of the variation in forwarder rut depth. Soil shear modulus explained 42% of harvester rut depth variation. After start-up instability, harvester motion resistance and wheel slip increased as soil shear modulus decreased (Figure 17). Their variation also increased with increasing soil softness. Net traction decreased sharply with decreasing soil strength, making it a viable variable for trafficability mapping. A net traction coefficient of 0.2 is typically considered the lower limit for trafficability. As Figure 16 shows, the trafficability of the test track would have been insufficient after 50 m - this is borne out by the test drive result, with the forwarder operator deciding to end the drive at 40 m.

Future efforts should be aimed at cost-effective measurement of ground speed, determination of maximum available tractive force from CAN data, and data transfer from harvester to cloud server and back to forwarder.

Principles and potential of forwarder load cycle guidance

Large differences – up to 300% – have been observed in forwarding productivity between operators (Ylimäki et al. 2012). Differences in productivity increase in challenging conditions such as steep terrain and soft soil. Moreover, the in-



Figure 17. Soil strength versus key harvester mobility variables on the test track. For mobility mapping, the lower limit of net traction coefficient (0.2) is marked with a red dashed line.

creasing diversification of timber assortments is making forwarding more challenging even for experienced operators. The objectives of Task 3 were to determine the potential effect of load cycle guidance on forwarding productivity and to identify efficient forwarding techniques.

Detailed positional and multi-source work study data on forwarding was gathered from three thinning sites. An experienced and highly skilled operator forwarded a total of 35 timber loads. The forwarding output as well as individual load cycles were compared to the results obtained by the vehicle routing problem (VRP) solution generated with the ArcGis application. The forwarder operator followed his usual forwarding technique with round trips on strip road loops and multi-assortment loads. It should be noted that the VRP optimization system simulated forwarding based on minimum travel on site, and therefore some loads would have been difficult to accomplish in reality.

A new forwarding technique closely following the VRP solution by minimizing driving, especially during loading, was introduced. Based on the information from the VRP optimization result for each load cycle trip, the compiled operator work time element formulas, and the restrictions imposed by the new minimized driving technique, results for the new forwarding technique were calculated. Compared to the operator's standard driving technique, the benefits included increased productivity, decreased soil compaction, lower fuel consumption and diminished strain to forwarder (Table 9).

Table 9. Key-figure comparison between standard forwarding and efficient forwarding using the minimal driving technique in accordance with VRP optimization (151 m³ timber removal).

Standard forwarding	Efficient forwarding	Difference %
3.22	3.08	-4.4
3.5	3+3, 3+4	-
9000	7502	-16.6
213.1	184.7	-13.3
0.613	0.568	-7.3
	Standard forwarding 3.22 3.5 9000 213.1 0.613	Standard forwarding Efficient forwarding 3.22 3.08 3.5 3+3, 3+4 9000 7502 213.1 184.7 0.613 0.568

The benefit and potential of the efficient forwarding technique can be estimated to be much higher than in this comparison, in which a highly skilled operator was used. With operators of average productivity levels, the productivity increase with the new forwarding technique and forwarder assisting system could be 10-20%. On a national level in Finland, the annual cost-saving potential could be more than 10 million euros of a total annual forwarding cost of approximately 210 million euros. To fully accomplish the benefits of efficient forwarding, forwarding should begin after the harvester has completed the majority of felling on the site.

6. Exploitation and impact of results

The aim of the work package was to develop technological and logistical solutions for intensified wood production and harvesting. The results improve the cost-efficiency of the wood supply chain through better harvesting productivity and new information sources for more efficient site conditions based planning and execution of wood procurement [Table 10]. The improvement of harvesting productivity through developed methods and piloted technology is assessed to be 5 – 10 %. The results also enhance the mechanization of silviculture, thus

Table 10. Estimation of the potential reduction of wood harvesting and forest regeneration costs, when assumed that the results have been exploited in full extend.

Research area	Impact of results, MEUR per year	Next steps in exploitation
 More effective wood harvesting Operator tutoring systems Tree mapping system Novel geographical information Load space constructions [energy wood] 	40 - 70	 Productisation of tutoring systems into harvesting machines Extensive piloting of tree map system in practice Implementation of new data sources in current planning systems (GIS) Productisation of adjustable load space construction
 Mechanization of planting and young stand tending Basics for continuously operating planting machine Concepts for mechanised tending 	15 - 20	 Extensive implementation of uprooting method in cleaning Developing and implementation of new cost-effective base machine concepts for pre-commercial thinning Prototype construction of continuously operating planting machine
Total	55 - 90	

decreasing forest regeneration costs and manpower requirements. The tested equipment and machine and system concepts enable new business opportunities for forest machine manufacturers, system providers and forest machine contractors. Several projects targeting into implementation of the achieved results have started or are in planning phase. Further research work concerning information management topics will be carried out in DIGILE's Data to Intelligence research program (Forest Big Data).

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NOVEL TWO-STAGE KRAFT OXYGEN-ALKALI PROCESS (KROXY)

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ABSTRACT

The objective was to develop a delignification process concept to provide substantial pulp yield improvements. This was accomplished by a two-step process comprising modified kraft cooking followed by flow-through oxygen delignification. The kraft cooking stage was performed at a high hydroxide ion concentration of 1.55 M in the presence of oxidative additives anthraquinone [AQ] and polysulfide [PS], and was terminated at a high kappa range of 50-80 to achieve the full yield-preserving potential of this cooking concept.

To achieve the standard kappa number for final bleaching, an unusually high degree and rate of oxygen delignification is required. This was achieved by increasing the temperature above that used in conventional oxygen delignification, and keeping the caustic and dissolved oxygen concentrations at a more constant level during delignification. A combination of the described methods with the addition of viscosity-preserving additives led to an extraordinarily high degree of delignification [>60%] for high-kappa brownstock pulp [kappa 50-70], while maintaining high lignin-cellulose removal selectivity.

By utilizing the described two-stage KrOxy concept, a chip-scale demonstration with Pinus sylvestris resulted in a pulp yield increase of more than 8 percentage points for bleachable grade pulp [κ -15] with higher intrinsic viscosity. In addition, an increased amount of dissolved polysaccharides was found in the spent black liquor for recovery and utilization.

Keywords: Kraft, high yield, oxygen delignification

1. Background

Conventional pine kraft pulping is associated with substantial yield losses, mainly owing to losses of galactoglucomannan (GGM). Significant stabilization against these peeling reactions has been achieved by utilizing higher alkali concentration combined with slightly elevated temperature at the beginning of the cook (Paananen, M. 2009, Master's Thesis: M. Paananen et al. Holzforschung 2010 pp. 64, 6, 683-692). Under these conditions higher pulping selectivity can be achieved by increasing the rate of delignification and protecting the carbohydrates against degradation by hindering peeling reactions. Furthermore, different pretreatments combined with the addition of agents protecting the reducing end-groups against alkaline peeling should substantially increase the pulp yield at kappa numbers 60-100.

In addition, there is a need to continue the delignification down to a kappa number of about 15 by reinforced oxygen delignification. However, applying conventional two-stage oxygen delignification would result in significant yield and viscosity losses. To compensate for this impairment, oxygen delignification should be changed to continuous mode to ensure low but constant dissolved oxygen and sodium hydroxide concentrations.

Oxygen delignification is now commonly used as an intermediate stage between kraft pulping and final bleaching to remove 40-60% of the residual lignin in brownstock pulp. The final degree of delignification is however limited by pulp strength considerations. In the KrOxy project it was investigated whether the addition of a small amount of recovery-compatible oxidant directly after the first oxygen delignification stage in alkaline conditions could increase the degree of delignification up to 80% after a following second oxygen stage while the pulp strength is preserved at an acceptable level.

2. Objectives

Two-step cooking-reinforced oxygen delignification is a new approach to increase the yield of softwood pulp by 4-7 percentage points. In order to reach this leap it will be necessary to combine different yield-saving strategies over the entire delignification process including a radically new application for the oxygen delignification phase. The objective of the first task [Task 4.1] was to develop a more selective initial pulping phase to kappa 60-100, with adaptation of appropriate reaction conditions. The aim was to hinder the carbohydrate degradation reactions during kraft cooking through high alkalinity and/or by adding oxidative chemicals (AQ, PS) to stabilize the reducing end groups of the carbohydrates. It was also shown that cellulose chain cleavage causes pulp strength loss during oxygen delignification due to attack by oxygen-based radicals generated by the reactions. The objective of the second part [Task 4.2] was to improve lignin-to-cellulose selectivity during oxygen delignification by using a novel flowthrough reactor with elevated temperature, more constant alkali and oxygen charge, as well as radical-scavenging polymeric additives that adsorb on the cellulose surfaces.

3. Partners and their contributions

A summary of the work package partners and their contributions is presented in Table 1.

Work package partners	Role of participating organization
Aalto University / Department of	• Modified high-yield kraft pulping (Task 4.1)
Forest Products Technology	• Novel, extended oxygen delignification process (Task 4.2)
VTT	Chemical characterization of pulps and liquors
	Verification cooks at chip scale
	Evaluation of pulp bleachability and strength properties

Table 1. Partner organisations and their research roles.

4. Research approach

About 75% of galactoglucomannans (GGM) are lost during cooking, 15% through dissolution, 45% from primary peeling and the residual 15% from alkaline hydrolysis followed by secondary peeling. The aim of the first project stage (Task 4.1) was to maintain as high as possible hemicellulose yield and then continue delignification by applying a novel oxygen delignification process (Task 4.2), as illustrated in Scheme 1.

4.1 Novel high-yield pulping

As a reference for the high-yield pulping study, kinetic experiments were executed at two different hydroxide ion concentrations: in conventional cooking conditions [0.50 M [OH-], 33% sulfidity, 2.00 M [Na+], 160°C], and in high-alkali cooking conditions [1.55 M [OH-], 33% sulfidity, 2.00 M [Na+], 160°C]. In addition to hydroxide ion concentration, the effect of ionic strength, temperature [130-160°C] and additives [anthraquinone and polysulfide) for stabilizing the reducing end-groups against alkaline degradation reactions were studied. In addition to the standard compositional analysis of pulps and liquors, low-molecular weight carbohydrate derived degradation products were detected from the black liquor by capillary electrophoreses to determine the yield losses through dissolution and degradation. The molar mass of the dissolved carbohydrates was determined to evaluate the utilization potential of the dissolved material in other applications (e.g. polymeric xylan for re-precipitation



Scheme 1. Novel two-stage kraft concept (KrOxy).

on pulp fibres, or other in bio-based products]. The results can also be utilized in the development of the Virtual Chemical Pulping Model (VIC) (WP5). The overall objective of this task was to increase pulp yield from 60% to 65-70% at a lignin content of about 10-12% based on pulp (kappa number 60).

4.2 Novel high-yield oxygen delignification process

The pulp produced in the high-yield pulping task with a kappa range of 60-80 was introduced to high-selective oxygen delignification in a flow-through Berty reactor. The range of operating variables for the Berty oxygen delignification experiments are summarized as T: 95-105 °C, P: 3-10 bar O₂ and an alkali level of 1.1-3.3 g/l. The kinetics of delignification and carbohydrate degradation is examined and presented in this report. The objective of the novel oxygen process was to preserve the overall yield advantages obtained from the preceding high-yield pulping process during the high degree of delignification >60% required for typical kappa levels entering into bleaching.

5. Results

5.1 High-yield pulping

Summary of the studied high-yield pulping process conditions:

Low alkalinity, 0.50 M [OH-]:

Reference series:

- Duplicate series to evaluate the repeatability of the experimental set-up: at 160°C, 33% sulfidity, 2.00 M [Na+]
- Without ionic strength adjustment (no added NaCl)

Anthraquinone (AQ) cooking:

 1,3 and 5% AQ odw (0.05; 0.15 and 0.25 gAQ/L), at 160°C, 33% sulfidity, 2.00 M [Na+] Polysulfide (PS) cooking:

- 6 g PS/L (as elemental sulfur) at 160°C, 33% initial cooking liquor sulfidity, 0.6 M [Na+]
 Polysulfide + anthraquinone (PSAQ) cooking:
- 6 g PS/L (as elemental sulfur) + 0.15 g AQ/L
- at 160°C, 33% initial cooking liquor sulfidity, 0.6 M [Na+]
- Low alkaline sulfite cooking with anthraquinone (LASA):
- 0.50 M [OH-]; 0.375 M [SO32-]; 0.15 g AQ/L at 160°C

High alkalinity, 1.55 M [OH-]:

Without additives:

- Temperatures of 130, 140, 150 and 160°C: 33% sulfidity, 2.00 M [Na+]
- Without ionic strength adjustment (no added NaCI)

AQ cooking:

- 1,3 and 5% AQ odw (0.05; 0.15 and 0.25 gAQ/L) at 160°C, 33% sulfidity, 2.00 M [Na+]
- 5% odw AQ, at 150°C, 33% sulfidity and 2.00 M [Na+]

PS cooking:

- 6 g PS/L (as elemental sulfur) at 160°C, 33% initial cooking liquor sulfidity, 1.86 M [Na+]
 PSAQ cooking:
- 6 g PS/L (as elemental sulfur) + 0.15 g AQ/L at 160°C, 33% initial cooking liquor sulfidity, 1.86 M [Na+]

High-alkaline sulfite cooking with anthraquinone (HASA):

 1.55 M [OH-]; 1.163 M [SO32-]; 0.15 g AQ/L at 160°C

5.1.1 Materials and methods

Wood residue characterization

For the high-yield pulping method studies Pinus sylvestris raw material was acquired from a single pine stem. Chipped wood was screened (SCAN CM 40:01) and milled with a Wiley mill to pass a 1 mm hole screen. The wood meal raw material was analysed for extractives according to SCAN 49:03, and the neutral sugars, acid insoluble lignin and acid soluble lignin according to NREL/TP-510-42618. The corresponding analyses results are shown in Table 2. For each test point, 40 g of oven dry wood meal was charged into the reactor together with 8 litres of cooking liquor. The solid residue from each experiment point was analysed for yield, extractives (SCAN 49:03), kappa number (SCAN-C 1:00), viscosity (SCAN-CM 15:99) and for neutral sugars and soluble/insoluble lignin (NREL/TP-510-42618). The liquor fraction from selected samples was analysed for sulfide (SCAN-N 31:94) and residual effective alkali (SCAN-N 33:94).

Black liquor characterization

Black liquor (BL) analyses were performed to distinguish the yield losses through polysaccharide dissolution and degradation. The dissolution of hemicelluloses was evaluated according to the carbohydrate composition of BLs. Low-molecular weight organic acids were detected by capillary electrophoresis (CE) to evaluate polysaccharide losses through peeling. Some scattering of the CE results was inevitable due to the low quantities of the detected acids as a result of the high liquor-to-wood ratio, as well as the salt (CI-, SO42-) removal treatment required before the CE measurements.

5.1.2 Effect of alkali concentration during kraft pulping

Wood residue

The wood residue was analysed with an emphasis on evaluating the effect of cooking conditions on delignification and carbohydrate retention. The increase in alkali concentration from 0.50 to 1.55 M [OH–] resulted in a substantial acceleration in the delignification rate, thus allowing a decrease in cooking temperature without substantially prolonging pulping time. In order to obtain 80% lignin removal in kraft pulping to a kappa number of 60 (κ -60), the H-factor was decreased from 1480 to just 300 when the hydroxide ion concentration was increased from 0.50 to 1.55 M. To obtain a kappa number of 30 (κ -30), which corresponds to the conventional production of bleachable softwood kraft pulps, the H-factor can be decreased from 2370 to only 480, owing to higher alkalinity. Regardless of significantly altered delignification rate, we could not observe any significant effect of hydroxide ion concentration and temperature on the yield of wood residue throughout the entire delignification phases. The present study revealed that the advantage of elevated [OH-] on GGM yield in the wood residue increased with temperature but was more pronounced at lower delignification degrees. At a Klason lignin content of 10% based on pulp, 1.55 M [OH–] results in a 1.4 percentage point higher glucomannan content compared to 0.50 M [OH-], regardless of temperature. Even though the overall pulp yield is hardly affected, the ratio of GGM to AX is substantially altered by kraft cooking at high alkali concentration. This is accomplished by substantial GGM stabilization, while AX is eventually lost through dissolution.

Black liquor

Black liquor samples throughout cooking were analysed to allocate the polysaccharide losses

Acetone extractives, % based on wood	3.2
Gravimetric lignin, % based on wood	25.9
Soluble lignin, % based on wood	0.7
Cellulose, % based on wood	41.7
Glucomannan, % based on wood	16.9
Xylan, % based on wood	8.2
Other sugars, % based on wood	1.4

Table 2. Composition of Pinus Sylvestris wood rawmaterial.

to either dissolution or degradation reactions. The main organic acids formed as a result of peeling reactions of pulp polysaccharides are the isosaccharinic acids (glucoisosaccharinic acid from cellulose and GGM, and xyloisosaccharinic acid from xylan), lactic acid, as well as 2-hydroxybutanoic acid (from xylan) and 2,5-dihydroxyoxypentanonic acid (from cellulose and GGM]. The total amount of carbohydrates in the black liquor was significantly increased due to dissolution promoted by elevated cooking liquor alkalinity. Prolonged cooking times at higher alkalinity resulted in a decrease in the amount of dissolved xylan, indicating an increase in competitive degradation pathways of the dissolved xylan. The maximum amount of dissolved polysaccharides was achieved at Klason lignin contents of 15% and 6% based on pulp for 0.50 and 1.55 M [OH–], respectively. For both studied alkali concentrations, the decrease in the amount of dissolved xylan is accompanied by enhanced formation of 2-hydroxybutyric acid and lactic acid. Xyloisosaccharinic acid formation via β -alkoxy elimination, followed by keto-enol tautomerization and benzilic acid rearrangement, was clearly enhanced at lignin contents below 6%, suggesting xylan degradation. However, the formation of glucoisosaccharinic acid originating from cellulose and glucomannan was also significantly enhanced at lignin contents below 4%. Clearly less lactic and 2-hydroxybutyric acids were formed at the same pulp yield or degree of delignification for all black liquors derived from 1.55 M [OH-], as compared to 0.50 M [OH-] cooking conditions. Overall, lower amounts of hydroxy acids were formed at high alkalinity between 140 and 160°C at a given degree of delignification. This suggests that the degradation of polysaccharides occurs to a lower extent, supporting our hypothesis that stopping reactions are enhanced over peeling reactions at higher alkali concentration. Therefore, maintaining an elevated alkali concentration and shorter cooking times is proposed to achieve a higher concentration of dissolved high molecular weight hemicellulose at the same kappa levels, thus providing a means of enhancing the re-deposition of dissolved hemicelluloses on fibres through liquor recirculation.

5.1.3 Kraft pulping in the presence of oxidative additives

Oxidative additives: Wood residue

The effect of AQ and PS on carbohydrate degradation and dissolution behaviour was evaluated separately and in combination at constant 33% sulfidity and 160°C temperature with 0.50 M [OH–] and 1.55 M [OH–]. Figure 1 illustrates a substantial pulp yield improvement when oxidative additives such as polysulfide and anthraquinone are present in the white liquor. The delignification rate was also greatly accelerated using AQ or PS in combination with 0.50 M [OH-]. However, only PSAQ at 0.50 M [OH-] reached the delignification rate as obtained at elevated base concentration without additives at 160°C (Figure 2). Our results indicate that even a small AQ addition provides a notable delignification rate promotion at moderate hydroxide ion concentration, whereas at elevated alkalinity, higher AQ concentrations are required to recognize a substantial additional H-factor reduction. The effect of AQ on carbohydrate stabilization was already evident at the lowest studied charge of 0.05 g AQ/L, but major differences clearly occurred at the highest charge of 0.25 g AQ/L. At κ -30 (4.5% lignin based on pulp), PSAQ together increased GGM retention to an impressive 70% at 0.50 M [OH-], whereas analogously 1.55 M [OH-] resulted in a 55% retention only, which corresponds to a 2.5 percentage point lower pulp yield (Figure 3). As expected, the removal of xylan was enhanced by increasing hydroxide ion concentration (Figure 4). Interestingly, at both studied hydroxide ion concentrations the combined addition of polysulfide and an-



Figure 1. Effect of alkalinity and oxidative additives on pulp yield, high and conventional [OH-] at 160°C.



Figure 3. Effect of alkalinity and oxidative additives on GGM retention, high and conventional [OH-] at 160°C.

thraquinone provided no additional improvement in xylan yield compared to polysulfide alone. Different to glucomannan, xylan is more retained by the presence of oxidizing additives at high alkalinity as compared to moderate alkalinity conditions. This is particularly true at higher kappa numbers.

Oxidative additives: Black liquor

Any charge of anthraquinone, polysulfide alone or in combination with anthraquinone, clearly increased the amount of dissolved polysaccharides (Figure 5) and decreased the amount of hydroxy acids found in the black liquor (Figure 6) at both studied hydroxide ion



Figure 2. Effect of alkalinity and oxidative additives on delignification rate, high and conventional [OH-] at 160° C.



Figure 4. Effect of alkalinity and oxidative additives on xylan retention, high and conventional [OH-] at 160 $^\circ\text{C}$

concentrations. Interestingly, high alkalinity without any additives provided higher carbohydrate content in BL for recovery and re-precipitation than could be obtained by addition of PS, AQ or AQPS in conventional cooking conditions at 0.50 M [OH-]. Additionally, in the presence of anthraquinone, the degradation of dissolved xylan resulted in a lower pulp lignin content than without AQ. Owing to this, the maximum xylan concentration in black liquor was detected at 4–6% pulp lignin content, as was also the case at high alkalinity without any addition of anthraquinone. Regardless of the alkalinity, the maximum molar mass of 35 kDa was detected at a pulp lignin content of 7%, after which the molar mass of the precipitated xylan decreased significantly. In GGM stabilization AQ was more efficient than PS regardless of the alkalinity. However, the xylan stabilizing effect of AQ and PS were comparable at both alkalinity levels. The combined use of PS and AQ at high alkalinity provided the highest BL polysaccharide content. The synergistic effect was most pronounced at a kappa number of 30, preserving up to 11% of wood carbohydrates in BL. At lower alkalinity the synergistic effect of AQ and PS was realized already at higher pulp lignin contents.

Efficient xylan stabilization in black liquor owing to oxidative additives was also supported by the lower amount of detected acidic degradation products. Based on total formation of the acidic degradation products, the combination of PSAQ at both alkalinity levels reduced the peeling reactions most efficiently. At conventional alkalinity, the combined effect of AQ and PS was insignificant compared to PS alone, whereas at high alkalinity AQ offered an additional beneficial effect. The formation of lactic acid was significantly reduced in the presence of PS, and both the high alkalinity and the combined use of AQ further reduced the formation. The formation of 2-hydroxybutyric acid originating from xylan was significantly reduced in all experiments including anthraquinone, and no 2-hydroxybutyric acid could be detected when anthraquinone and high alkalinity were combined. The beneficial effect of anthraguinone and polysulfide was also evident according to the degradation products originating from cellulose and galactoglucomannan. Although high alkalinity alone had no effect on the formation of 2,5-dihydroxypentanoic acid, this degradation product was not detected at all in the presence of either AQ or PS. In general, anthraguinone addition also reduced the total formation of lactic and isosaccharinic acids. However, it was not as effective as PS in preventing the formation of isosaccharinic acids. In this case, the combination of PS with high alkalinity or AQ provided no additional benefit.



Figure 5. Dissolved polysaccharides in BL as a function of pulp lignin content in presence of AQ and PS.



Figure 6. Total content of low-molecular weight acids originating as a result of peeling reactions (xylo- and glucoisosaccharinic acids, lactic acid, 2-hydroxybutyric acid and 2,5-dihydroxypentanoic acid).

Overall mass balance

Over the entire studied kappa number range, higher alkalinity conditions during kraft cooking revealed a substantial net carbohydrate yield increase (pulp + black liquor) compared to conventional kraft cooking conditions. This was mainly owing to the significant enhancement in the dissolved hemicellulose fraction. At a temperature of 160°C and a pulp lignin content of 9% (corresponding to κ -60), the studied hydroxide anion concentration of 1.55 M had a 3.4 percentage point higher net carbohydrate yield than conventional 0.50 M. At κ -30, the advantage of elevated alkalinity was even further emphasized, as the net carbohydrate yield describing the amount of recoverable polysaccharides was 4.1 percentage points higher compared to the amount at 0.50 M hydroxide ion concentration. In the presence of oxidative additives, the amount of dissolved polysaccharides in black liquor was further increased regardless of alkalinity. However, in analogous conditions and kappa number, the studied higher alkali concentration always resulted in black liquor with a higher dissolved polysaccharide fraction. The increased amount of dissolved polymeric sugars found in the black liquor offers an improvement in overall carbohydrate yield. Enhanced utilization of the dissolved polysaccharide fraction would require recycling of the carbohydrate-enriched lye into the later pulping stages to initiate partial re-precipitation or to collect the dissolved polysaccharides by other means, for example, membrane filtration.

5.1.4 HALT pulping

As previously shown, at 160°C, the studied conditions of 1.55 M [OH-] without and in the presence of oxidative additives provide a significantly accelerated delignification rate compared to 0.50 M [OH-] conditions. At the given kappa number of 60, compared to reference conditions at conventional alkalinity at 160°C, experiments at higher hydroxide ion concentration and significantly lower cooking temperature (HALT) revealed a yield increase of 7.8% and 11.3% in the presence of single and two oxidants, respectively. The obtained yield increase was a result of improved retention of all softwood carbohydrates in the pulp. As a consequence of the improved overall carbohydrate retention and, especially, cellulose retention, an increase in pulp viscosity was also observed. In addition, compared to conventional pulping conditions, under HALT conditions it is possible to protect the dissolved polysaccharides against degradation, thus increasing the amount of polymeric polysaccharides in the black liquor. While having somewhat high molecular weight, the amount of dissolved polysaccharides increased from 2.2% to 5.5% at kappa number 60, and even further as the pulping was extended to lower kappa numbers. It should be emphasized that without increasing the cooking time, the described HALT pulping concept did not only provide pulp with increased yield and viscosity, but it also reduced carbohydrate degradation in the pulp and black liquor, thus decreasing the amount of produced hydroxy acids.

5.1.5 Recovery of dissolved wood fraction

The dissolved wood polymers, e.g. xylan, were recovered from the high alkalinity black liquor by diafiltration, as illustrated in Figure 7. Black liquor was first fed from the black liquor tank to an ultrafiltration unit, where membranes were used to quantitatively retain the dissolved wood polymer while a maximum amount of lignin passed the membrane. To minimize NaOH losses, the retentate R1 was diluted with water so that the xylose-to-NaOH ratio in the retentate 2 (R2) approached a maximum while keeping the polymer well in solution. The results of the diafiltration experiments are presented in Table 3.

The results confirm that the xylan can be quantitatively retained with relatively open membranes. Thereby, the sodium hydroxide losses are lower than 5% of that in the feed concentration. Although lignin separation could be improved with increased membrane cut-off, a relatively high proportion of lignin remained in the retentate. The retentate R2 can be utilized as an alkali source for oxygen delignification whereby a high proportion of the xylan precipitates on the fibre surface. However, to ensure efficient delignification, low lignin contents should be reached in the R2 retentate: 14 g/l lignin with a 100 kDa membrane is approximately low enough. Alternatively, the xylan can be precipitated by addition of a non-solvent alcohol, e.g. isopropanol, or mineral acid, e.g. sulfuric acid. Preliminary tests showed that xylan can be quantitatively precipitated by either type of non-solvent comprising a lignin content lower than 20% in the crude sample without further purification, ensuring that the precipitation pH remains high enough. Relatively pure lignin, >75%, can be obtained by the addition of a surplus of nonsolvent to the supernatant of the first precipitate. The permeates P1 and P2 can be recycled as a sodium hydroxide source for the cooking and oxygen delignification stages.

Table 3. Diafiltration results with ultrafiltration membranes of three different cut-offs.

Membrane		l0kDa ^A	20 kDa [∆]	50 kDa ^A	100 kDa [∨]
T, °C		30.0	30.0	30.0	50.0
p inlet, bar		6.0	4.4	4.4	3.0
VR1		0.73	0.67	0.64	0.81
VR2		0.81	0.82	0.82	0.90
Dilution of F2	H ₂ 0:R1	5.48	4.21	3.34	6.02
NaOH, %*	P1	75	68	66	78
	P2	14	24	23	19
	R2	3	5	5	3
Xylan,%*	P1	1	2	4	3
	P2	1	1	4	7
	R2	91	95	100	60 [#]
Lignin, %*	P1	26	29	38	52
	P2	13	20	25	27
	R2	58	49	36	14

VR, the volume recluction = Vp/V0, *compound in percentage of the amount in feed, F1 ^A filtration with Alfa Laval LabStack M20 at Aalto, ^vfiltti with Alfa Laval DSS M20 at VTT, [#]Part of the xylan might he precipitated on the surface of the membrane due to very low alkali concentration [3 g NaOH/I] in retentate 2.



Figure 7. Diafiltration concept for the recovery of xylose from black liquor.

5.2 Novel oxygen "pulping"

5.2.1 High-yield oxygen delignification of high-kappa pine kraft pulp with Peroxymonosulfate (Px)

The advantages of multistage oxygen delignification are explored in comparison with a single-stage process at the same level of total caustic charge. A pine kraft pulp with a kappa number of 65 was bleached and the effect of NaOH charge distribution compared to kappa number, pulp yield, and selectivity of oxygen delignification was studied. The effect of initial chelation with ethylenediaminetetraacetic acid (EDTA) and interstage pulp treatment with peroxymonosulfuric acid (Px) were also investigated. It was shown that the chelated pulp with high kappa number can be delignified in three stages to K-15, but at an unacceptable reduction in pulp viscosity and yield has to be taken into account. In contrast, interstage treatment of the chelated pulp with Px at a total charge of 2% active oxygen (based on pulp) in combination with three stages of oxygen delignification enables a kappa number reduction to about 15 at acceptable viscosity and a total yield advantage of about 0.5% (based on wood) compared to oxygen-delignified kraft pulp with K-24 derived from the same pine wood chips.

The results show a kappa number reduction of 76% for a pine kraft pulp of initial kappa 65 after a three-stage oxygen delignification with interstage peroxymonosulfate (Px) treatment (i.e. OPxOPxO) and 6% total alkali charge and 2% (as active oxygen) total Px charge in the batch reactor (air-bath digester). The operational conditions are shown in Table 4.

By distributing the alkali over the three stages and applying an initial chelation stage, a pulp with kappa 15.4 and adequate viscosity level of 910 ml/g was produced. Without Px interstage treatment (i.e. OOO) a less delignified and lower viscosity pulp (kappa 19.9 and viscosity 840

 Table 4. Conditions of different oxygen delignification experiments (peroxymonosulfate applied represented as Px).

Sequences	NaOH charge (% od)	PMS charge (% od)	Reaction time (min)	T[°C]	P(bar)
OPx, O HK (high kappa 65)	6	2	30	85	7
OPxO, OO HK (high kappa 65)	4	2	30	85	7
	2	0	60	95	7
ОРХОРХО, ООО НК	3	2	30	85	7
	2	2	60	95	7
	1	0	60	95	7
00 –LK (low kappa 24)	3	0	30	85	7
	1	0	60	95	7

ml/g) was obtained. The lignin-free yield of the OPxOPxO pulp derived from the high-kappa (65) pulp is about 0.5% higher than that of a conventionally kraft cooked [kappa 24] and then oxygen delignified pulp derived from the same wood furnish at a final kappa number of about 15. The Px treatment leads to increased delignification and [delignification/cellulose degradation] selectivity owing to higher reactivity of residual lignin. Removal of heavy metal ions from pulps prior to the O-stage by means of a chelating agent treatment improves the selectivity for the sequences with Px treatment. The selectivity plot for these reactions is shown in Figure 8.



Figure 8. Effect of alkali distribution, chelation and Px treatment on selectivity for high-kappa pulp.

6. Exploitation plan and impact of results

We have shown that the proposed novel pulping concept significantly improves mill economy through enhanced raw material utilization and decreased energy consumption. From the biorefinery point of view, the dissolved hemicellulose fraction provides an opportunity to replace oil-based platform chemicals.

So far, the majority of research on pulping and oxygen delignification has been directed at finding the most suitable conditions for increasing pulp yield. Future work is needed to demonstrate our concept at larger than lab scale. Process scale-up is therefore needed to overcome the technological limitations at the mill scale. Additionally, the bleachability of the high-yield pulp must be investigated using both ECF and TCF sequences as well as the mechanical properties of the final bleached pulps.

7. Publications and reports

Journal papers:

Jafari, V., Sixta, H. and van Heiningen, A. High yield oxygen delignification of high-kappa pine Kraft pulp with Peroxymonosulfate (Px). Holzforschung, accepted.

Jafari, V., Sixta, H. and van Heiningen, A. Kinetics of oxygen delignification of high-kappa pulp in a continuous flow-through reactor. Submitted to Industrial & Engineering Chemistry Research Journal.

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Nieminen, K., Kuitunen, S. and **Sixta, H.** Novel insight in lignin degradation during Kraft cooking. Submitted to Industrial & Engineering Chemistry Research Journal, 2013.

Nieminen, K. and **Sixta, H.** Comparative evaluation of different kinetic models for batch cooking: A review. Holzforschung 2012, 66, 791-799. **Paananen, M., Liitiä, T.** and **Sixta, H.** Further Insight into Carbohydrate Degradation and Dissolution Behavior during Kraft Cooking under Elevated Alkalinity without and in the Presence of Anthraquinone. Industrial & Engineering Chemistry Research Journal, 2013, 52, 12777–12784.

Paananen, M., Tamminen, T., Nieminen, K. and **Sixta, H.** Galactoglucomannan stabilization during the initial kraft cooking of Scots pine. Holzforschung 2010, 64, 6, 683-692.

Conference presentations:

Jafari, V., Sixta, H. and van Heiningen, A. Kinetic study of oxygen delignification of highkappa pine Kraft pulp in a through-flow reactor. The 17th International Symposium on Wood, Fibre and Pulping Chemistry, June 2013, Vancouver, Canada. (oral presentation)

Jafari, V., T., Sixta, H. and van Heiningen, A. Bleachability of conventional and high alkali cooked softwood kraft pulps of different kappa numbers during oxygen delignification in a flow-through reactor. Appita conference, 2013, Melbourne, Australia. [poster]

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Jafari, V., Sixta, H. and van Heiningen, A. Comparison of oxygen delignification of high-kappa pine Kraft pulp in through flow and batch reactors, 4th NWBC conference proceeding, 2012. [poster] **Paananen, M., Liitiä, T., Rovio, S.** and **Sixta, H.** Effect of alkali concentration in polysulfide pulping with and without anthraquinone on polysaccharide degradation and dissolution. Conference proceedings in 17th International Symposium of Wood, Fibre and Pulping Chemistry, June 2013, Vancouver, Canada. (oral presentation)

Paananen, M., Liitiä, T., Rovio, S. and **Sixta, H.** Effect of alkali concentration in kraft pulping with or without anthraquinone addition on polysaccharide degradation and dissolution. 12th EWLP conference proceedings, 2012. [poster]

Reports and Thesis:

Montagna, P. N. Master's Thesis: Kinetics and effective capillary in Kraft impregnation of Pinus sylvestris. ITC, Instituto de Tecnologia Celulosica, University of Santa Fe, Argentina, 2013.

Toivari, T. Master's Thesis: Modification of the displacement batch cooking concept to highalkali cooking. Department of Forest Products Technology, Aalto University, 2013.

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VIRTUAL CHEMICAL PULPING MODEL

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EFFFIBRE PROGRAMME REPORT

ABSTRACT

In this project, unit operation models for batch and continuous digesters using wood chips as raw material were developed. The unit operation models consist of phenomenon-based sub-models for delignification and carbohydrate degradation kinetics, reaction equilibria, fibre wall ion exchange, and mass transfer. The stoichiometry and Arrhenius parameters for lignin degradation reactions are based on the literature. The parameters for dissolution of degraded lignin from the fibre wall into the liquor were regressed using data from the KrOxy project. Comparison between the model output and experimental data show that the modelling of lignin removal is not yet sufficiently reproduced by the model. On the other hand, carbohydrate degradation, based on the experimental data produced in KrOxy, was very well modelled. The mass transfer model was validated with the literature data concerning kraft liquor impregnation of eucalypt wood.

Many features of the cooking-related model (VIC) can be applied to specific pulping chemistry considerations. Besides describing pulp manufacturing phenomena, the VIC model serves as a solid basis to be extended to more generally applicable model covering wood raw material fractionation into main wood components (biorefinery platform). The developed mass transfer model is available for digester studies. The simplification and significant speeding up of calculation procedures has made the developed VIC model much more attractive and useful for modelling bleaching stages and whole bleaching plants.

Keywords:

modelling, pulping, mass transfer, reaction kinetics, wood, ion exchange, digester

1. Background

The Virtual Pulping (VIP) model developed in the first stage of the EffTech programme is a virtual tool for simulation of pulp bleaching bevond experimental limits. The model combines state-of-art knowledge on physicochemical phenomena (mass transfer, chemical equilibriums, reaction kinetics, etc.) with the practical unit processes (mixing, washing, retention towers, etc.] used in the building of complete bleaching sequences. The VIP model thus serves as an encyclopaedia of pulp bleaching that is an equally powerful tool for the scientist in exploring unknowns as it is for the engineer in predicting the effects of process design and parameters on bleaching results and process impacts.

The Virtual Chemical Pulping (VIC) model broadens the scope of VIP to chemical pulping of wood. Similar to bleaching modelling, the motivation for the extension is two-folded. Gathering state-of-art knowledge in a virtual encyclopaedia makes it possible to broaden and deepen understanding of pulping at the phenomenal level. This understanding is needed for the creation of new pulping concepts that meet the criteria for lower production costs (pulping yield, energy consumption, etc.) and/or higher product value (pulp, energy, chemicals, etc.). In addition, the model can be used in predicting the outcomes of various process designs and changes in pulping parameters.

2. Objectives

The primary objective of work package was to create in-depth knowledge on existing and possible new cooking processes. The delignification of wood chips is a difficult process to comprehend for many reasons. Even in the hypothetical case of perfect chip penetration, at least 50% of the cooking liquor and chemicals remain outside the chips. The additional mass transfer between the free liquid and the already impregnated chips, mostly by diffusion, thus limits the rate of delignification. The fast chemical reactions, in comparison with the slow mass transfer, produce steep gradients in the chemical environment (e.g. pH) that lead to uneven delignification within the chips.

In addition to the boundaries set by the slow mass transfer, the innumerable simultaneous chemical reactions taking place among the wood constituents limits our ability to fully understand the complex nature of delignification. In order to achieve deeper knowledge, computational modelling and simulation of the transport phenomena and chemical reactions are needed. Thus, the secondary objective of work package was to create a tool for understanding the behaviour of wood chips and their constituents in a wide variety of chemical conditions.

3. Partners and their contributions

A summary of the work package partners and their contributions is presented in Table 1.

Work package partners	Role of participating organization
Aalto University, Department of Forest	Work package 5 and Task 5.1 were coordinated by
Products Technology (Professor Tapani	Professor Tapani Vuorinen. Olesya Kuzmenko and
Vuorinen]	Elina Warsta (until end of October 2011) worked as
	doctoral students. The team specialized in the che-
	mistry of pulping and bleaching.
Aalto University, Department of Bio-	Professor Ville Alopaeus was responsible for Task
technology and Chemical Technology	5.2 and Susanna Kuitunen and Juha Visuri (until the
[Professor Ville Alopaeus]	end of 2011) worked as doctoral students. The team
	specialized in the modelling of chemical processes.
University of Jyväskylä, Laboratory of	Professor Raimo Alén was responsible for Task 5.3.
Applied Chemistry (Professor Raimo	Teemu Paloheimo (until the end of December 2011)
Alén]	and Vesa Nykänen (from February 2012) worked as
	doctoral students. The team specialized in wood and
	non-wood analyses, characterization of process stre-
	ams and chemistry of delignification and bleaching.

Table 1. Partner organisations and their research roles.

4. Research approach

The topics studied in this project were related to modelling of 1) irreversible chemical reactions, 2) mass transfer, and 3) continuous digester unit.

Irreversible chemical reactions [1] leading to delignification and carbohydrate losses are the core of the model. The reaction stoichiometry and rate laws were examined from the literature. When available, the Arrhenius law kinetic parameters were taken from the literature. If the parameters were not available, experimental data from the literature, own experimental data, or data provided by the KrOxy project were used for regressing the parameters so that the model behaviour corresponded to the experimental observations. Real compounds were favoured in the modelling, although lignin and some of its degradation products needed to be modelled as pseudocompounds closely resembling the real structures detected directly from lignin-polymer or from lignin model compound studies. Furthermore, the aim was to build the reaction chemistry library from elementary reactions, but some of the reactions combine many elementary reaction steps to keep the library simple enough. Not all possible reaction routes were included in the model, but rather the aim was to identify the most relevant ones. The main focus was on kraft pulping reactions, since this the most used pulping process in the world.

In comparison to the reaction rates, mass transfer in chips is rather slow and limits the availability of chemicals at the reaction site. Its precise modelling is therefore important. Modelling of mass transfer in chips is based on the novel idea that chips consist of two liquid phases (lumen liquid and fibre wall liquid). Chip-scale mass transfer takes place only in the lumen, and at each location inside the chips, the mass transfer takes place also between the lumen and fibre wall liquid. This new approach enables modelling of the ion exchange property of wood. Ion exchange influences the local chemical concentrations in the fibre wall and, consequently, the reaction kinetics. The concentration profiles in the rectangular particles are traditionally solved based on equations written for several grid points, resulting in high computer CPU and memory consumption. These challenges were overcome by applying modelling methods traditionally applied in the catalyst field.

In order to apply the modelling results in practice, models for commonly used digester units, such as the continuous digester model, are needed. For modelling momentum balances, which determine the chip column and the liquor velocities in the continuous digester unit, the models published in the literature were applied. To further facilitate use of the models, the graphical user interface developed by the VIP project was updated with VIC models. Additionally, to enhance communication between scientists and engineers, several engineering parameters were computed from the detailed chemical composition of liquors and wood/ pulp. These include kappa number, intrinsic viscosity, brightness, yield, active alkali, effective alkali, sulfidity, higher heating value, etc.

5. Results

5.1 Chemical reaction library

The chemical reaction library includes reactions for lignin, galactoglucomannan, xylan, and cellulose.

The components required for the pulping simulation were added to the component database, and the reactions were implemented in the VIC simulator.

5.1.1 Reactions of lignin in alkaline conditions

The work carried out for modelling delignification in alkaline conditions can be divided into three parts. Firstly, the literature concerning reactions of lignin model compounds in soda and kraft pulping conditions was studied to determine the relevant reaction mechanisms. The Arrhenius reaction kinetic parameters were obtained directly from the literature, or the experimental data available in the literature were used in the parameter regression. Secondly, a model of the dissolution of degraded lignin polymers from the fibre wall into the filtrate was developed and the necessary model parameters were regressed using experimental data produced in the KrOxy project. Data published in the literature was then used for model validation. Thirdly, the reactions of non-phenolic lignin units with α -carbonyl functionality were studied experimentally, and the model was further developed based on these results.

In view of the complexity of the real lignin structure, consisting of various types of ether and carbon-carbon bonds, a large number of possible degradation reactions are possible. For modelling purposes, a simplified lignin structure and a reaction scheme was used. Units in the lignin polymer were modelled as monomeric pseudo-compounds [single phenylpropane units]. Each pseudo-compound represents a group of real lignin structures with similar chemical properties. In the model, the native lignin is assumed to consist of phenolic and non-phenolic units, in addition to a few lignin structures with α -carbonyl functionality. The aim of the modelling was to determine the reactions of these native lignin structures and their reaction products.

Degradation of lignin units

In the development of a kinetic reaction library for the degradation of lignin, several literature sources were used. The main focus was to determine the mechanisms and kinetics of the following reactions (see Figure 1):

- degradation of phenolic lignin units in soda, soda-AQ, and kraft pulping conditions
- degradation of non-phenolic lignin units in soda pulping
- degradation of -carbonyl group containing lignin in kraft and soda pulping
- demethylation of lignin units

The simulation results with the developed model and measured values are given in Figures 2-4.

Dissolution of lignin units

The macromolecular nature of lignin was taken into account in modelling the dissolution of lignin. The logic used in developing the lignin dissolution scheme was the following:

- the dissolving "lignin polymer fragment" is the sum of monomeric units participating in the "dissolution reaction"

- this polymer fragment needs to include
 - non-phenolic units that are no longer
 - attached to the next lignin unit via $\beta\text{-carbon}$
 - ionized phenolic unit that makes the fragment water soluble

The dissolution stoichiometry was fitted with Kinfit software using data from the KrOxy project. For model validation, the data published by Bogren et al. (2007) were simulated. Both



Figure 1. Lignin unit reactions included in the model: a) phenolic lignin units in soda and kraft pulping; b) non-phenolic units in alkaline pulping; c) non-phenolic units with α -carbonyl in soda and kraft pulping; and d) demethylation of lignin units.



Figure 2. a) Formation of β -aroxy styrene structure from reaction of phenolic lignin model compound [phenolic lignin units] 0=3.4 mmol/l, [OH-]=0.1 mol/l (experimental from Kondo et al. (1987]); and b) formation of thiirane structure from phenolic lignin model compound [phenolic lignin units] 0=3.4 mmol/l, [OH-]=0.1 mol/l, [HS-]=0.005 mol/l (experimental from Gierer and Ljunggren (1979a) and Gierer and Ljunggren (1979b)].



Figure 3. Formation of oxirane intermediate from degradation of non-phenolic lignin structures a) in soda pulping (experimental from Miksche (1972) and Miksche (1975)] [non-phenolic lignin unit]=3.3 mmol/l dissolved in water and ethylene glycol monomethyl (70% and 30%, respectively), [OH⁻]=0.52 mol/l]; b) in soda-AQ pulping (experimental from Brunow (1981)] [non-phenolic lignin unit] = 8,2 m mol/l dissolved in water and dioxane (60% and 40%, respectively), [OH-]=1 mol/l, [AHQ diacetate]=0.18 mol/l.

setups used wood meal as raw material. Thus, it was assumed that mass transfer restrictions were negligible.

Below (Figures 5–9), the simulated lignin yields are compared with the measured data. The simulations reveal that the assumption of equal reactivity of all ether bonds between the non-phenolic lignin units is not sufficient and that the reaction kinetic parameters obtained from the model compound studies need to be modified. The stoichiometry used in the dissolution reactions should be further validated with measurements of the kraft lignin chemical structure.

Experimental study on reactions of non-phenolic lignin units with α -carbonyl functionality

Experiments were carried out with a lignin dimer (adlerone) representing α -carbonyl containing β -O-aryl ether structures in native lignin. It is well known that these structures react very rap-



Figure 4. Demethylation (experimental from William et al. (1965), William et al. (1967), and William et al. (1968)] in kraft pulping. Western hemlock meal with methoxyl value 4.51% (1.45 mol/kg fibres), [HS-]=0.96 mol/l, [OH⁻]= 3 mol/l.



Figure 5. Experimental (Bogren et al. (2007) and simulation results [OH]=0.78 M, [SH]=0.26 M).



Figure 6. Experimental (Bogren et al. (2007) and simulation results [OH]=0.26, [SH]=0.26).



Figure 7. Experimental (Bogren et al. (2007) and simulation results [OH]=0.1 M, [SH]=0.25 M, 168 and 154 C.



Figure 8. Experimental (Bogren et al. (2007) and simulation results [OH]=0.25, [SH]=0.1).



Figure 9. Experimental and simulation results (Kroxy project, [OH]=1.55, [SH]=0.31, [CI]=0.14, 168 and 154 C).

idly in alkaline pulping conditions. The content of α -carbonyl groups in wood is low; however, this work represents how a possible increase in the amount of carbonyl groups in wood lignin could enhance the speed of the pulping process with commonly used nucleophiles.

Knowledge of the kinetics of α -carbonyl lignin unit degradation is not complete. Thus, the kinetics of adlerone's reactions with different nucleophiles, i.e. hydroxide, hydrosulfide, sulfite, and hydroperoxyl ion were studied. The following reaction scheme was assumed:

Firstly, adlerone reacts with hydroxide ion and water is eliminated, forming an unsaturated product. This is a relatively stable compound. When strong nucleophiles (hydrosulfide, sulfite, or hydroperoxyl ions) are added, the elimination product reacts further.

The kinetics of the first reaction step was first followed. After adlerone's initial conversion, the strong nucleophile was added and the kinetics of this secondary reaction step was then followed. Kinetic parameters for these secondary reactions have not been previously reported.

The kinetic experiments and the analyses conducted with GC-MS confirmed the reaction mechanism previously suggested by Gierer et al., although the existence of parallel mechanisms could not be fully excluded. Based on results from GC-MS, β -aryl ether bond cleavage takes place immediately after addition of hydrogen sulfide ion into the solution containing the β -elimination product.

5.1.2 Reactions of carbohydrates in alkaline conditions

Reactions of galactoglucomannan

Native galactoglucomannan (GGM) polymers were modelled as a reducing end group (GGMR), non-reducing end group (GGMNR) and units in the middle of the chain (GGMM). One end of the GGM polymer was assumed to be reducing and the other end non-reducing. The degree of polymerization of GGM in wood was assumed to be equal to 102.

In alkaline conditions, the reducing end groups and middle units have reversible acid-base equilibrium reactions. According to Young and Liss (1978) and Paananen et al. (2010), ionized reducing end groups undergo peeling or stopping reactions. The bond next to the ionized middle units may also encounter an alkaline hydrolysis reaction. However, this scheme, presented in the literature, did not give satisfactory results. By adding a new stopping reaction, the formation of metasaccharinic acid is possible also at lower alkali concentration. This significantly improved the fit of the model to the experimental data.

The experimental data used in the parameter optimization were obtained from the KrOxy project. When the parameters for GGM degradation were regressed, alkali-catalyzed deacetylation and cleavage of uronic acid esters were included in the simulation. The model output and the experimental values are shown in Figure 10.

Reactions of xylan

Xylan degradation follows similar routes to galactoglucomannan. In the model, it was assumed that arabino [4-O-methylglucurono] xylan has two kinds of side-group. Of these, 4-O-methylglucuronic acid side groups were treated as individual compounds, whereas the arabinose side group was considered as a leaving group that enhances the conversion of a reducing end group in xylan into a stable metasaccharinic acid group through the stopping reaction. During pulping, a considerable amount of xylan is dissolved as a polymer. This feature was included in the model as well.

Measurements concerning the uronic acid



Figure 10. Simulated (lines) and experimental (points) GGM yield.

content of wood can be found from the literature. In the modelling, the degree of polymerization of xylan was assumed to be 106. One end was assumed to be reducing and the other non-reducing.

Like galactoglucomannan, xylan was modelled to react according to stopping, and alkaline hydrolysis mechanisms.

The stoichiometries slightly differ from those used for galactoglucomannan. The formation of

arabinose in peeling and alkaline hydrolysis reflects the fact that every 10th xylan middle unit has an arabinose side-group. When the arabinose unit is in the reducing end unit it enhances the formation of stable xylometasaccharinic acid.

Dissolution of xylan as a polymer was described such that all components and their ionized forms dissolve at an equal rate. The dissolved xylan degrades at the same rate as the fibre-bound xylan. Possible readsorption of dissolved xylan was not considered.



Figure 11. Simulated (lines) and experimental (points) xylan yield in fibre wall (upper) and in filtrate (lower).



Figure 12. Simulated (solid lines) and Bogren-model (points) HexA concentrations as a function of time. The effect of temperature and hydroxyl ion and hydrosulfide ion concentration on evolvement of HexA.

sim [OH-]=0.77

sim [OH-]=0.51 mol/kg

sim [OH-]=0.25 mol/kg

Bogren-model [OH-]=0.51 mol/kg,

Bogren-model

Bogren-model

<u>sne</u>

[OH-]=0.77 mol/kg,

[OH-]=0.25 mol/kg,

mol/kg

Simulation of the degradation and dissolution is shown in Figure 11. The experimental data used in the parameter optimization were obtained from the KrOxy study. As can be seen, the xylan yield is not well modelled at lower alkalinity. Furthermore, the xylan polymer concentration in the filtrate is not well modelled.

4-Deoxy-4-hexenuronic acid (HexA) is formed from 4-O-methylglucuronic acid (MeGIc) and degrades as 4-oxo-4H-pyran-2-carboxylic acid (comanic acid, COMA). In alkaline conditions, both uronic acids are in dissociated form (their pKa values are about 3) contributing to the ion exchange (the Donnan effect).

HexA profiles simulated with the VIC model (reaction kinetic parameters were taken from Bogren et al. 2008) fit better with Bogren's experimental data than the HexA profiles simulated with the Bogren model (Figure 12). The Bogren model output deviates from the measurement data such that it deviates from the VIC model output. A better fit is probably due to the fact that the VIC model takes into account the decreasing Donnan effect as hexenuronic acids are cleaved from the fibre wall, whereas in the Bogren model the Donnan effect was assumed to be constant throughout the simulation.

Reactions of cellulose

Native cellulose polymers were modelled to consist of non-reducing end group (CNR) and units in the middle of the chain (CM). The degree of polymerization of cellulose in wood was assumed to be equal to 10,000.

Cellulose degradation was modelled using the original galactoglucomannan degradation scheme:

 Reducing end groups (CR[f]) in cellulose chains have two reversible acid-base equilibrium reactions leading to formation of CR[-f] and CR[-2f]. The units in the middle of the chains (CM[f]) have a reversible acid-base equilibrium reaction leading to CM(-f).

- According to Young and Liss (1978), both CR[-f] and CR[-2f] undergo peeling reaction to form isosaccharinic acid. CR[-2f] also reacts through a route forming a stable metasaccharinic acid end unit.
- The bond next to CM(-f) cleaves according to Paananen et al. (2010), dividing one chain into two new chains.

Peeling reactions lead to consumption of alkali and cause yield losses. Cleavage of the bond next to CM[-f] leads to an intrinsic viscosity drop. The experimental data used in the parameter optimization were obtained from the KrOxy project.

The simulated intrinsic viscosity is compared to measured values in Figure 14. Intrinsic viscosity measurement was not used in the parameter optimization. Changing the assumption concerning the initial degree of polymerization of cellulose chains should improve the fit of the model to the experimental data.



Figure 13. Simulated (lines) and experimental (points) cellulose yield.



Figure 14. Simulated (lines) and experimental (points) intrinsic viscosity.

5.1.3 Alkali-catalyzed deacetylation of hemicelluloses

The modelling results for the impregnation kinetics of eucalypt wood meal have been reported in Kuitunen et al., 2013 (VIC publications / The role of Donnan effect in kraft liquor impregnation and hot water extraction).

The experiments [Inalbon et al. 2009] were conducted at low hydroxide ion concentration. The modelling work showed that due to low ionic strength in the system, ion exchange effect was significant. Addition of sodium chloride suppressed the ion exchange, increased the hydroxide ion concentration in the fibre wall liquid (actual reaction site), and thus accelerated the deacetylation rate.

In untreated wood, some uronic acids are present as lactones or esters or otherwise inaccessible and so do not contribute to ion exchange. When they encounter hydroxide ions, the esters and lactones are hydrolyzed and inaccessible acids become accessible. Thus, the negative charge in the fibre wall increases and the ion exchange effect becomes stronger.

5.1.4 Reactions of polysaccharides in acidic conditions

The acid-catalyzed hydrolysis reactions of wood polysaccharides in water are modelled in the FuBio JR2 programme. The developed models are available also for simulation of acidic/neutral pulping processes.

5.1.5 Link to the VIP chemistry library

There is a need for continuum between the chromophore chemistry modelled in alkaline pulping (VIC) and that in bleaching (VIP). In the VIC model it was assumed that catechols are formed from phenolic units through demethylation by strong nucleophiles. In the VIP model, it is assumed that catechols are converted into ortho-quinones in the presence of oxygen. Ortho-quinones are very reactive with alkali and react according to the scheme developed in the VIP project. Thus, in the VIC model, demethylation was considered to be responsible for the chromophore formation in alkaline pulping.

5.2 Experimental study on the reactions of wood hemicelluloses under alkaline conditions

The alkali-catalyzed degradation of glucomannan and xylan was separately investigated in a Master's thesis entitled "Formation of hydroxy acids in alkaline cooks" by Järvinen. The cooks were performed at two alkali levels (0.31 and 0.93 g NaOH/L) and at 100, 130 and 160°C. In each case, the chemical composition of the soluble fraction of hydroxy acids was determined in detail and the formation of acids by different reaction routes was considered. However, so far the models developed were not fully verified by the results and, therefore, further verification studies will be conducted.

5.3 Neutral sulfite treatment

Changes in the chemical composition of kraft pulps were studied in neutral sulfite treatments (as a continuation of work started within the EffTech programme). As a strong nucleophile, the sulfite demethylated and dissolved lignin and depolymerized cellulose and hemicelluloses. Hexenuronic acid was quantitatively degraded.

The observation of the cleavage of glycosidic bonds by neutral sulfite is of novel interest. It is believed that the cleavage follows the S_N^2 mechanism. Although cleavage of glycosidic bonds is unwanted in pulping processes, the reaction could potentially be utilized in making specialty pulps.

Because sulfite and other strong nucleophiles are able to demethylate lignin and thus increase its phenolic group content, a separate study was undertaken to assess the phenolic group content of lignin by Raman spectroscopy. Changing pH from neutral to alkaline causes a shift in the aromatic Raman band, and this shift can be attributed to the ionization of the phenolic hydroxyl groups. UV resonance Raman spectra of several lignin model compounds in solution and wet pulp samples were collected in neutral and strongly alkaline conditions.

Change in pH induced a recordable shift in the aromatic band position in the UVRR spectra. The shift was 25-35 cm⁻¹ with model compounds without para substituent, 8-12 cm⁻¹ with model compounds with para substituent, and ca. 2-7 cm⁻¹ with pulp samples. Increasing amount of phenolic hydroxyl groups increased the UVRR band shift in pulp samples. The shift and the amount of phenolic hydroxyl groups measured with the conventional NalO₄ method resulted in similar trends but could not be directly connected with each other. However, some specific structures, such as phenolic guaiacyl units, could be identified from the model compound spectra, and also from pulp samples.

5.4 Chip-scale mass transfer model

A wood chip was modelled to consist of two liquid phases: liquid in the lumen and liquid bound to the fibre wall. The usage of two liquid phases enables modelling of the ion exchange [Donnan effect] property of the wood fibres. When uronic acids (pKa ~ 3) or phenolic groups (pKa ~ 10) dissociate, they create a negative charge in the fibre wall causing ion exchange. Ion exchange is modelled with the Donnan theory. As a result of the ion exchange, the concentration of positively charged ions is higher in the fibre wall liquid than in the lumen liquid and the opposite is true for negatively charged ions. This has an influence on the reaction kinetics.

Macroscale mass transfer was modelled to take place only in the lumen, and microscale mass transfer to take place between the fibre wall liquid and the lumen liquid. A mass transfer model developed for electrolytes was used in both macroscale and microscale mass transfer computations. The flux of a component (i) is due to 1) diffusion (concentration gradient), 2) the potential field caused by movement of other charged components, and 3] convection:

$$N_{i} = -D_{i} \left(\frac{dc_{i}}{dr} - c_{i} z_{i} \left(\frac{\sum_{j=1}^{n} z_{j} D_{j} \frac{dc_{j}}{dr}}{\sum_{j=1}^{n} z_{j}^{2} D_{j} c_{j}} \right) \right) + c_{i} v$$

where N=flux $(mol/(m^2 s))$, c=concentration (mol/m^3) , r=coordinate (m), D=diffusion coefficient (m^2/s) , z=charge (-), v=velocity (m/s).

Chip volume is assumed to be constant throughout pulping. As lignin and carbohydrates dissolve from the fibre wall, the empty space left behind is filled with water. The water flowing from the liquid phase external to the chips causes the convection (3rd term in the overall flux).

The assumption concerning the constant chip volume was tested using the fibre saturation point (FSP, kg water/kg fibre) data reported as a function of pulping yield. Reasonable agreement was found between the porosity (volume fraction of empty space in the fibre wall filled with water) computed from the amount of removed lignin and carbohydrates and the porosity computed from the fibre saturation point data (Figure 15).

Changes in porosity influence the fibre wall mass transfer and reaction rates. The porosity $[\epsilon, -]$ together with tortuosity $[\tau, -]$ and liquid viscosity $[\mu, Paxs]$ were used for correcting the reported diffusion coefficients (D(298.15K), m²/s):

$$D(T) = \frac{\varepsilon}{\tau} \frac{\mu(298.15K)}{\mu(T)} \frac{T}{298.15K} D(298.15K)$$

In the reaction rate equations, concentrations of the fibre wall compounds were computed by relating their molar amount to the mass of water bound to the fibre wall.

The above equation also shows the effect of temperature and liquid viscosity on the value of the effective diffusion coefficient (D(T)). Raising the temperature from 25°C to 170°C increases the effective diffusion coefficient seven-fold. Several different liquid viscosity models, taking into account for instance sodium hydroxide concentration or total dissolved solid and temperature, were implemented in the VIC simulator. For the tortuosity, the value was adopted from the pulp bleaching simulations.



Figure 15. Porosity computed from FSP measurement (Scallan and Tigerström 1992) data and from yield of carbohydrates and lignin.

For most components, the diffusion coefficients are reported in the literature. Wilke-Chang correlation was used for estimating the diffusion coefficients of lignin and carbohydrate polymers. The molar volume of solute (A) at its normal boiling point temperature, needed for the Wilke-Chang correlation, was estimated with Le Bas method. A very steep drop in diffusion coefficient was observed when the degree of polymerization (DP) was increased from 1 to 10 [D=8×10-10 m²/s -> 2×10-10 m²/s], after which the diffusion coefficient approached a value of 10-10 m^2 /s when the DP was further increased. There was minimal difference in the diffusion coefficients of lignin and carbohydrate polymers having the same DP, because the molar masses of lignin and carbohydrate monomeric units are quite similar.

The concentration profiles of different components as a function time in lumen can be solved with the equation:

$$\frac{dc_{i,r}^{Lumen}}{dt} = -\frac{1}{r^{\Omega}}\frac{d}{dr}\left(r^{\Omega}N_{i,r}\right) + R_{i,r}^{Lumen}$$

where Ω =shape factor of the particle (sphere=2, cylinder=1, and slab=0), R=rates due to the reaction and mass transfer between the lumen and fibre wall liquid (mol/(m³×s)).

The evolution of component concentrations in the fibre wall liquid were modelled by the following equation:



Several mathematical methods can be used to solve the resulting set of partial differential equations (PDEs) presented above. In this project, the finite volume, finite difference, and moment methods were tested. In the moment method, the concentration profiles (c vs. r) are approximated with polynomials. The evolution of the values for polynomial coefficients is solved as a function time. In comparison to the discretization methods used for solving PDEs, the number of variables needed is much smaller. The drawback is that when very steep concentration profiles develop inside the chips, polynomials attain negative values. This causes various problems in the simulation. The number of variables needed when using finite volume and finite difference methods are similar, but finite volume conserves mass better than the finite difference method. As the result, it seems at present, that finite volume is the most suitable method for solving the chip model. A report has been written concerning the finite volume solution, and a scientific manuscript addressing the moment method and finite difference solution has been written and submitted to the Industrial & Engineering Chemistry Research journal.

The model was tested by comparing the simulation results to the experimental results obtained from kraft liquor impregnation of eucalypt wood chips (Figure 16). The numerical values for experimental concentrations (published by Inalbon et al. 2011) were kindly provided by Prof. Zanuttini.



Figure 16. Simulated [lines] and experimental [points] concentration profiles of hydroxide ion [green] and acetyl groups [red] in eucalypt wood chips after 15 min [solid lines and filled points] and 30 min [dashed lines and empty points] impregnation with kraft cooking liquor. Temperature 110°C, 0.25 mol NaOH/L, and sulfidity 25%.

5.5 Modelling of a continuous digester unit

Equations for continuous digester behaviour as a function of height have been presented in a report by Abdulwahab, 2012. In short, ordinary differential equations are needed for chip column compaction pressure and liquor phase pressure drop. The chip phase [1] compaction pressure is due to gravity acceleration, wall friction, and drag between the chip and liquor phases. For the liquor phase [2] pressure drop, the acceleration due to gravity and the drag between the chip and liquor phases are taken into account.

The novelties of the developed continuous digester model in comparison to published models are:

- Kappa number is obtained using comprehensive, elementary reaction kinetic models
- Digester diameter variation is taken into account
- Possibility to take into account density and dynamic viscosity variations caused by dissolved solids and temperature on the momentum balances

5.6 Usability of the simulator

Usability of the simulator was facilitated by two means. Firstly, the graphical user interface, developed during the VIP project, was updated with the most recent Simantics platform features, user feedback, and the new models implemented in the VIC project. Secondly, several improvements were made to the actual simulator code. These include:

• Total revision of the code. Modularity and clarity of the code was improved. Microsoft Visual Studio's performance analysis tool was used for detecting CPU time consuming parts of the code. Alternative implementations significantly reduced the CPU time consumption. • New method to solve reaction and phase equilibrium (Kuitunen and Alopaeus, Robust method for solving reaction and phase equilibrium in aqueous wood fibre systems). With this new method, the equilibrium composition of process feed flows are solved in more robust way.

6. Exploitation and impact of results

Virtual Chemical Pulping Model contributes to the EffFibre programme goals through creating better understanding of the basic phenomena involved in chemical pulping of wood. Based on this knowledge, radically new and efficient pulping processes could be developed. Several new concepts have been proposed and evaluated within the programme.

7. Publications and reports

Scientific journals:

Kuitunen, S., Vuorinen, T. and **Alopaeus, V.** The role of Donnan effect in kraft cooking liquor impregnation and hot water extraction of wood, Holzforschung, 2013, 67, 511-521.

Liu, Z., Suntio, V., Kuitunen, S. and Alopaeus, V. Modelling of mass transfer and reactions in anisotropic biomass particles with reduced computational load, Submitted to Industrial & Engineering Chemistry Research in October, 2013.

Nieminen, K., Kuitunen, S., Paananen, M. and Sixta, H. Novel Insight in Lignin Degradation during Kraft Cooking, Submitted to Industrial & Engineering Chemistry Research in September, 2013.

Warsta, E., Lähdetie, A., Jääskeläinen, A.-S. and Vuorinen, T. Effect of pH on lignin analysis by Raman spectroscopy, Holzforschung, 2012, 66, 451-457.

Warsta, E. and Vuorinen, T. Neutral sulfite treatment of lignocellulosic pulps, BioResources, 2013, 8, 3468-3478.

Conferences presentations and posters:

Jakobsson, K., Kuitunen, S. and Alopaeus, V. Implementation aspects of modelling equilibrium reactions and thermodynamic data in a flowsheet simulator, Computer Aided Chemical Engineering, 2013, 32, 127-132. Poster presentation, 23rd European Symposium on Computer Aided Process Engineering [ESCAPE23], Lappeenranta, Finland, June 9-12, 2013.

Kuitunen, S., Suntio, V. and Alopaeus, V. Modelling mass transfer in wood chips. Poster presentation, 9th European Congress of Chemical Engineering (ECCE9), Hague, Netherlands, April 21-25, 2013. **Kuitunen, S.** and **Alopaeus, V.** Robust method for solving reaction and phase equilibrium in aqueous wood fibre systems, Poster presentation, 9th European Congress of Chemical Engineering (ECCE9), Hague, Netherlands, April 21-25, 2013.

Kuitunen, S. and **Alopaeus, V.** The role of Donnan effect in kraft liquor impregnation and hot water extraction, Oral presentation, 12th European Workshop on Lignocellulosics and Pulp (EWLP), Espoo, Finland, August 27-30, 2012.

Kuzmenko, O., Kuitunen, S. and **Vuorinen, T.** The reactions of non-phenolic lignin units with alfa-carbonyl functionality with soda and kraft pulping chemicals, Poster presentation, 12th European Workshop on Lignocellulosics and Pulp [EWLP], Espoo, Finland, August 27-30, 2012.

Kuitunen, S., Pulkkinen, I. and **Alopaeus, V.** Modelling of fibre swelling, Oral presentation, 5th International Colloquium on Eucalyptus Pulp, Porto Seguro, BA Brazil, May 9-12, 2011.

Warsta, E. and **Vuorinen, T.** Sulfite treatment of lignocelluloses, Poster presentation, 7th International Conference on Renewable Resources and Biorefineries, Bruges, Belgium, June 8-10, 2011.

Reports:

Abdulwahab, M. and **Kuitunen, S.**, Compaction pressure and velocity profiles in kraft pulping digester, Report of summer trainee work, Aalto University, 2012.

Master of Science theses:

Jafri, Y. Estimation and Modelling of Black Liquor Heat Content, Master of Science thesis, Aalto University, 2013.

Järvinen, A. Hydroksihappojen muodostuminen alkalisessa keitossa (Formation of hydroxy acids in alkaline cooks), Master of Science thesis, University of Jyväskylä, 2013.

8. Literature cited

Bogren, J., Brelid, H. and **Theliander, H.** Reactions kinetics of softwood kraft delignification - general considerations and experimental data. Nordic Pulp and Paper Research Journal, 2007, 22(2), 177-183.

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FUTURE PULP MILL CONCEPTS

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ABSTRACT

The Future Pulp Mill Concepts work package served as the key industrializing link for the novel pulping concepts developed in the EffFibre programme. The work package initially comprised seven concept tasks and three special tasks aimed at improving pulp usability, functionality and mill runnability. Several complex sub-processes were investigated as part of the unit operation oriented research. A total of five mill concepts in addition to a reference mill were finally evaluated by means of the developed technoeconomic evaluation platform.

The performed concept evaluations clearly showed that modern pulp mill investments are divided fairly evenly across several departments, such that significant savings in one department have only a minimal influence on total mill investment cost. The motivation for process modification should therefore be based on the following considerations: improving resource efficiency (particularly use of wood raw material), improving the sales value of new and existing products, and achieving marginal production increases in existing production lines.

The potential for a higher than 10 percentage point yield increase was identified, thus enabling significant savings in wood consumption. Medium kappa concepts (cooking kappa 40–45) are available for retrofitting existing pulp lines, although the option of increasing production through moderate new investments offers very a short payback time compared to retrofits. The best cooking and oxygen delignification techniques require further research and demonstration before being made available to industry. The proposed novel pulping concepts can significantly improve mill economy owing to enhanced raw material utilization and decreased energy consumption. From the biorefinery point of view, the dissolved hemicellulose fraction provides an opportunity to replace oil-based platform chemicals.

The developed concept evaluation practices and tools can be utilized for many kinds of industrial studies. Retrofitting of existing lines offers the most benefit until totally new lines emerge. Since the principles of the concept evaluations have been commonly built and agreed, individual evaluations are widely comparable. The applied research on unit operations of high-kappa pulping paves the way for the piloting and industrial demonstration of more effective high-kappa pulping technologies. This is absolutely needed whenever a substantial cooking kappa increase is to be implemented at the mills.

Keywords:

modelling, pulping, high kappa, resource efficiency

1. Background

Chemical pulp production based on the alkaline kraft process has remained largely unchanged for more than two decades. Contrary to the remarkable advances in bleaching practices, which have been driven mainly by environmental considerations, the pulping process has not undergone any fundamental transformations beyond rather modest modifications of the conventional cooking process.

The world market for short-fibre pulp has become dominated by South American eucalyptus, which has seriously weakened the competitiveness of birch or other naturally growing hardwood based short-fibre pulp production. It is foreseeable that a similar trend will affect the competitive position of pulp produced using Northern softwoods. The crucial factor is the cost of softwood raw material, which is particularly high in Northern Europe. The Finnish pulp industry therefore faces a bleak future if a business-as-usual approach is followed.

Our main vision for changing this paradigm is to radically reduce the wood consumption of the pulp manufacturing process, which was the key goal of Novel Two-Stage Oxygen-Alkali process work package. This is achieved in practice by a novel alkaline delignification process, KrOxy, which is able to significantly improve the yield of hemicelluloses by increasing the selectivity of lignin removal in cooking and oxygen delignification.

Another building block in our vision to develop a Sustainable and Resource Efficient Pulp Mill is a fundamental understanding of the properties of wood and the basic phenomena of the pulping process, which is the key focus area of Virtual Chemical Pulping Model work package. Although the main vision of EffFibre is based on retaining carbohydrates in the final pulp to the maximum extent desirable and possible, Virtual Chemical Pulping (VIC) modelling also enables us to study in advance how new carbohydrate or lignin-based side streams can and should be withdrawn to serve as feedstocks for new value chains without compromising the objective of producing chemical pulp fibre. In this respect, this target has a direct link to the FuBio programme. In addition, the synergy of new products with the pulp mill heat and material balance is of great interest.

The programme results were translated into practical mill concepts and form the basis for techno-economic analysis of the concepts. In the Future Pulp Mill Concepts work package, in addition to KrOxy-based concepts, a selection of other pulp manufacturing conceptual approaches were assessed using the same and comparable tools in order to evaluate alternative cash flow generating routes based on softwood raw material.

The developed future pulp mill concepts have deep roots in research carried out during the first phase of the EffTech programme (VIP modelling – the "father" of the developed VIC model, Short Pulping – the vision for yield-saving and new tools). In addition, scientific work in other cooking-related projects has contributed to the development of a new vision for the purpose of the delignification process.

At the industrial scale, general efficiency targets consist of a somewhat abstract list of complex problems with multiple parameters. In programme an extensive concept matrix has been created to solve these complicated problems with a single methodology. The resources of a single company are typically too limited for such a large undertaking, and only relative dependencies can be determined in a single small-scale project, thus leaving multiple issues unresolved. In contrast, in a large, sufficiently resourced concept study of this kind, complicated efficiency dependencies, such as energy
vs. yield or yield vs. quality, can be solved simultaneously and using the same tools.

As regards unit processes, the requirements are somewhat different. The key problem is that when a process entity (e.g. bleaching) is installed and optimized, the inefficiency of a single subfunction can impact the entire process, and unit operation inefficiency has to be compensated with additional investment. At the mill scale, typical examples are separation, filtration and mass transfer efficiency. In the worst case, an inefficient sub-function has a significant hedge function at the industrial scale. For example, insufficient mass transfer can lead to several reaction vessels instead of one, or insufficient material separation can escalate to investments in additional separation equipment.

Unit operation inefficiencies that have persisted for years can be extremely difficult to isolate and are therefore commonly treated as a generic efficiency problem. Unit operation optimization is a strategic means of reducing investment intensity. To ensure practical utilization of the programme results, efficient technological solutions for the concepts are essential.

The pulp industry generally lags far behind other chemical industry in terms of chemical, physical and mathematical modelling. Use of inaccurate modelling tools at all process levels, from overall concept to single physical or chemical phenomena, can prevent a company from investing in new technology. Fortunately, this weakness has been recognized and in recent years several dedicated projects, such as VIP, have addressed and improved the situation.

2. Objectives

The main challenges of the Future Pulp Mill Concepts work package are how to utilize existing pulp mills more profitably and how to respond to the process modification needs arising from the new value chains of cellulose, hemicelluloses and lignin-based products. The objectives are to connect fundamental knowledge with techno-economic pulp mill concept evaluation and to provide new and effective tools for short and long-term pulping process developments.

Specific objectives of the work were:

- To develop radically new energy and resource efficient production technologies to reduce the capital intensiveness of the cluster. By utilizing or modifying existing capital assets as envisioned in the present approach, these assets will be fully utilized until the end of their life cycle. It is also foreseeable that a significant simplification in, e.g., recovery island and fibre line processes will allow more compact pulp mills with a lower investment burden.
- To increase the use of domestic wood, specifically northern softwood. The foreseeable lower unit consumption of wood raw material for pulp production may alleviate possible competition against sawn timber or fuel production in the event of limited wood supply.
- To support sustainability: More products [pulp, energy and bio-products] produced from less wood further improves the sustainability of this renewable resource based industry.
- To create systematic modelling practices for chemical pulping as a basis for investment planning and decision making.

3. Partners and their contributions

The partners and their roles in the future pulp mill concepts research area of the EffFibre programme are presented in Table 1.

4. Research approach

Many new development programmes focus on testing and optimizing specific parts of a process or generating a new process without taking the mill environment into account. As a consequence, many new and promising process applications cannot be utilized at the mill scale because no investigation of how the new process can be installed in the existing mill infrastructure is carried out. In addition, there is often a lack of research and support teams to pursue mill-scale development. In the work package, the approach was to integrate partial optimization work with mill-scale development as early as possible and to generate single process stage influence to mill balance.

When a new process application is developed, the new development step typically affects one single mill department without the overall mill balance being considered. In modern pulp mills, the design parameters of the differ-

Table 1. Partner organisations and their research roles.

Organization	Role
VTT Technical Research	Evaluation platform
Centre of Finland	Literature survey of concepts
	Experimental work of concepts
Andritz Oy	Evaluation platform
	Evaluation base data and parameters
	Pre-hydrolysis concepts
	High-kappa pulp operation parameters
	Frozen chip smelting model
Kemira Oy	Control of hydrophobic substances in water circuits
Metso Oy	Techno-economic evaluations
Metsä Fibre Oy	WinGems basic pulp mill model
	New reinforcement pulp manufacturing concept
	Highly thermally stable cellulose pulps
UPM-Kymmene Oy	Sulfur-lean concept
	Lignin separation cooking concept
Stora Enso	Chip quality measurement and process control

ent departments and the overall energy balance of the mill are highly optimized, so any additional load from one department can have a significant impact on all other process sections. It is therefore highly beneficial to test the new process or application's effect on the mill balance as early as possible in order to understand its real process potential.

One approach was to determine the Capex between the main process departments, as the final operating cost is a function of operating and capital expenses. For the Capex analysis, the cost structure of the reference mill model was calculated by Pöyry. Each concept's Capex was calculated based on the cost of a single department in the reference mill, and if the investment of department was for different capacity, the new cost was calculated according to evaluation platform formula 1:

The calculation was made only for process departments, with sub-functions inside the department left unchanged.

The following concept development stages were applied to each concept:

• Literature survey of current knowledge and practices regarding the concept.

• Laboratory testing to define the main operating parameters for the process and concept. The amount of data generated in lab testing was less than a normal development process because the balance calculation partially supported the results.

• Use BAT and known main design parameters as reference for main mill departments where the WP does not develop new technology. Apply this data also to the reference pulp mill model and to the new concepts.

• Generate the main balance for the known reference mill.

- Run the mill balance for each sequence.
- Calculate Opex and Capex for all sequences.

In the work package the studied concepts were in fiberline applications and partially new. The chemical recovery departments were for alkaline pulping and generated according to general and published data on typical parameters and solutions for a modern Scandinavian mill. In this project the main objective was to define the operational parameters for the new pulping concept, and then to simulate the process in a modern pulp mill environment to determine the feasibility of each process.

As a uniform model universally applicable to all modern mills, the generated model is expected to serve the industry for a number of years. The model enables companies to test new applications and processes in the mill environment and to utilize the results in strategic decision making in the early phases of process development.

An evaluation platform was generated for a reference mill and the following concepts were evaluated:

- · Reference mill concept
- Sulfur-lean concept
- EneMi, energy mill concept
- Urmas concept
- KrOxy concept
- Pre-hydrolysis concept

Two concepts were not evaluated: the neutral sulfite concept gave fairly good pulp results, but the evaluation platform was not suitable for sulfite mill evaluation; in the lignin separation concept, lignin mass transfer did not succeed after dissolution, rendering the concept unfeasible.

5. Results

5.1 Evaluation platform

The aim of the evaluation platform task was to create a framework and tools for the evaluation of pulping and biorefinery process concepts.

At the beginning of the programme, reference kraft mill model concepts were developed for softwood and hardwood. The reference pulp mill model was created based on Metsä Fibre's WinGems fibre line model and modified Andritz' recovery model. Unit operations were updated or rebuilt to represent the latest design and BAT requirements for pulp mills. The material and energy balances of the studied process concepts were calculated using a WinGems-based simulation model, as described in detail by Kangas and Kaijaluoto (2013). A toplevel view of the simulation model is shown in Figure 1. The model is composed of the following departments:

- Debarking (highly simplified)
- Cooking and oxygen delignification
- Bleaching
- Drying
- Wastewater treatment (highly simplified)
- Evaporation
- Recovery boiler and bark gasification (gasification highly simplified)
- White liquor preparation
- Turbine plant

Techno-economic analysis was based on variable production costs (wood, chemicals, energy, waste). In order to further define the analysis, the investment costs of the reference pulp mill were included. The investment costs of a modern Nordic kraft pulp mill (2000 adt/day) were estimated by Pöyry (2012). The estimate



Figure 1. Top-level view of the simulation model.

was made for a 640,000 adt/a mill. Costs were divided into 18 different mill departments from wood handling to pulp drying and from evaporation to recovery boiler. The design capacity was defined for each department. The mill site infrastructure was also included. The costs of each department were further divided into 8 sub-categories, such as civil works, main machinery or automation. The overall cost of a new pulp mill in Finland was estimated at EUR 760 million excluding in-direct costs or contingency. This cost data was further utilized when calculating the investment cost comparison of the different concepts using formula 1:

$$\frac{C_a}{C_b} = \left(\frac{A_a}{A_b}\right)^n$$

where C_a and C_b = costs, A_a and A_b = design capacities, and n = 0.6 [typical value].

When estimating the total production cost of adt pulp, the following cost fractions were taken into account: wood, chemicals (includes utilities and waste), labour, maintenance (includes spare parts), depreciation, other costs (such as insurances, local taxes and administration) and logistics (transportation from Nordic countries to Central Europe). This was a modification of the cost structure presented by Turton et al. (2010), and very similar to that used by Pöyry (2012).

In addition to building the concept evaluation platform, a virtual pulp bleaching model (VIP) was implemented on the Simantics graphical simulation platform (Figure 2).

5.2 Unit operations

Unit operations form the core of mill operations and generate the sub-processes that make up the mill. Thorough understanding of unit processes is essential to achieving the resource and energy efficiency targets of the future pulp mill.

As part of the work package, Andritz conducted a unit operation project focussing on three functions: chip defrosting, filtration and wash-



Figure 2. Implementation of the VIP bleaching model in the Simantics simulation platform.

ing. Heat transfer phenomena and defrosting of frozen wood chips is an important unit operation in pulp mills located in arctic conditions. The results expand our understanding of heat movement inside the chip column during the pre steaming.

The motivation for the filtration study was to further examine lime mud and fibre suspension behaviour during filtration with the aim of increasing the capacity of the filters and washers. The two- and three-phase models were constructed for the filtration. The fibre content in the permeate was also investigated.

The first subtask of the washing study was to develop a method for estimating washing efficiency along the cake during counter-current cake washing. This made it possible to determine at which point along the cake optimal washing efficiency is achieved. In the later part of the washing task data reconciliation methods were developed for estimating washing efficiency by means of online measurement of dissolved dry solids content around the washer. This aids optimization of washer and washing plant operations. As a side result of the data reconciliation methods, two new equivalent washing efficiency parameters were developed.

Heat transfer and defrosting phenomena of frozen wood chips

This subtask examined the defrosting of frozen wood chips and steam movement inside the frozen chip column. An insulated cylinder measuring approximately one metre tall and 0.3 m in diameter was built to study temperature development inside the frozen chip column. Thermocouples were installed inside the wood chips and also in different parts of the test environment. Figure 3 schematically illustrates the test environment and thermocouple positions.



Thermocouples

- Three thermocouples 3,7, and 11 are placed inside wood chips.
- Thermocouples 2,6 and 10 monitor chip surface temperature.
- 15 and 16 measure temperature at both ends of the bed.
- Rest are placed on opposite sides of bed and 13 is positioned top middle, the remainder on opposite sides of the bed at top part.

Figure 3. The schematic view of the test environment and positioning of the thermocouples.

Figure 4 shows the temperature development inside individual wood chips of different thicknesses. The temperature rose to steady state within three to four minutes. Figure 5 shows an example temperature development at different heights in the chip column: the first layer was about 0.25 m, the second 0.50 m and the third 0.75 m from the steam injection point (bottom), respectively. It took about 15 minutes to heat the whole column to steady-state temperature.

Filtration

Filtration is a common unit operation in pulp mills and is used for the separation of solids

from liquid by interposing a medium through which only the liquid (permeate) can pass. Filtration takes place in digesters, pulp washers, drying machines, lime mud filters, white liquid filters, dregs filters, etc. One of the key questions of filtration studies is filter capacity, i.e. how much solids and/or permeate can be handled in unit time. A key motivation is try to minimize the required surface area, i.e. filter size, with respect to production requirements.

We created a mathematical model for lime mud filtration that accounts for the special filter precoat function of lime mud. However, the mod-



Figure 4. Temperature development in the middle of a wood chip.





el is not restricted only to lime mud filtration and can be used also for other porous material when the required parameters are known. The lime mud model consists of three phases: solids [lime], liquid (white liquor) and gas. The model is based on an immiscible flow of gas and liquid inside the porous material, described by Darcy's law. The liquid is incompressible, the gas compressible. The model gives the dry solids content and the liquid and gas velocities as a function of time in different parts of the cake and the pre-coat. As an example, Figure 6 shows the average dry solids content of a "typical" lime mud cake as the cake is cyclically formed above the pre-coat and later dewatered.

Similarly, Figure 7 shows the instantaneous volumetric fluxes of gas and liquid during the filtration cycle.

The modified filtration model was also applied to the cake forming analysis of the pulp suspension. Visual markers were placed inside the filtration chamber and monitored by camera. Figure 8



Figure 6. Average dry solids content of lime mud cake.



Figure 7. Instantaneous volumetric fluxes of liquid and gas in the lime mud cake.

shows the movement of the markers during cake formation at 3% initial pulp consistency.

The second task of the filtration study was to determine which operation parameters affect the fibre content of the leaving filtrate (permeate) during filtration. The fibre content of filtrate flows not returning to the fibre line is an important parameter as a key measure of material loss. A laboratory test environment was built to study the filtration process and the fibre content in the filtrate. As Figure 9 shows, the most important parameters affecting the fibre content of softwood pulp are feed consistency and pressure shock against the filter medium.

Washing studies

The washing studies comprised three tasks. In the first task, a counter-current cake washing model was developed in which the washing stages are divided into sectors and each sector has own dimensionless Peclet number for characterizing washing efficiency. The aim was to determine the washing efficiency of the dif-



Figure 8. Marker movement during cake formation at initial 3% consistency pulp suspension in the chamber.



Figure 9. Fibre content of the filtrate at different feed consistencies and pressure shock against the filter medium. ferent sectors. As an example, the model was applied to 7-stage [8 sectors] counter-current cake washing, see Figure 10. The results in Table 2 show the Peclet number for each stage. The developed method makes it easier to estimate washing efficiency along the cake. To enable washing efficiency to be improved, it is beneficial to know where the washing efficiency is low and high. The model was also applied to segregated filtrate circulation in cake washing.

The aim of the second task was to develop two equivalent washing efficiency parameters: equivalent wash yield Y_{st} and general equivalent displacement ratio $DR_{cost,ctst}$. The equivalent washing efficiency parameters make it possible to compare different washers easily. The equivalent wash yield is based on the standardized feed consistency to the washer and is an intuitively clear parameter. The general equivalent displacement ratio is an extension of the EDR value and enables feed and discharge consistencies to the hypothetical washer to be freely selected.

In the third subtask, a method was developed to estimate washing efficiency based on online-measured dissolved dry solids data from around the washer, and data reconciliation. As an example, the estimated standardized wash yield of a washer using different data reconciliation methods is shown in Figure 11.



Figure 10. Dissolved dry solids content in 7-stage counter-current cake washing.

 Table 2. Estimated Peclet numbers (P) and calculated dissolved dry solids content of the inflows and outflows of different stages in the belt washer.

Stage, i	P	$C_{c(i-1)}$ (%)	$c_{c(i)}$ (%)	$c_{w(i)}$ (%)	$c_{f(i)}$ (%)
1	1,39999	12.49239	9.33223	8.63000	11.61618
2	3.09593	9.33223	5.52248	5.03000	8.63000
3	1.66686	5.52248	3.38479	3.01000	5.03000
4	1.44184	3.38479	1.89264	1.60000	3.01000
5	0.74582	1.89264	1.00370	0.76000	1.60000
6	0.00434	1.00370	0.58627	0.36556	0.76000
7	0.00000	0.58627	0.21000	0.01000	0.36556



Figure 11. Online equivalent wash yield Y_{10} based on the different data reconciliation methods.

Processability of high-kappa pulps

The target of the KrOxy concept study was to determine the processability of kappa 50-90 pulp after the blow line and before oxygen delignification. The findings are useful in all high-kappa pulping applications. In this section of the pulping process, the pulp is passed through a mild mechanical refiner to reduce its shive content. This subjects the pulp to mild mechanical defibration, which can have an impact on pulp properties and cause pulp deformation. In the present study, unwashed industrial kappa 70 pulp was run through a 12" laboratory disk refiner. The refiner was equipped with mild effect plates and two 2 mm and 5 mm plate distances were used. The pulp was passed six times through the refiner and a pulp sample was taken after each pass. Drainage, SR°, shive content, curl, kink and fibre length were measured. Drainage was measured by a DD tester similar in function to the Rauma-Repola tester and Perti wash tester used in the thick cake filtering tests.

The results (Figures 12 and 13) show that, with the marked exception of a dramatic change in drainage, pulp properties are not remarkably influenced by refining according to conventional pulp analyses. A surprising finding was that the pulp shive content did not reduce below 2% after 6 passes. The results show that mechanical defibration causes a remarkable reduction in drainage and this has a negative impact on equipment size.

In the final oxygen stage, the pulp reached a kappa number of below 20 and final brightness of 89 in ECF bleaching. No significant differences in physical properties were distinguished between the defibrated and reference pulp.

Unit process research is one of the core pillars of wood-based process industry renewal, and there is a need for continuous unit operation research in all key areas of the pulp industry. Development of unit processes is key to securing the future competitiveness of pulp, biorefinery and paper production.

5.3 Medium kappa KrOxy concepts

The basic idea of the medium kappa level KrOxy concepts was to evaluate possibilities to increase pulp yield by performing high-al-



Figure 12. Drainage and SR° as a function of refiner passes for kappa ~70 pulp.



Figure 13. Shive content as a function of refiner passes for kappa ~70 pulp measured by a Somerville screen with 0.15 mm slots.

kali kraft cooking with or without cooking additives at kappa number 45 and continuing delignification with conventional oxygen delignification. The main expected economic effects were reduced wood consumption per produced tonne of pulp and/or increased pulp output and significantly enhanced reinforcing properties of the fibres for product added value. In addition, performing the proposed process modifications of the kappa 45 concept would make it possible to utilize the existing pulp mill capacity to the end of its natural life cycle. In the course of the project it was decided to combine the medium-kappa KrOxy concept with a new reinforcement pulp manufacturing concept, resulting in the joint Urmas concept reported below.

5.4 New reinforcement pulp manufacturing concept

The objective of Urmas was to generate an industrial concept for reinforcement pulp manufacture based on the yield-saving pulping modifications developed in WP4 [KrOxy]. The new reinforcement pulp was dedicated to wood-containing printing papers. The main hypothesis regarding optimal reinforcement pulp quality was to preserve and optimize the distribution of the hemicelluloses fraction between the cell wall and fibre surface by means of a combination of the yield-saving delignification process and targeted additions of hemicelluloses in the process.

The project comprised the following tasks:

- Define the critical product quality and performance criteria for optimal reinforcement pulp.
- Increase the hemicellulose yield of spruce pulp utilizing conventional and SuperBatch cooking with additives (AQ and PS) at kappa level 30 and 45 and KrOxy ideology-based cooking modifications at kappa level 45 and 60 including oxygen delignification and bleaching trials.
- Test the reinforcement ability of bleached pulps with wet strength and runnability testing.
- Define a cost-efficient manufacturing concept (particularly wood consumption vs. energy production).

The laboratory cooks, except the SuperBatch variants, were carried out at VTT in a 30-litre water-jacketed circulation digester (Verdi). The SuperBatch and SuperBatch PS-AQ cooks were conducted at Experimentis Oy, Rauma. Oxygen delignification was performed in two modes: conventional one or two-stage oxygen delignification according to the cooking kappa; novel continuous oxygen delignification system adapted and up-scaled from the high-kappa KrOxy concept. The bleaching sequences for these pulps were DEopDP according to the cooking kappa. The target brightness was 90% ISO.

Selected bleached pulps were refined after laboratory drying in a Voith LR 1 refiner with conical fillings and tested for basic pulp and sheet properties at Labtium Oy. Impact testing was also carried out for the selected pulps in order to evaluate the wet strength properties of pure reinforcement pulp. Based on the yield and chemical composition results of the bleached pulps and the results of Impact testing, three pulps were selected and evaluated at VTT Jyväskylä for wet strength and runnability (Impact and Kisu).

Pulping studies for increased hemicellulose yield

The highest total yield increase, mainly in the form of GGM, was achieved in conventional and SuperBatch cooking with the use of PS-AQ. At kappa number 30 the yield increase was about 2.5 percentage points and at kappa number 45 about 6% compared to the reference kraft at kappa number 30 (Figure 14). With SuperBatch cooking, the yield gain was slightly lower than with the conventional kraft.







Figure 15. Cooking yield vs. kappa number (High alkali-type modifications).

The use of polysulfide in high-alkali cooking offers an interesting possibility to increase cooking yield. A yield increase of 3 percentage points over high-alkali cooking was obtained at kappa level 60. The highest yield, almost 60%, was got with low temperature additive cooking (Figure 15).

The yield benefit was well preserved up to the bleached pulp. Figure 16 shows that the yield benefit comprises mostly of the higher glucomannan (GGM) content of the Urmas pulps. Particularly, when polysulfide is used, the GGM retention can be maximized. The highest bleached yield and the highest GGM percentage were obtained by high-alkali cooking with additive to kappa level 45 at low temperature (EA 80 ADD π 46 LT).

Analysis of the results of Voith refining and testing of the bleached pulps (Figures 17 & 18) reveals some interesting observations. By far the best beatability and runnability performance seemed to be obtained by medium-kappa PS-AQ cooking. However, high-alkali cooking with additive at low temperature [EA 80 ADD π 46 LT] gave nearly the same performance – and without AQ.

As the dewatering properties, freeness (CSF) and water retention (WRV), should logically follow the hemicellulose content of the fibres, it



Tensile stiffn. Ind, kNm/g

8

7

6

5

4

3

200 300 400 500 600







Figure 18. Tensile stiffness vs. CSF (runnability).

CSF, ml

700 800

REFRO

O PS-AQ K43

EASO x60 Cont

A EASO PS x60 Cont

△ EA80 PS k48 Conv

EA80 PS #46 130C Con

A EASO PS x60 130C Cont

could be anticipated that a higher yield would have a negative effect on water removal. However, the clearly faster refining response was shown to compensate to a large extent, e.g., the influence of higher WRV (Figures 19 & 20).

Reinforcement fibre (RIF) performance

The objectives of the "Runnability effects of reinforcement fibre" literature review were to collect available knowledge on the runnability of reinforcement fibre (RIF) and identify the relevant RIF properties and their measurement methods. Runnability was examined in its broadest sense in regard to web runnability, excluding process parameters. As a furnish component, RIF influences dewatering during forming, the press section and drying, as well as the wet and dry strength properties of the web. According to a follow-up study, web breaks typically occur at the press section and at the start of the drying section of the paper machine. Despite this, wet strength properties are often excluded from runnability analyses, which focus mostly on dry paper strength and defect resistance. Based on the literature review, the following conclusion regarding the characterization and testing of reinforcement pulp was drawn: paper machine runnability, including the wet strength of paper, is the key concern regarding the performance of reinforcement fibres. Basic wet strength testing with the Urmas pulps was therefore performed using the Impact device, and runnability effects were tested on the semi-pilot KISU system with a typical paper machine furnish mix [kraft pulp refined to tensile index 60 or 90, fibre composition: 15-17% kraft and the rest TMP from paper mill, filler content 10%].

The pulps having the best combinations of high pulp yield and GGM content and good performance in impact testing and standard paper technical testing were selected for the trials. The tested pulps were named REF (kappa 30), PS-AQ M (kappa 43) and HA ADD M (EA 80 ADD π 46 LT). The runnability performance of the pulps was estimated by measuring dewatering properties, wet and dry paper strength, and critical structural properties of sheets.

There were no significant differences between the stocks in regard to dewatering of the web in the forming section. In wet pressing, increased refining for the same tensile mainly decreased the solids content of the webs.

REF and HA ADD M had approximately 20% higher MD tensile strength than PS AQ. The lower density of PS AQ indicates a lower degree of bonding in the fibre network. In practice, high MD tensile strength correlates with less web breaks in unit processes where dry paper web is run, and is therefore a critical factor in regard to runnability.



Figure 19. CSF vs. SRE





Formation was at same level for all the furnish mixes, while pure TMP had slightly better formation due to the absence of longer fibres. There were no significant differences in the fibre orientations of the webs. Pulps cooked with the modified methods generated a more porous structure.

The results indicate that the best overall performance in regard to paper and printing machine runnability is produced by stocks made with REF and HA ADD M pulps. In this trial, PS AQ reinforcement fibre did not improve the critical runnability properties of the web. However, as the trial stocks were composed of 85% of mechanical pulp, it is evident that the differences in performance between the reinforcement fibres were small, and more pronounced effects would be more likely if the content was increased to actual levels [20-40%] used in LWC base papers.

Techno-economic evaluation of the Urmas concepts

Techno-economic evaluation comprised the following cases: reference kraft cooking; kappa 30 (REF30) and PS-AQ cooking; and medium-kappa cooking (PS-AQ κ 43) – i.e. Urmas cases.

The reference mill utilizes \sim 3.4 million m³ of wood to produce 640,000 of spruce pulp annually. The Urmas concept needs only 3.2 million m³ of wood to produce the same amount of

pulp [~5% less]. If fixed wood consumption is assumed for the Urmas concept, pulp production is increased to ~670,000 t annually. The following three cases were evaluated in this study: [1] spruce reference, [2] Urmas with lower wood consumption, and [3] Urmas with fixed wood consumption and thus higher pulp production. The wood balances of the reference spruce mill are shown in Figure 21 and the Urmas mill in Figure 22, respectively.

Figure 23 gives an overview of the key operational parameters of the Urmas mill.

A summary of the total manufacturing costs (OPEX & CAPEX) is presented in Figure 24.

It can be said that the Urmas concept with medium kappa seems a technically feasible and economically viable option for producing reinforcement pulp. The total production cost of pulp in a new Nordic pulp mill is ~540 €/ adt. There is no significant difference between the spruce reference mill and the Urmas concept: what is gained in lower wood consumption is spent by using more chemicals and by producing less electricity. However, the sole benefit of the Urmas concept is increased pulp production if the amount of available wood is fixed. In these cases the Urmas mill could produce pulp with slightly lower costs and increase pulp production by 5%.



Figure 21. Wood balance of the REF mill.

Figure 22. Wood balance of the Urmas mill.



Figure 23. Overview of key operational figures of the Urmas concept



Figure 24. Total manufacturing cost comparison for the Urmas cases.

The economically most attractive way to apply the Urmas concept – and any proposed "medium kappa" KrOxy case – would be to retrofit an existing SW pulping line for a PS pulping modification to a medium kappa number of ca. 45 and then continue with selective oxygen delignification to about kappa 15. No such retrofitting cases were within the concept study scope of EffFibre due to several reasons, most importantly the fact that the base case for retrofitting had to be an existing line with its specialities and peculiarities (e.g. production bottlenecks). The SWOT analysis, in which the industrially relevant aspects were considered, is shown in Table 3. It should be noted that the Urmas concept will generate a fibre product that is different from the current reinforcement pulps. Many benefits are foreseen for the paper industry, e.g. in the form of better runnability at the machine as well as calculable savings in refining energy and chemical pulp dosing. However, there is also a risk that the new fibre will be initially considered as a "new animal" and thus less attractive to papermakers.

Table 3. SWOT analysis of the Urmas concept

Strengths	Weaknesses
 High and medium kappa additive based cooking give significantly higher yield Medium kappa (κ45) economically attractive for existing pulp mills Improved paper machine performance (lower refining energy, reduced chemical pulp demand) 	 So far lacking effective and selective practices for industrial high kappa oxygen delignification
Opportunities	Threats
 Tailoring reinforcement fiber performance by hemicellulose manipulation Optimizing the charasteristics of Nordic softwood fibres New pulping machinery concepts for sale 	 New fibers slowly accepted by the paper manufacturers AQ will stay on the list of forbidden chemicals

5.5 Sulfur-lean cooking concept

Today, the kraft process remains the dominant chemical pulping process. Recently, however, the utilization of side-products of chemical pulping has (again) been proposed in order to fully utilize their biorefinery potential and to produce more products than just pulp and energy. This has renewed interest in sulfurfree pulping processes, as the side-products produced are virtually sulfur-free. Traditional side-products include energy, tall oil, lignin and turpentine. A sulfur-free process could enable easier black liquor gasification with significantly increased energy production, as sulfur-free liquor is potentially easier to gasify. Sulfur-free lignin may have significant utilization possibilities in both energy production as well as as a precursor for carbon fibre production. Kraft lignin is already used in certain applications, but new applications may require the removal of incorporated sulfur.

In the current research programme, soda cooking was studied and compared with kraft cooking. The soda process, with its straightforward recovery process, is currently the most realistic alternative to kraft. Addition of anthraquinone (AQ) significantly speeds up cooking and increases the cooking yield. As some sulfur enters the process together with the wood raw material a totally sulfur-free process is not a reality with today's closed recovery systems. By means of process simulation, it has been estimated that sulfur entering the process from wood raw material would generate a sulfidity of about 8-10%. However, with methods such as using "white liquor" as alkali in bleaching and replacing MgSO, with Mg[OH]₂, sulfidity as low as 1-2%, i.e. virtually sulfur-free, can be achieved. In this study, soda, soda-AQ and soda-AQ with 10% sulfidity were compared with kraft cooking.

The cooking results were expected. Although AQ speeds up delignification, soda-AQ cooking is still slower than kraft cooking. The yields [after bleaching] were, however, comparable, even somewhat high for sulfur-lean cooking. The yield from pure soda cooking without any additives was significantly lower, by >3 percentage points. As can be seen from figures 25 and 26, there is a distinct difference in the carbohydrate composition between soda-AQ and kraft



Cook 57 O, delign. 56 55 Bleached 54 Xylan vield 53 % 52 51 50 Kraft 10% sulph 49 Soda-AQ 48 49.0 50,0 52,0 53,0 54.0 55.0 51,0 Yield on wood, %

58

Figure 25. Cellulose yield after cooking, oxygen delignification and final pulp.

Figure 26. Xylan yield after cooking, oxygen delignification and final pulp.

cooking. Soda-AQ cooking resulted in pulp with higher xylan content, probably due to the stabilizing effect of AQ on xylan. The cellulose yield is higher for kraft cooking. This is likely due to the shorter cooking time, which also gives kraft pulp the highest viscosity of all pulps. Regarding bleachability (O-A/DEopDP sequence), it was observed that the soda-AQ pulp was more difficult to bleach than the kraft pulp, requiring more chlorine dioxide and peroxide in order to reach the target brightness of 88% ISO.

Refining and testing of the pulps showed some interesting differences between them (both never-dried and dried pulps were tested). The pure soda pulp had poor beatability, while the other pulps developed tensile strength equally. Despite the fact that the sulfur-lean pulps had higher xylan content than the kraft reference pulp, they dewatered more easily than the kraft reference measured as WRV at a given tensile index. This is surprising, as xylan is known to bind water. It may be speculated that at least part of the xylan in the sulfur-lean pulps is surface xylan, whereas the kraft pulp xylan may consist more of fibre wall xylan. Regarding other strength properties, the kraft reference performed slightly better. Even low sulfidity seems to increase individual fibre strength, measured as wet zero-span tensile strength. This was also illustrated when comparing the fracture toughness index at a given tensile index. The trend is similar for tear strength, although this is not so clear between the different cases. The kraft reference also performed best on Scott bond strength. This may partly be attributed to a higher WRV value. Sulfur-lean pulps had higher bulk, stiffness and light scattering.

The cooking performance and paper technical properties of kraft and soda-AQ are fairly close, although the higher bleaching chemical demand increases the production cost of soda-AQ. Simulations show that that the productions costs would be quite similar between kraft and soda-AQ, with a difference of about 16 €/adt of birch pulp in favour of the kraft process. However, if black liquor gasification is used, soda-AQ generates more energy than kraft and production costs are lower. Sulfur-free lignin would offer a potential revenue stream but at the cost of reduced energy production.

A major draw-back of the soda-AQ process is higher investment cost. All alkali has to be regenerated in the lime cycle, and thus the lime cycle must be about 30% larger for soda-AQ compared to kraft. A larger lime kiln naturally also consumes more energy. Black liquor gasification is also estimated to be a bigger investment than a traditional recovery boiler, as the technology is still not mature.

A summary of SWOT results for the sulfur-lean concept is given in Table 4.

Table 4. SWOT analysis of sulfur-lean cooking.

STRENGTHS	WEAKNESSES
 Possibility to isolate sulfur-free lignin & side-products Less S emissions Better dewatering in press section compared to kraft pulp Higher bulk and stiffness compared to kraft pulp (never-dried pulps) 	 Higher causticizing demand compared to the kraft process Poor bleachability of pulp Lower strength compared to kraft pulp (never-dried pulps)
OPPORTUNITIES	THREATS
 Significantly increased energy production by BL gasification 	Regulation and cost of AQLow demand for sulfur-free side-

5.6 Lignin separation cooking concept

The objective of this task was to determine whether separation of dissolved lignin from pulping liquors during cooking could improve the efficiency of the cooking stage. In the case of sulfur-free cooking, isolation of sulfur-free lignin could generate a sellable side-product.

A literature review was carried out to evaluate current knowledge on lignin dissolution and separation. The amount of lignin liberated during kraft cooking is fairly well documented. However, the characteristics of black liquor lignin are much less known. Generally, it is known that as cooking proceeds, large lignin molecules can be extracted. However, the true molecular size distribution of the lignin dissolved at various phases of the cook and the stability of the dissolved fractions is not well documented, nor are the possibilities for separation of different lignin fractions.

The experimental part of the study shed some new light on lignin behaviour during cooking, but there were doubts regarding the suitability and reliability of the available analytical tools. The fact that it has been claimed and shown that some dissolved lignin fractions present in the cooking liquor have a positive effect on the speed of delignification makes the entire "lignin separation cooking" concept somewhat twosided. As one of the main ideas was to generate a new valuable side-stream in chemical pulping, it seemed risky to proceed in the chosen direction when some negative side-effects of delignification may occur, especially when the characteristics of this specific lignin (fairly low molecular mass) make it challenging to separate economically. In this sense, it may be more attractive to separate lignin from the liquor going to the chemical recovery island (e.g. the Lignoboost process). The study was thus terminated after finalizing the initial experimental part.

5.7 Energy mill concepts

In the energy mill scenario, the energy production of the pulp mill is given a greater importance compared to traditional production. As the price of energy continues to outpace the price of pulp, there is growing interest in lowkappa (<10) pulping. Low-kappa pulping using yield-preserving methods is preferable because the heating value of carbohydrates [cellulose and hemicelluloses] is lower compared to lignin. In the present study, AQ and polysulfide were utilized as cooking aids in both conventional and SuperBatch kraft cooking. Cooking to a low kappa number enables the oxygen stage, and possibly also a chlorine dioxide stage, to be omitted, resulting in savings in investment and energy costs. Both the ECF and TCF bleaching options were evaluated.

According to the experimental results, cooking to kappa number 10 with over 40% yield is achievable by adding PS-AQ to the cook (Figure 27). PS-AQ effectively increased the stability of glucomannan also when cooking to low kap-



Figure 27. Development of pulp yield in cooking, oxygen delignification and bleaching as a function of kappa number for selected EneMi concepts.

pa numbers and throughout both ECF and TCF bleaching. The glucomannan to xylan ratio was thus different, 7.5 : 9.5 vs. 11.5 : 6.5, for the reference kraft pulp and EneMi PS-AQ pulps.

The pulp produced in the energy mill ECF (with PS-AQ) concept was of reasonable quality. Moreover, some properties were equally good or even better than the properties of the reference pulp. For example, the tensile properties of the EneMi PS-AQ ECF pulps were better than those of the reference kraft (Figure 28), although the advantage over the reference was lost in TCF bleaching.

More wood was consumed by the EneMi concepts, and costs were 17 €/adt higher. The chemical costs of the EneMi TCF case were ~85 €/adt higher than all ECF cases due to the use of expensive peracetic acid and chelate as well as increased consumption of other bleaching chemicals, such as NaOH. The variable production cost per adt of pulp (when revenue from by-products is included) was somewhat higher for the EneMi ECF concept, at 267 €/adt, compared with the reference mill using kraft cooking, 257 €/adt. Variable production costs of EneMi TCF were much higher, at 350 €/adt. The increased electricity production of the EneMi concepts did not compensate the losses due to higher wood and



Figure 28. Tensile stiffness index as a function of specific refining energy. Voith LR1 with conical fillings, never-dried pulps.

chemical costs. If the electricity price is set to the same level as electricity produced by wind, the energy mill ECF concept is economically very close to the reference kraft mill.

The investment cost of the EneMi ECF mill was slightly lower than the reference mill, at ~EUR 9 million, whereas the EneMi TCF mill cost was equivalent to the reference mill. The omission of oxygen delignification reduced the investment cost, but this was offset by the larger recovery boiler. The total production cost of the reference mill was 536 €/adt compared to 543 €/adt for the EneMi (ECF) mill (Figure 29). If the December 2012 price of 610 €/adt for softwood pulp is assumed, the revenue was positive for the reference and EneMi (ECF) concepts. If CHP (combined heat and power production) can be utilized, the economy of both concepts could be improved during winter months. This is due both to improved mill energy efficiency and the favourable price of district heating. However, it is important to note that district heating is a seasonal product and cannot be sold year-round. The SWOT analysis of the energy mill concept is presented in Table 5.

5.8 High-yield nucleophile cooking

The objective of this concept was to study the effect of sulfite cooking at pH 6-8 on total pulp yield. The idea is that in neutral cooking conditions strong nucleophiles such as sulfite ion are able to dissolve lignin while the carbohydrates are preserved to a high extent. In neutral conditions, the acid-base-catalyzed degradation of carbohydrates is significantly slowed, leading to high pulping yield.

The cooking experiments produced high-kappa pulps with high yield. The target kappa number of 50 was reached by cooking pine chips



Figure 29. Total production costs/ adt pulp of EneMi concepts.

 Table 5. SWOT analysis of the energy mill [EneMi] concept.

Strengths	Weaknesses
 Slightly lower investment costs (no oxygen) More simple process Low extractive content 	 High wood consumption Wood consumption reduction depends partly on AQ charge No reduced chlorine consumption Large digester needed Larger recovery boiler needed Heat usage in summer (if CHP) Must be situated near heat users
Opportunities	Threats
 Increasing energy prices + "Green energy" benefits and economy Easy TCF bleaching 	 Economics Green energy benefits do not include pulp mill energy production Pulp properties do not meet requirements

at 180 °C for 240 minutes using 30% Na_2SO_3 charge and 0.1% AQ-charge, and initial pH 8. Lower kappa numbers were obtained by using a higher chemical charge, higher temperature or longer cooking time. The advantages of these pulps are a high cooking yield of 55% at kappa number 50 (Figure 30) as well as high brightness at over 60% ISO. However, pulp viscosity was decreased considerably during cooking.

The pulp cooked to kappa number 59 was del-



Figure 30. Comparison of total yield (% od wood), in neutral sulfite cooking [NS] vs. reference kraft cooking.

ignified further with one and multiple oxygen delignification stages. This decreased the brightness of the pulp to below 40% ISO, but the yield and viscosity were maintained. The bleaching sequences tested, DEDED, DEpDP, and DED, resulted in pulps with brightness of 89% and 87% ISO. The disadvantage of the bleaching was high chemical consumption, although the pulp properties were maintained. The total yields of the bleached pulps based on od wood were between 48-53%.

Chemical composition analysis of the selected pulp samples confirmed that the yield increase was mainly due to high retention of glucomannans. The sulfonic acid groups content measured from the cooked pulps were 250-300 mmol/kg, indicating that the sulfonic acid groups are also attached to the carbohydrates. Near-neutral sulfite cooking seems to be a very promising alternative for producing high-yield pulp with high brightness after cooking. The high sulfonic acid group content of the pulp provides new application potential. Furthermore, the utilization of lignosulfonates from the residual cooking liquor will improve the economics of the possible neutral sulfite biorefinery concept.

5.9 H₂O₂ boosted cooking

The main object of this concept was to direct degradation product formation of glucomannans selectively towards glycolic and 3,4-dihydroxybutyric acids by alkaline hydrogen peroxide pre-treatment prior to kraft pulping. Approximately 75% of glucomannans are lost during kraft cooking, forming various degradation products. Extraction of these degradation products from black liquor is a complex process. Alkaline peeling of the glucose end group forms a 4-deoxy-2,3-hexodiulose intermediate, which is cleaved in the presence of hydrogen peroxide to glycolic and 3,4-dihydroxybutyric acids. The goal was to determine optimal conditions for pre-treatment for selective product formation and to study its effect on the cooking process.

Pre-treatments were performed with spruce chips and coarse first-stage TMP to reduce mass transfer difficulties. To minimize decomposition of hydrogen peroxide due to heavy metals, acid washing or chelation with DTPA was performed on the chips and TMP. In addition, the pre-treatments were performed in 11 autoclaves coated with zirconium. The experiments compared the effects of temperature, time, alkali and hydrogen peroxide addition on degradation product formation. The degradation products were identified by a gas chromatography-mass spectrometer and quantitative analysis was performed by a gas chromatograph with flame ionization detector. The highest formation of the desired degradation products (0.71% glycolic and 0.58% 3,4-dihydroxybutyric acid, od wood basis) from spruce chips was achieved with 18% NaOH and 7% hydrogen peroxide additions at a liquid to wood ratio of 1:6 (Figure 31). To reach a kappa number of 30, the pre-treated chips required 4% less effective alkali in the cooking stage and slightly lower H-factor when the sulfidity was 35% and temperature 165 °C.

The results demonstrated that the pre-treatment directed degradation product formation selectively towards glycolic and 3,4-dihydroxybutyric acids, while formation of other degradation products was limited. However, the product concentrations obtained were low compared to chemical consumption. Further development of the process should be focused on preventing the consumption of hydrogen peroxide by side reactions.



Figure 31. Formation of glycolic and 3,4-dihydroxybutyric acid as a function of NaOH addition at a constant 1:2 ratio of H_2O_2 and NaOH.

5.10 Pre-hydrolysis pulping concept

The pre-hydrolysis kraft pulping concept typically produces dissolving pulp for the textile market, but due to growing interest in the hydrolysate of this concept in recent years, it was of interest to determine how Scandinavian softwood can be utilized in pre-hydrolysis pulping. Specifically, due the low yield of dissolving pulp, it was of interest to determine the actual production costs when produced using modern technology. In the simplest scenario, all removed organic material, hydrolysate and black liquor are burned for energy production. In such a system electric power production is maximized and only cellulose is utilized to final product. There are various options for refining the hydrolysate further, but as the technology for treating the hydrolysate is under development, only the yield and chemical composition were analysed.

Dissolving pulp is produced using the sulfite method or the pre-hydrolysis kraft method but from different hardwood species. In this study, the pulping properties of softwood were studied to determine the pulp quality of softwood dissolving pulp. Today, softwood is widely used in paper pulp and has a good market position, but in the future, softwood could also be used in dissolving pulp manufacture. For example, logs from first thinnings could be suitable for the pre-hydrolysis kraft process.



Figure 32. Batch pre-hydrolysis yield as a function of P-factor.

The softwood chips used in the study were EffFibre reference chips, as used in reference cooks in several other work packages. The prehydrolysis, cooking, oxygen delignification and conventional bleaching were carried out at Aalto University and the ozone stages at the Kemira R&D centre in Espoo. The p-factor influence on softwood pulp was also tested to determine the influence of the different pre-hydrolysis treatments on pulp and hydrolysate properties.

The pre-hydrolysis was conducted as an autohydrolysis process, requiring only a small amount of water and a temperature of about 150-170 °C as the wood moisture and direct steam used for heating is sufficient for hydrolysis. The wood acidity reduces the pH to the level required for hemicellulose degradation to commence. Furthermore, as many hydrolysate reaction products are acidic, the pH reduces further to around 3.5, at which the process becomes auto-buffering.

Results

Batch hydrolysis testing showed the relationship between P-factor and yield (Figure 32). It can be seen that during the optimized autohydrolysis process about 15-20% of wood material is degraded before alkaline cooking is started (Figure 33).



The batch results were used as a reference

Figure 33. Sugar components in the hydrolysate.

when the cooking conditions were defined. For the cook in which pulp bleaching and quality parameters were determined, the following conditions were selected:

- + P factor 500 in pre-hydrolysis and, total 710, 160 $^{\circ}\mathrm{C}$
- Alkali charge 23,5% on wood (EA) as NaOH
- Sulfidity 37,8%
- H-factor 1640

The results show that the cooking is quite similar to a normal kraft cook.

The cooking and oxygen stage results are shown in Table 6. The cooking kappa and oxy-

gen kappa were selected according to viscosity. The pulp R10 and R18 (cellulosic residue in 10% and 18% NaOH solution) values are high, indicating that the pulp dissolving properties are at the right level. The kappa can be further decreased, but doing so results in too low viscosity. The pulp bleachability is quite similar to softwood kraft pulp (Table 7). Because the kappa is lignin kappa, the chemical consumption of bleaching depends on the kappa number. The R10 and R18 values are maintained in the reference bleaching sequence D-Eop-D-P, so it is flexible between paper pulp and viscose pulp.

Softwood is suitable for dissolving pulp manufacture at reasonable manufacturing cost. Dis-

Table 6. Main pulping parameters and results for pre-hydrolysis pulp.

Initial pulp	
Kappa Number	20,7
Viscosity, mL/g	1024
ISO Brightness, %	26,2
R10, %	93,7
R18, %	95,1

O Stage	
Temperature, °C	95
Time, min	60
Pressure, bar	5,5
NaOH, %	2
Final pH	12,1
Kappa Number	9,7
Viscosity, mL/g	734
ISO Brightness, %	37,1

Table 7. Main bleaching parameters.

	SW
Карра	9,7
CIO ₂ , kg/adt	10,7
H_2O_2 , kg/adt	3,6
NaOH, kg/adt	19,8
H ₂ SO ₄ , kg/adt	3
Viscosity, ml/g	489
R10, %	94
R18, %	96,5

solving pulp yield is significantly lower than bleached kraft pulp, which has a significant impact on investment and production costs (Figure 34). In our example, the hydrolysate is burned with the black liquor, which maximizes the mill energy production compared to pulp production. Due to increased organic load on the recovery system, the evaporation and recovery boiler must be designed for higher capacity than in the kraft case. This has a significant effect on investment calculations. If the hydrolysate is recovered and treated separately, the size of recovery can be reduced accordingly. The balance calculation clearly shows the role of the hydrolysate in a modern pre-hydrolysis mill and provides an indication of its chemical composition.

The concept study shows that Scandinavian SW is a good raw material for the pre-hydrolysis process. In addition to the textile industry as the main user of dissolving pulp, the material industry is also showing interest in dissolving pulp as a potential raw material in a number of applications, such as micro- and nanofibrillated cellulose (MFC and NFC).

To conclude, the SWOT analysis of the pre-hydrolysis concept is presented in Table 8.

Strength	Weakness
 Flexibility, possible to produce several products Highest electric power production Hydrolysate can be one product Good technology applications available globally 	 Hydrolysate is difficult to refine to further products Mill is more complex than kraft mill Raw material quality must be better than with paper pulp Recovery island must be designed for higher capacity
Opportunities	Threats
 Global textile market is about 70 Mt/a and the market is growing. Pulp can be used in several products, not only to dissolving Cotton farming is limited globally capacity needed for food production Dissolving pulp could be produced from wood fractions which has low extention 	 Rayon capacity will stay limited globally, other products do not became competitive Oil base textiles stays competitive and increases the market share

Table 8. SWOT analysis of the pre-hydrolysis concept.

	1000			
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5	800	the second se		
2	700	-		the second se
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t c	500	_		
Jen	400			
stu	300			
2	200		1000	100 C 100 C
-	100			
	100		and the second se	the second se
	Ū	Ref. DP	Ref. SW	Ref. Poyry
Mill site infrastucture [ME]	70	70	70
Effluent treatment [Mé	9	24	24	25
Fresh water treatment	[M€]	7	7	7
■ Turbine plant [M€]		72	59	47
Bark gasification [M€]		33	29	29
Lime kiln [MC]		24	19	19
■ Recausticising [M€]		39	32	32
■ Recovery boiler [M€]		173	138	140
■Evaporation [M€]		84	66	52
■ Pulp warehouse [M€]		4	4	4
■ Pulp drying [M€]		114	114	114
■ Bleaching [M€]		52	52	52
Oxygen delignification	[M€]	24	25	25
Brown stock washing 8	screening [M€]	30	30	31
■ Cooking [M€]		69	58	59
Chip screening [MC]		5	5	5
Chip storage [M€]		24	20	20
Debarking & chipping	(M€)	36	31	30
Total		885	783	760



Figure 34. Dissolving pulp production and investment costs versus the reference mill (recovery system hydrolysate excluded from investment costs).

5.11 MiHa chip quality measurements and process control

In the Continuous Digester Runnability [MiHa] project, new ways to analyse continuous digester runnability problems were studied. The project aimed to decrease continuous cooking runnability disorders attributable to raw material through:

- Identification of main runnability signals and failure mechanisms and development of robust continuous cooking process runnability indices.
- Establishing the connection between raw material (amount and properties in qualitative manner) and continuous cooking runnability.
- Creation of process control strategies utilizing the enhanced raw material information.

Early detection and identification of process faults is crucial in preventing unplanned production interruption and shutdowns in largescale processes. Corrective actions could be made if the incipient process faults leading to lost production could be identified at their early stage. When considering chemical pulping and the role of raw material in runnability failures, corrective actions could be made if information were available on raw material quality changes and variation. The effect of these changes on the cooking process also needs to be known. The information from other frontend processes is crucial in this case.

Soft sensors

The runnability of a continuous digester is often defined as 1] lack of disturbance, 2] high production rate, and 3] adequate pulp quality. Alternatively, these production factors can be described as high product quality and high process performance. However, when a continuous process shifts to a 'gray area' it can be challenging to observe the state of the process through direct measurement. In order to observe and evaluate the behaviour of the process, soft sensors were developed for continuous cooking. The soft sensors were based on process statistics, mass and energy balances, fluid mechanics, pulping kinetics and process dynamics, and were used to present refined, combined information from (direct) measurements. Soft sensor indices were formulated to detect specific disturbances in the chemical pulping line. Special attention was given to developing soft sensors to describe raw material properties and front-end process behaviour.

The processed information acquired from the soft sensors was further developed as runnability indices. The indices were scaled between -1 and 1 using soft sensor specific statistical values for mill operating data:

$$I_n = \frac{[x_n - mean(x_i)]}{2 \cdot stdev(x_i)}$$

where I = runnability index, x_n = current value of the soft sensor, and $mean(x_i)$ and $stdev(x_i)$ = average and standard deviation values of the soft sensor, respectively.

In the scaling operation, index value 0 represents the normal situation. With the described scaling operation, each runnability index was adjusted with respect to process sensitivity. The statistical values of the soft sensors were calculated using normal production periods covering several months of a pulp mill operating data. The purpose of scaling was to facilitate the use of statistical data analysis methods.

Monitoring

Soft sensors and runnability indices were developed to observe and evaluate the state of the process. From the application point of view, an important question was how to compress and present the [measurement] information. The focus was therefore shifted to monitoring, runnability failure detection and identification. The core of the monitoring system was formed from continuous measurements and modelbased indices. Based on this information, the characteristics of runnability failures were defined for systematic detection of runnability failures. Runnability failures were detected quantitatively and qualitatively. In quantitative detection, runnability failures were detected from the continuous digester operation according to production rate decreases; when the production rate decreased significantly within a given time frame, the drop was interpreted as a deterioration in process runnability. In qualitative detection, a change or increased variability in digester quality parameters (e.g. Kappa#, residual alkali) indicated runnability problems.

By combining the runnability indices and runnability failure detection, a performance monitoring system for the chemical pulping line was developed. An eight-hour time window was selected for runnability failure monitoring, consisting of six hours of data before and two hours after the triggered failure. Performance indices were visualized (Figure 35) to enable intuitive interpretation of the results. Numerical values of all sub-process indices could also be given. The performance indices and visualization of process runnability could be used for supporting operators, enhancing process operability, and fault type classification in the plant. The defined concept is not restricted only to the chemical pulping line, but could also be utilized in various other complex pulp and paper processes.

Analysis

One approach to analysing runnability failures and root causes was to use engineering skills together with the presented monitoring application. However, this approach proved to be time-consuming, as profound process knowledge was required to analyse the behaviour of the pulping line. An alternative option was to use statistical data analysis methods, i.e. clustering and classification.

Early runnability failure detection and runnability index analysis can be accomplished with data analysis methods. First, the index data was divided in two groups, i.e. digester data and front-end data. Digester data included all runnability indices from cooking, while the front end data covered all indices from the earlier process stages. Digester index data was divided into different clusters according to the numerical values of the runnability indices. Hence, each cluster had a physical interpretation based on the cluster centre point value. With the clustered fault cases, front-end process data could be classified into fault classes using classification algorithms. Digester faults leading to lost pro-

	Production	Deviation in_chip blow_pipe pressure	Deviation in_chip dry_matter content	Deviation in_wood total_yield in_impregnation	Deviation in_L/W_ratio prior_to digester	Unstable digester chip_level	Screes	Channeling	Difficulties in pulp_discharge	Blow_Kappa
Case 2011-03-04_00 04.03 00:30 04.03 10:40	Change: -17 % Max: 1230 Avgs 1080 Min: 0546 187	0.55	0.79	8.78	0.78	0.45	0.82	0.41	0.52	0.51
Case 2011-06-23_06 23.06 06:50 23.06 17:00	Changes +9 % Mex: 1280 Avg: 1170 Mm, 750 Std: 175	0.61	0.73	0,31	0.76	0.85	0.83	0.73	0.71	0.57
Case 2011-07-22_06 22.07 06:20 22.07 16:30	Change: -14 % Msx: 1110 Avg: 970 Mn: 650 Std: 147	0.63	0.48	0.55	0.48		0.43	1.49	0.48	M

Figure 35. Monitoring chemical pulping line runnability failures with runnability indices.

duction were identified and predicted based on the front-end process data.

The results could be utilized in operational practices to adapt to changes in raw material in order to increase process stability. In addition, knowledge of key raw material measures could be used in the development of relevant raw material measurement applications.

5.12 Removing hydrophobic substances from high-kappa pulps

The aim of the "Effect of cooking additives on yield and extractives" study was to test different cooking aids and their influence on cooking yield and extractive content in kraft cooks of three different raw materials. The additive dose was 2 kg/tonne of wood. The tested aids were:

- Anthraquinone
- Fennodispo 5700 (polyethoxylene ether phosphate)
- HEDPA (1-hydroxyethylidene diphosphonic acid)
- Fennodispo 5900 (alkylsulfonic acid)
- Sodium borohydride (NaBH4)
- DTPA (diethylene triamine pentaacetic acid)

In Figure 36, the cooking kappa number and yield is shown for the tested chemicals. As expected, AQ had the largest effect on yield and

kappa number for softwood, whereas HEDP had the largest effect for birch. Figure 37 shows the total amount of extractives analysed from the black liquor. As the figure shows, cooking with FD 5700 results in higher extractive content in the black liquor for both birch and eucalyptus.

AQ is usually added at the beginning of the cook. In this study, AQ was added at 4 different ways:

- at the beginning of the cook
- when the temperature reaches cooking temperature
- 50% when the temperature reaches cooking temperature and 50% at half cooking time
- divided into three points, as can be seen from Figure 38

As a comparison, the results for cooks without AQ are shown, both for cooks with and without black liquor. As can be seen, AQ addition in the early phase of the cook gave higher yield and also faster cooks than the reference. The results also showed that adding all of the AQ at the beginning of the cook gave basically the same result as cooking with black liquor.

The results showed that the influence of the time of addition of AQ on the cooking results is small. The influence of particle size offers pos-



Figure 36. Kappa number and cooking yield for different cooking additives

sibilities to tailor the influence of AQ, achieve a faster cook or increased yield.

on yield and cooking speed showed that none of the tested additives can be used to substitute AQ as a cooking additive giving both yield increase and increased cooking speed.

The results showed that the extractive content of the pulp can be lowered by using cooking additives (surfactants). The effect of these



Figure 37. Extractive content in black liquor after cooking with different additives.



Figure 38. Yield as a function of kappa number for different AQ addition points.

5.13 HitPulp – highly thermally stable cellulose pulps

Cellulosic fibres are used in a number of packaging solutions to reduce their environmental impact. Despite the wide industrial utilization of kraft pulp fibres, the effect of fibre processing (e.g. bleaching, refining and enzyme treatments) on thermal degradation of paper has still not been properly investigated. High thermal stability of pulp is essential in several packaging applications, but the potential to improve the thermal stability of fibres has not yet been realized. A solution to these challenges was sought by the Highly Thermally Stable Cellulose Pulps (HiTPulp) project, with a focus on the impact of refining, residual lignin and cellulase treatment on the thermal degradation of kraft pulp sheets. The objectives of HiTPulp were to define the mechanisms behind the thermal degradation and to develop a method to improve thermal stability (resistance to yellowing and strength loss) of kraft pulps.

A new device and described procedure was adapted for evaluating the thermal stability of

different papers at the laboratory scale. The development of optical, strength and chemical properties was monitored as a function of thermal treatment time. The experiments provided empirical data that enables the prediction of the thermal behaviour of papers in an atmospheric oven chamber at high temperature over 175 °C in the presence of water vapour. We have shown with the implemented method that there was a systematic decrease in brightness, strength and degree of polymerization (DP) of different papers with increasing temperature, time and volume fraction of water vapour. An additional significant finding of this study was that the thermal treatments performed with an oven induced similar changes in the conjugated double bond region of ultraviolet resonance Raman (UVRR) spectra to other known thermal treatment methods.

The developed thermal treatment procedure is an effective method for studying the thermal degradation of papers at high temperatures in the presence of water vapour. The major advantage of this type of device is that large sample amounts can be exposed simultaneously to high temperature.



Figure 39. UVRR spectra of refined softwood kraft paper sheets at an excitation wavelength of 244 nm from bottom to top before and after 60 minutes of thermal treatment at 225 °C: FB before thermal treatment; FB after thermal treatment at $\Phi H_2 O$ 1%; FB after thermal treatment at $\Phi H_2 O$ 75%; OD after thermal treatment at _[H_2 O] 1%; OD before thermal treatment; OD after thermal treatment at $\Phi H_2 O$ 75%. The heights of the spectra were normalized at 1097 cm⁻¹.

A second study was conducted to examine the effect of mechanical treatment on the thermal degradation of kraft fibres. Homolytic degradation of cellulose occurs when -1,4-glycosidic bonds are activated by an external mechanical force, e.g. refining. Thereby formed free radicals are highly reactive and enhance the degradation of cellulose, which results in reduced viscosity and strength properties of the pulp. On the question of the homolytic degradation of cellulose and its further reaction into carbonyl groups, this study found that the DP of the pulps decreased as a result of refining. The decrease was enhanced with enzyme treatment prior to refining.

Comparing the untreated pulp with the cellulase treated pulp, the study showed that cellulase treated fibres were more swollen (WRV), fibrillated (Simon's stain) and cut (fibre length) already before refining. The Simon's stain method with light microscopy showed that the mechanical degradation of the cellulase treated pulp proceed differently compared to the untreated pulp, as a brush-like fibre end configuration was observed with the cellulase treated pulp fibres. In addition, after intensive refining, the thermal stability of the untreated pulp sheets was superior estimated with the strength and optical properties. It is important to stress that the cellulase treatment reduced refining energy consumption significantly, but caused a significant reduction in strength properties after thermal treatment. Overall, the study implied that intensive refining of the chemical pulp introduced changes to the chemical composition of the pulp and enhanced the thermal degradation of the pulp sheets in terms of strength loss and thermal yellowing.

A third study investigated the role of lignin in the mechanical and thermal degradation of kraft pulp. We hypothesized that chemical changes during refining lead to the production of mechanoradicals, which are responsible for the DP decrease in pulp. This degradation may be inhibited by lignin due toits known ability to act as a radical scavenger.

The burst strength results of the unbleached, oxygen delignified and fully bleached pulps clearly indicated that the thermal degradation of the lignin-free pulp sheets proceeded faster (Figure 40, 41).



Figure 40. Burst strength (kPa) of unbleached (UNB), oxygen delignified (OD), fully bleached (FB), and fully bleached and refined pulp sheets after treatments at 225 °C, and Φ H₂O 1 and 75%.



TM-1000_0919 2012.09.25 14:03 D3,4 x3,0k 30 um TM-1000_0939 2012.09.25 14:47 D3,5 x3



The first study showed that water vapour enhances hydrolytic thermal degradation of fully bleached papers evaluated based on brightness, light absorption coefficient, burst strength, DP and UVRR spectra. However, lignin-containing paper behaves in a different manner. Our findings substantially add to the current understanding of how residual lignin is able to resist both thermal yellowing and strength decrease caused by increased water vapour. Additionally, the UVRR spectra and light absorption coefficient results indicate that lignin degradation occurs predominately in an oxygen-rich atmosphere. Furthermore, during the thermal treatments, the lignin content of the residual lignin-containing pulps remained constant, while carbohydrates were degraded.

The study also concluded that alkali lignin assisted laboratory refining positively influenced the properties of the bleached pulp sheets. Additionally, scanning electron microscope imaging revealed distinct deposition of lignin onto the fibre surface after sheet formation. Based on the higher burst and tensile strengths of the residual lignin treated pulp sheets compared to fully bleached pulp sheets after refining and thermal treatments, lignin was proposed to work as a scavenger against mechanically and thermally induced radicals. This study showed that lignin on the surface of the fibre matrix has a positive effect in preventing mechanical and thermal degradation.

These hypotheses have been validated and the mechanisms of thermal degradation studied using a novel approach based on new equipment. Our understanding of the chemical and physical mechanisms of refining combined with thermal treatments on pulp properties has increased. Representative results were achieved with the different refining methods at both the laboratory and industrial scale. The results are also applicable to differently treated kraft fibres and commercially available papers.
6. Exploitation and impact of results

In chemical pulping, wood raw material is the biggest single cost factor. The results of this study can be directly applied in the pulp and paper industry to achieve rapid improvements in the cost efficiency of existing mills. This strengthens the ability of producers to weather the current turmoil in the industry and to significantly extend the lifetime of existing capacity. Another main outcome of the programme is profound new understanding of fundamental pulping process principles. This enables us to simulate and foresee how processes need to be modified to meet the challenges of new value chains and products, and to adapt basic paper fibre production in line with the enduring need for reduced costs. The leading position of Finnish machine, industrial service and chemical suppliers can be maintained and strengthened by the programme outcomes.

The ultimate goals of the EffFibre programme – profitability, resource efficiency and energy efficiency – are challenging, but achievable. In this respect, the results of the present project, such as increased yield through high-kappa cooking combined with smart use of additives, are very promising. However, these goals will be more easy to attain with strategic collaboration between the EffFibre and Fubio programmes. The synergies created through this combination could open up new value chains and provide new approaches to utilizing side streams in the pulping process.

The results of the EffFibre programme will lead to constructive common agreement on the basic design data, parameters and methods to be determined for pulp mill. The strength of expertise of the EffFibre research community enables the developed platform and model to serve as a 'new industrial practice' for future mill and concept verifications. Modern decision-making processes utilize mathematical modelling and balance calculations as an integral part of risk analysis. This project combines experimental data and modelling in a synergic approach to mill concept development.

In the unit process studies, the focus has not been at the concept level, but on characterizing the complex problems that exist in the mill. Improvements in washing, separation and chip steaming require deep understanding of basic process phenomena. In addition, deeper understanding of unit operations enhances the predictability of processes and, in the long-term, reduces investment costs through better efficiency.

Anthraquinone (AQ) plays a significant role in the new vision for pulping. The key finding of the AQ study was that by determining the influence of particle size and point of addition. This also opens up possibilities to tailor the influence of AQ, for faster cook or increased yield. However, during the project AQ lost its position in BFR and, consequently, mills selling pulp for food packaging face difficulties in using AQ.

The evaluation platform and concept study was very successful to compare the new technology and generated basic dimensioning data and operation parameters for several concepts. However the next steps when the concept is developed to mill scale and industrialized, the development will require more resources and money.

The project demonstrated that with innovative approaches and new R&D methods, pulp manufacturing research is capable of bringing new ideas to the industry and significant improvements to the manufacturing process. As with all innovations, not everything is new, but the result is generated from basic knowledge of the phenomena at play and modern, innovative ways of interpreting the results bring opportunities to improve products and industry performance. The project highlights several new technologies for improving efficiency and opportunities for utilizing new processes, improving process efficiency and manufacturing products that break the boundaries of the conventional product portfolio.

7. Publications and reports

Scientific journals:

Kangas, P., Kaijaluoto, S., and Määttänen, M. Evaluation of Future Pulp Mill Concepts – Reference Model of Modern Nordic Kraft Pulp Mill Nord Pulp Pap Res J 2013. (submitted).

Tervola, P. An equivalent wash yield and extension of the equivalent displacement ratio in chemical pulp washing, Appita Journal, (submitted).

Tervola, P. An online data reconciliation and estimation of washing efficiency in chemical pulp washing, Appita Journal, (submitted)

Tervola, P. Fourier series solution for cake washing with different washing efficiencies along the cake, Appita Journal, (submitted).

Tervaskanto, M., Yli-Korpela, A., Luukkainen, S., Timonen, O. and Ikonen, E. Identification and monitoring of failure pathways in a chemical pulping line. Nordic Pulp and Paper Research Journal, 27(3), 2012.

Conference presentations:

Kangas, P., Kaijaluoto, S. and Määttänen, M. Three Concepts for Intensification of Nordic Kraft Pulp Processes (poster). AIChE 2013 Annual meeting, San Francisco, CA, US, 2013.

Kalliola, A., Kangas, P. and Kuitunen, S. Simulating A/D Stage Bleaching Chemistry. EWLP 2012 - 12th European Workshop on Lignocellulosics and Pulp, vol. 2008, Espoo, Finland, 2012.

Yli-Korpela, A., Tervaskanto, M., Luukkainen, S., Timonen, O. and Ikonen, E. Identification of Runnability Failures in Continuous Cooking. Control Systems 2012 Conference, April 22-25. New Orleans, Louisiana. Vänskä, E., Luukka, M. and Vuorinen, T. Thermal degradation of intensively refined fibers in paper. 245th ACS National Meeting and Exposition, Spring 2013, New Orleans, USA.

Vänskä, E. and Vuorinen, T. Development of thermal stability of intensively refined kraft pulps for specialty paper applications. The 17th International Symposium on Wood, Fiber and Pulping Chemistry, June 12-15, 2013, Vancouver (BC), Canada.

Vänskä, E., Vihelä, T. and Vuorinen, T. Stabilization effect of lignin in refined and thermally treated pulp sheets. Marcus Wallenberg Prize Event Special Sessions for Younger Researchers and Scientists, September 23-24, 2013, Stockholm, Sweden

Master's thesis:

Hanhikoski, S. High yield nucleophile cooking of wood chips, 28.8.2013, Aalto University, School of Chemical Technology, Degree Programme of Forest Products Technology

Purhonen, J. Hydrogen peroxide pre-treatment prior to kraft cooking, 2013, Aalto University, School of Chemical Technology, Degree Programme of Forest Products Technology.

Koistinen, A. Defrosting and heating of softwood chips with saturated steam, Oulu University, May 2012

Savela, M. Environmental impact study of softwood prehydrolysis kraft pulping and bleaching, 2013, Aalto University, School of Chemical Technology, Degree Programme of Forest Products Technology **Luukka, M.** Effect of water vapor on thermal degradation of cellulose, 14.8.2012, Aalto University, School of Chemical Technology, Degree Programme of Forest Products Technology.

Vihelä, T. Antioxidant properties of lignin in paper, 15.7.2013, Aalto University, School of Chemical Technology, Degree Programme of Forest Products Technology.



EffFibre program focuses on improving availability and supply of high-quality raw material from Finnish forests and developing new production technologies for chemical pulping. The program provides new research based solutions to improve the competitiveness and quality aspects of forest based raw materials and to develop radically new energy – and resource efficient production technologies for chemical pulping including biorefinery aspects.

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