Impregnation of Norway spruce (*Picea abies* L. Karst.) wood with hydrophobic oil

Thomas Ulvcrona
Faculty of Forest Science
Department of Silviculture
Umeå

Doctoral thesis
Swedish University of Agricultural Sciences
Umeå 2006

Acta Universitatis Agriculturae Sueciae
2006: 88

ISSN 1652-6880
ISBN 91-576-7137-0

Tryck Arkitektkopia, Umeå 2006

Abstract

Ulvcrona, T. 2006. Impregnation of Norway spruce (*Picea abies* L. Karst.) wood with hydrophobic oil. Doctor's dissertation.
ISSN 1652-6880, ISBN 91-576-7137-0.

The overall goal of this thesis was to develop new knowledge to facilitate the production of Norway spruce wood-based materials that are more durable and more homogenous than current materials by using a hydrophobic oil impregnation process. Linseed oil uptake and its dispersion were studied at both macroscopic and microscopic levels, when used to impregnate Norway spruce wood with the Linotech process. The effects of raw material properties, and treatments prior to impregnation, on retention of oil were also quantified. Furthermore, resulting crack development, modulus of elasticity and dimensional stability after impregnation was analysed. Finally, the possible use of Near-Infrared Spectroscopy measurements to estimate global oil uptake in impregnated wood was evaluated as well. There were significant differences in uptake between different wood tissues. Water-filled porosity (water volume divided by porosity volume) and basic density had significant effects on oil retention levels within tissue types. In addition, treatments prior to impregnation affected the oil retention levels. Furthermore, there were relatively good fits of the derived linear regressions predicting the retention of oil within all tissue types (0.85, 0.79, 0.66 and 0.69 in r^2 for mature sapwood, juvenile sapwood, mature heartwood and juvenile heartwood, respectively). Heartwood/mature wood and heartwood/juvenile wood showed 10-20 % weight increases due to linseed oil uptake, compared to 30-50 % in sapwood/mature wood in one experiment. One overall regression equation predicting oil uptake, based on Near Infrared Spectroscopy measurements, showed an r^2 as high as 0.89. No surface cracks due to impregnation were found in sapwood samples and only occasional cracks in heartwood samples. Internal cracks with a radial orientation occurred more frequently in heartwood samples than in sapwood samples. There was a clear negative correlation between the initial water-filled porosity and the abundance of cracks that developed. The flexural modulus was largely unaffected by impregnation; however, the variation seems to become lower for impregnated sapwood specimens than for control specimens. The effects of the impregnation on dimensional stability, tested in a water soaking experiment, were strongest in dried mature sap and heartwood tissue specimens (Anti-swelling efficiency 29.2-22 % and < 20 %, respectively). However, it also had positive effects on specimens representing other tissue and pre-impregnation treatment combinations. The treatments eliminated much of the variation in swelling and shrinking coefficients observed between untreated tissues.

The results of the presented studies imply that it is possible to fully impregnate Norway spruce wood with a hydrophobic oil in a commercial process and to produce durable wood-based materials with homogenous properties. Furthermore, it seems possible to develop evaluation methods that allow adequate setting of process parameters and rapid and accurate measurements of oil uptake during the impregnation process. Thus, wood-based materials could be created according to set specifications.

Key words: Oil uptake, cracks development, mechanical properties, dimensional stability, Near Infrared Spectroscopy, new products development, material properties, supply chain.

Author's address: Thomas Ulvcrona, Svartberget Research Station, Vindeln Experimental Forests, SE-922 91 Vindeln, SWEDEN.

Contents

Appendix

Papers I-V

The present thesis is based on the following papers, which will be referred to by their roman numerals.

I. Ulvcrona, T., Lindberg, H. & Bergsten, U. 2006. Impregnation of Norway spruce (*Picea abies* L. Karst.) wood by hydrophobic oil and dispersion patterns in different tissues. Forestry 79(1), 123-134.

II. Ulvcrona, T. & Bergsten, U. 2006. Possibilities for compositional tailoring of Norway spruce (*Picea abies* L. Karst.) wood using a hydrophobic oil impregnation process. Holz als Roh- und Werkstoff URL: http://DOI10.1007/s00107-006-0137-5.

III. Ulvcrona, T., Lindberg, H. & Bergsten, U. Effects of initial water-filled porosity on macroscopic cracks and mechanical properties of Norway spruce (*Picea abies* L. Karst.) wood tissues after impregnation with a hydrophobic oil. Proceedings Fifth IUFRO Workshop: Connections between Forest Resources and Wood Quality: Modelling Approaches and Simulation Software. Waiheke Island Resort, Auckland, New Zealand November 20-27, 2005. (In Press).

IV. Ulvcrona, T. Determination of shrinkage/swelling coefficients of various types of Norway spruce (*Picea abies* L. Karst.) tissues after impregnation with a hydrophobic oil. (Manuscript).

V. Lindeberg, J. & Ulvcrona, T. Potential use of Near-Infrared Spectroscopy to evaluate hydrophobic oil uptake in wood. (Manuscript).

Papers I, II and III are printed with the kind permission from the publishers.

Introduction

General background

As pointed out by various authors, e.g. Hovgard & Hansen (2004), forest industries must continue to develop innovative new raw materials, processes and products in order to remain competitive. Similarly, Bowyer (2000) argues that there is a need to invigorate the wood science and technology profession. Solid wood is generally characterized as an anisotropic, heterogeneous and hygroscopic material nowadays (Kollman & Coté, 1984; Kifetew *et al.,* 1998; Thuvander, Kifetew & Berglund, 2002). Furthermore, its properties are affected by both silvicultural practices and subsequent processing (Mathias *et al.,* 1991; Rowell, 1996; Kifetew *et al., op. cit.*; Megnis *et al.,* 2002). Therefore, there is a need to develop inter-disciplinary approaches and to integrate forestry and silvicultural practices with both wood and process technologies in order to increase the efficiency of the development of innovative new solid wood products.

The businesses that are most likely to develop and launch new products successfully are those that have a clearly articulated new product development (NPD) strategy (Carbonell, Escudero & Aleman, 2004). Tzokas, Hultink & Hart (2004) state that the NPD process involves the generation of new product ideas, the development of initial product concepts, assessments of their commercial attractiveness, the development of the actual products, market tests, and launches of the products in the marketplace. When the NPD process starts is it essential to have an understanding of the manufacturing process and associated costs, the process technology and equipment, and the feasibility of the development and manufacture of the product (Story, Saker & Smith*,* 1998). According to Hart *et al*. (2003) and Carbonell, Escudero & Aleman (*op. cit.*), technical feasibility is the most frequently used go/no go criterion in the early stages of the NPD process. In subsequent stages, i.e. during product testing phases, managers need to ascertain that the product has been developed according to set specifications (Tzokas, Hultink & Hart, *op. cit.*). Carbonell, Escudero & Aleman (*op. cit.*) argue that placing importance on customer acceptance criteria at every stage of the process correlates positively with the ultimate success of new projects. In-depth knowledge of material properties within both the stem wood and processed wood-based materials is an essential element of the NPD process. Future production and use of wood-based materials with homogenous material properties in successful NPD processes might function as strategic resources within supply chains that use them. Thus the development of wood-based materials with homogenous properties might provide valuable strategic resources for NPD processes within supply chains that use (or could use) wood. The most obvious ways in which the use of such materials could benefit the industry are perhaps that they could allow product specifications to be more strictly defined by designers and tighter tolerances could be applied in the following manufacturing process, thereby reducing variability, waste and costs, while maintaining (or increasing) customer satisfaction.

Wood tends to decay when it is exposed to conditions that favour the development of rot fungi and bacteria. The moisture content of the wood, the temperature of the surrounding environment, and the relative humidity are the most important factors governing fungal establishment both on and within wood (Råberg *et al.,* 2005). Furthermore, the natural durability of wood varies between species and tissue types (Anon., 1994) and the durability of wood from several tree species needs to be improved in order to make it suitable for use in exposed conditions. Durability is generally increased by treatment with some sort of industrial impregnation process. In other words, wood is impregnated in order to increase the life time of wood products that would otherwise tend to decay rapidly. Wood is also often impregnated with fire retardants and/or substances intended to increase its dimensional stability. Impregnation is therefore an important tool in many NPD processes.

Impregnation for wood preservation

To preserve wood a number of impregnation methods and impregnants are commonly used, singly or in combination. Generally they either shift one or more specific variables (notably moisture, oxygen or nutrient contents, or temperature) to a range in which fungal development is inhibited or they have a directly toxic fungicidal action.

Impregnation with toxic substances

Many preservative systems have a directly toxic fungicidal action or promote the absorption by the fungi of toxic materials in sufficient quantities to prove fatal (Richardsson, 1993). Important features of toxic systems are that the treatment must be long-lasting, providing good resistance to losses by leaching, volatilization and oxidation; yet the toxic agents must remain available to the attacking fungi. Copper-chrome-arsenic (CCA) preservatives have proved to be very reliable toxic systems in practice, thus CCA can be viewed as a common type of preservative that is internationally used for wood preservation. Many CCA wood preservatives are now available, with varying proportions of copper, chromium and arsenic components that are incorporated in the wood as oxides or salts (*ibid.*).

A general objective of recent research regarding toxic preservation systems has been to increase knowledge regarding the use of new raw materials and new formulas of preservatives. Toxic preservatives that have been considered in recent research include (*inter alia*) boron and copper-chromium boron (CCB) (Abdurrohim, Nurjaman & Hadiane, 2003; Furuno *et al.,* 2003; Baysal & Yalinkilic, 2005; Yamagushi, 2005; Dhamodaran & Gnanaharan, 2006), CCA (Lee *et al.,* 2001; Diaz *et al.,* 2002), Creosote (Diaz *et al., op. cit.*), Tanalith-C (Usta & Guray, 2001) and pentachlorophenol (Schmidt & Westberg, 2001). Another important element of any preservative treatment is the mode of application, since ideally the preservatives should penetrate deeply into the wood. Preservative substances can be applied by simply spraying the wood, immersion or by various pressure and/or temperature treatments. These alternatives are briefly discussed below.

Water-based application techniques

Generally spray applications are most effective, i.e. achieve best penetration, with preservatives of low viscosity and low volatility (Richardsson, *op. cit.*). Wood porosity has a profound influence on both penetration and loading, but the surface texture has particular significance for spray treatments; a rough sawn surface allows far greater loading of low viscosity preservatives than a smooth surface. Generally, preservatives for spray applications are prepared in low viscosity, non-polar organic solvents, which penetrate far more efficiently into dry wood than polar or aqueous systems. Furthermore, it has been shown experimentally that a conscientiously applied flood spray is equivalent to 10-15 seconds immersion. It is therefore frequently argued that spraying is as efficient as a brief dip treatment (*ibid.*).

Simple low-viscosity preservative solutions are often used in commercial immersion treatments (*ibid.*). They penetrate rapidly and the immersion period is adjusted to achieve the desired penetration. Immersion for a period up to about 10 minutes is usually described as dipping, whereas longer immersion treatments are known as steeping. Diffusion treatments involve immersion or spray procedures to load the surface of the wood with a preservative that will subsequently diffuse slowly through the wood, to achieve the desired distribution. Freshly sawn green wood with moisture contents exceeding 50 % is immersed briefly in the preservative and then close-stacked and wrapped to prevent losses of water by evaporation. Storage periods of several weeks, or even months for thick pieces, are required to distribute the preservative throughout the wood. This treatment achieves relatively good penetration in very impermeable wood, apparently because the radial penetration pathways are still open in green wood, whereas they are closed in the dry wood that is typically used for pressure impregnation treatments (*ibid.*).

Several vacuum and pressure treatments are used commercially, both “full-cell” and “empty-cell”. In the traditional full-cell process a sequence of vacuum and pressure treatments is employed to thoroughly impregnate as much as possible of the porous spaces within the wood (*ibid.*). Normally the wood is introduced into a cylinder and a vacuum of 90 % or more is drawn for 15 minutes up to several hours depending upon the permeability and cross-section of the wood involved. The vacuum removes most of the air from the porous spaces within the wood, and is maintained while the cylinder is flooded with preservative; water-borne preservatives are generally used at ambient temperatures and warmed only to prevent freezing, crystallization or sludging. When the cylinder is full the vacuum is released and the preservative starts to flow into the porous spaces in the wood under the influence of atmospheric pressure. In order to encourage penetration pressure is then applied, typically 0.7 MPa - 1.4 MPa, and maintained for as long as necessary to achieve the desired penetration and retention, typically 1 – 5 hours, but occasionally several days, depending on the wood’s permeability and cross-section. After the required period the pressure is released and the preservative is removed from the treatment cylinder. Typically, a final vacuum is then applied in an attempt to remove excess preservative and avoid subsequent bleeding when in service. A more important function of the final vacuum may be to relieve the compressed state of the wood, thus allowing any excess preservative to be properly absorbed. In empty-cell processes wood is impregnated with preservative under high pressure on top of air trapped within the wood. This trapped air is later permitted to expand, ejecting preservative from the porous spaces but leaving the cell walls impregnated, or coated, with preservative (*ibid.*).

With empty-cell processes it is far easier to apply preservatives in such a way that they do not bleed in service, but empty-cell processes can only be used when the required retention can be achieved despite the recovery of preservative from the spaces within the wood (*ibid.*). In the Rüping process, after the cylinder has been loaded and sealed air pressures of 0.2 MPa to 0.4 MPa are usually applied for 10 to 60 minutes, depending on the permeability and sizes of the pieces of wood in the charge. The cylinder is then flooded with preservative without releasing the pressure, which is then increased up to perhaps 1.4 MPa, and this pressure is maintained until the required gross absorption of preservative is obtained. The pressure is then released and the preservative is removed from the cylinder, permitting the air trapped within the wood to expand and eject excess preservative from the porous spaces. In practice a vacuum of about 60% is drawn during this stage to encourage expansion of the trapped air. If the pressure is not released in this way there is a danger that the remaining pressurized air will cause continuing bleeding of preservative at the surface of the wood, but the final vacuum will reduce the pressure of trapped air to below atmospheric levels. Generally, empty-cell processes are employed for creosote treatments (*ibid.*).

An important task for the future is to develop effective treatments that do not involve the use of copper/chromium preservatives (Megnis *et al., op. cit.*; Humar *et al.,* 2004) or other toxic substances, due to ongoing pressures to avoid, or at least minimise, the amounts of toxins in wood products. This is, accordingly, a driving force for new research concerning the preservation of wood and related issues. For instance, methods to dispose of toxic waste from CCA-treated wood have been studied and discussed by Kakitani, Hata & Imamura (2003) and Kartal & Imamura (2003). New and innovative substances are also being evaluated; for example, the possible use of chitosan has been studied by Alfredsen *et al.* (2004) and Eikenes *et al.* (2005 a, b). Use of chitosan as a wood preservative would also probably provide interesting opportunities in the fishing industry. New processes are also being investigated; for example the main effects of supercritical fluid impregnation of conifer heartwood and wood from broadleaved species have been investigated by Schneider, Morrell & Levien (2005).

Preservation of wood with hydrophobic oils and permeability in wood

One way to decrease the adverse environmental impact of various impregnation processes is to use non-toxic impregnants like hydrophobic oils. These oils are expected to keep the moisture content below the critical level for wood-decaying fungi (Van Eckeveld, Homan & Militz, 2001). The oil fills the cavities in tracheid lumens, rays, and possible cracks in the wood, including those resulting from drying processes. Impregnation of Scots pine sapwood with various oils, including linseed oil, coco oil, and tall oils, has been reported to significantly increase its waterrepellence (*ibid.*). One process for impregnating wood with hydrophobic oil is the Linotech process, which has been shown in previous studies to allow Scots pine wood to be impregnated with various levels of Linogard, a linseed oil derivative (Olsson *et al.,* 2001, Megnis *et al., op. cit.*).

The permeability of different types of wood differs substantially. Thus, their treatability by impregnation processes also differs. Permeability is a measure of the ease with which a specified fluid can flow through a porous material under the influence of a pressure gradient (Siau, 1972). Hansmann *et al.* (2002) concludes that permeability varies within trees, between trees and between geographic locations, so general characterisations of the permeability of a given species have limited meaning. The flow of liquids into wood is influenced by many variables, such as the strength of viscous and capillary forces, the polarity of the liquid, the back pressure due to the compression of air inside the wood, the impregnation time, the length of the specimen in the flow direction, and both the permeability and anatomical structure of the wood (Siau, *op. cit.*). Theoretical models of permeability in wood are often initially based on Darcy's Law, which allows permeability coefficients to be calculated from flow rates obtained from Pouiseille's Law, specimen dimensional and viscosity data and pressure gradients (Hansmann *et al., op. cit.*). However, the physical processes governing flows in wood are complex, and there are important restrictions on the direct applicability of Darcy's Law to wood imposed by variables such as the presence of pores plugged by particles, air bubbles, molecular slip effects and gas compressibility during gas flow (*ibid.*). In addition, as pointed out by Siau (*op. cit.*), the presence of molecular slip flow in the pit openings complicates the construction of reliable models of gas and liquid permeability in wood. Furthermore, the dimensions of molecular flow paths inside the wood should be key determinants of its permeability, and the permeability of wood in different directions may vary substantially (Bramhall, 1971). Bolton (1988) states that the gaseous conductivity per length unit in Norway spruce heartwood show a non-linear pattern with a stable value at a specimen length equivalent to ~ 9 cells in series. Siau (*op. cit.*) impregnated wood of Douglas fir (*Pseudotsuga menziesii*) and Loblolly pine (*Pinus taeda*) with oil, and found that latewood penetration exceeded earlywood penetration in nearly all specimens and sapwood penetration was higher than heartwood penetration. Clearly, therefore, the anatomical features of the wood to be treated, the viscosity of the oil, treatment time and specimen size are all important factors to consider when discussing the amounts of oil retained, and its distribution, after impregnating wood with oil.

Spruce wood as a raw material for impregnation

A specific problem associated with Norway spruce (*Picea abies* L. Karst.) wood is that it is difficult to impregnate cost-efficiently using currently available commercial processes (Wardrop & Davies, 1961; Bailey & Preston, 1969; Banks, 1970; Boutelje, 1983; Vinden, 1984; Anon., 1994). The permeability of wood is strongly dependent on its moisture content (Hansmann *et al*., *op. cit.*), as well as the principal direction of the grain (Bramhall, *op. cit.*; Bolton, *op. cit.*) and various physical and chemical properties (Wardrop & Davies, *op. cit.*; Banks, *op. cit.*; Baines & Saur, 1985; Hansmann *et al*., *op. cit.*). A very large reduction in the permeability of spruce occurs during drying (Banks, *op. cit.*), largely due to permanent structural changes that occur in the wood during the drying process, mainly as a result of the aspiration of bordered pits (Vinden, *op. cit.*). In Norway spruce, a ray cell's relative porous area is estimated to comprise only 5 % of the total cell wall area, compared to 50 % in Scots pine, a non-refractory species (Nyrén & Back, 1960). Furthermore, the parenchymatic cell wall is thicker in Norway spruce than in Scots pine (Liese & Bauch, 1967). Ray tracheids in spruce are also often interrupted by a parenchyma cell at the junction of the annual ring, which may explain why penetration often stops abruptly at a particular annual ring (Baines & Saur, *op. cit.*). Heartwood is usually much less permeable than sapwood due to pit aspiration and incrustation in it (Comstock, 1965; Siau, 1984). Another major difference between the wood tissues/types sapwood and heartwood that directly affects permeability is that the moisture content of sapwood is naturally much higher than that of heartwood (Hansmann *et al., op. cit.*). Bordered pits are larger (Siau, *op. cit.*) and more abundant in the earlywood than in the latewood of *Picea* tracheids. The majority of bordered pits in Norway spruce occur in the radial walls and tangential pitting is not uncommon. Normally in a growth ring tangential pitting is restricted to the last four or five tracheids in a radial row of tracheids, i.e., in the latewood (Laming & Welle 1971). In latewood some pits remain open even after drying (Baines & Saur, *op. cit.*). Permeability in latewood is generally higher than in earlywood when in a dry state (Wardrop & Davies, *op. cit.*).

In summary, the extensive commercial use of wood from Norway spruce implies that altering its material properties could be commercially important. However, mechanisms governing the possible retention of oil by different types of woody Norway spruce tissues need to be better understood before treated wood materials can be readily produced from them using a hydrophobic oil impregnation process.

The Linotech process is the hydrophobic oil impregnation process that has been used in this thesis to impregnate Norway spruce wood with various levels of Linogard. The process includes, in principle, a pre-acclimatisation phase, a pressure phase and a pro-acclimatisation phase with samples still in the oil; however, without any pressure applied and an eventual final vacuum phase. The linseed oil derivative Linogard is manufactured from cold-pressed linseed oil in several steps including, for example, heating, addition of inorganic acid and alkali as well as separation of precipitated materials (Anon., 2003). Linogard essentially consists of linoleic, linoic and oleic acids, mainly in the form of triglycerides; its content of free tocoferol being less than about 100 ppm (*ibid.*).

Objectives

The overall goal of the work underlying this thesis was to develop new knowledge to facilitate the production of Norway spruce wood-based materials that are both more durable and more homogenous than current products by using a hydrophobic oil impregnation process. Some essential intermediate objectives were therefore:

1. To quantify the amounts of a hydrophobic linseed oil derivative taken up, at both macroscopic and microscopic levels, when used in the Linotech process to impregnate Norway spruce wood. Since anatomically and chemically distinct wood tissues are likely to differ in their responses to the impregnation process (see above) we also compared uptake patterns in (i) heartwood and sapwood, (ii) mature wood and juvenile wood and (iii) earlywood and latewood. In addition, the dispersion of the oil within the year rings and tracheid cells was studied. (Paper I).

2. To quantify the effects on oil uptake in Norway spruce wood of its water-filled porosity, its basic density and various pre-treatments designed to change its moisture contents. Since anatomically and chemically distinct wood tissues should differ in their responses to impregnation processes we compared these effects in (i) heartwood and sapwood, and (ii) mature wood and juvenile wood. (Paper II).

3. To test the hypotheses that macroscopic cracks resulting from the impregnation process differ between wood tissues with different water-filled porosities, and that the impregnation process does not affect the wood's flexural modulus and bending strength in longitudinal direction: i.e., (i) impregnation causes more cracks to develop in heartwood wood than sapwood and (ii) neither the flexural modulus nor bending strength in longitudinal direction are affected by the impregnation process. In addition, the drying effect of the impregnation process was studied. (Paper III).

4. To examine the dimensional stability of specific pre-treated, dried and green tissues of Norway spruce and to quantify the effects of impregnation on their stability. (Paper IV).

5. To evaluate the potential use of NIR spectroscopy for evaluating oil retention levels. (Paper V).

Summary of the papers

Paper I

In this study the amount of the hydrophobic linseed oil derivative Linogard taken up was quantified, at both macroscopic and microscopic levels, when used in the Linotech process to impregnate Norway spruce wood. In addition, its dispersion within the year rings and tracheid cells was studied.

Samples were taken from fifteen trees. To determine the level of uptake after impregnation, the linseed oil was extracted from the impregnated wood using methyl-tertiary-butyl-ether. The uptake was quantitatively analysed by comparing the weights of specimens following impregnation both before and after oil removal using both standard weight measuring techniques and x-ray microdensitometry.

Furthermore, scanning electron microscopy was used to examine oil dispersion and cell walls in the treated wood.

In earlywood, the initial moisture content had a clear effect on the impregnation results. Six times more oil was taken up when the moisture content was greater than ~150 % than when it was less than 30 %. Theoretical calculations, based on density levels, suggest that the water-filled porosity of the wood (water volume divided by porosity volume) was positively correlated with the linseed oil uptake, and more strongly correlated in earlywood than in latewood. There were also significant differences in uptake between different wood tissues; heartwood/mature wood and heartwood/juvenile wood showed 10-20 % weight increases due to linseed oil uptake, compared to 30-50 % in sapwood/mature wood. Examination by scanning electron microscopy confirmed these uptake patterns.

The moisture content after impregnation was about 5 %, irrespective of the Linotech process parameters, tissue type, and initial moisture content. In conclusion, the impregnation process used here resulted in high levels of well-dispersed linseed oil uptake (i.e., full impregnation of Norway spruce was found to be possible) and should facilitate drying.

Paper II

The effects of the water-filled porosity and basic density of Norway spruce wood, and treatments prior to impregnation, on its retention of oil were quantified. Specimens (in total 168) of four distinct tissue types were sampled from a single tree. The Linotech process was then used to impregnate specimens with hydrophobic linseed oil to the largest span possible regarding retention of oil. Oil retention levels were quantified and the values obtained were used to generate linear regression equations for predicting oil retention in each wood tissue type from the two physical characteristics (the water-filled porosity and density of the wood) examined.

The amount of oil retained differed significantly between tissue types. Juvenile sap- and heart-wood retained up to 62 % and 34 %, respectively, per dry weight of the wood, while mature sap- and heartwood tissue types retained up to 125 % and 27 %, respectively. In addition, treatments prior to impregnation affected the oil retention levels. Dried specimens retained less oil than specimens with higher water-filled porosity. Furthermore, water-filled porosity and basic density had significant effects on oil retention levels within tissue types. The relatively good fits of the derived linear regressions predicting the retention of oil within all tissue types (r^2 0.85, 0.79, 0.66 and 0.69 for mature sapwood, juvenile sapwood, mature heartwood and juvenile heartwood, respectively) indicate that there is scope for producing compositionally tailored wood materials with specified properties.

Paper III

This study tested the hypotheses that macroscopic cracks resulting from the impregnation process differ between wood tissues with different water-filled porosities and that the impregnation process does not affect the flexural modulus or bending strength in longitudinal direction, i.e., (i) impregnation causes more cracks to develop in heartwood wood than sapwood and (ii) neither the flexural modulus nor bending strength in longitudinal direction are affected by the impregnation process. In addition, the drying effect of the impregnation process was studied.

Cracks were measured followed impregnation at three different vertical locations in 40 sapwood and 18 heartwood samples (500 x 25 x 25 mm; oil uptake levels ranging from 3.2 to 43.9 %). The flexural modulus after impregnation was measured in longitudinal direction with loads applied both radially and tangentially to 18 sapwood specimens with oil uptake levels ranging from 5.8 to 43.9 %. Fourteen sapwood specimens were used as controls.

Significant differences in the cracks were found between different vertical locations in the samples. No surface cracks were found in sapwood samples and only occasional cracks in heartwood samples. Internal cracks with a radial orientation occurred more frequently in heartwood samples than in sapwood samples. There was a clear negative correlation between the initial water-filled porosity and the abundance of cracks that developed. The flexural modulus was largely unaffected by impregnation; mean values in longitudinal direction, measured with radially and tangentially applied loads, did not differ much between impregnated sapwood and control specimens, and the initial water-filled porosity had no effect on the resulting flexural modulus. However, the variation was lower for impregnated sapwood specimens than for control specimens.

Paper IV

In this study the principal characteristics of specific pre-treated, dried and green tissues of Norway spruce were examined, and the effects of impregnation on their dimensional stability were quantified. For this purpose, shrinkage/swelling coefficients (S) were monitored at each experimental stage and statistically analysed with regard to both wood tissue types and treatments.

Ten samples of each of four types were taken, corresponding to four distinct tissue types: heartwood/mature wood, heartwood/juvenile wood, sapwood/mature wood and sapwood/juvenile wood. From each sample five specimens (one for each treatment, see below) were prepared. This procedure was replicated twice, generating 10 series of sets of five paired samples, in order to allow more statistically rigorous analysis of the data. The dimensions of each specimen were 40*30*10 mm (longitudinally*tangentially*radially).

Mean S-coefficients following the impregnation process were between -12.1 and -13.7 % for pre-treated and green specimens, and +1.4 % for dried specimens, indicating that cell wall bulking may have occurred in the latter. The effects of the impregnation in terms of Anti-Swelling Efficiency (ASE), tested in a water soaking experiment, were strongest in dried mature sap and heartwood tissue specimens (29.2-22 % and < 20 %, respectively; a significant difference). However, it also had positive effects on specimens representing other tissue and pre-impregnation treatment combinations.

The treatments eliminated much of the variation in S-coefficients observed between untreated tissues. This means that the treatments applied in this experiment diminished much of the natural variability within the wood, and the remaining differences can be regarded as differences between different wood-based materials. Thus, there appears to be scope to develop wood materials with tailored dimensional stability characteristics.

Paper V

Here, the potential use of Near-Infrared Spectroscopy measurements to estimate global oil uptake in impregnated wood was assessed.

Nine mature sapwood specimens were selected for analysis from a total of 152 impregnated juvenile and mature heart- and sap-wood specimens obtained from a single Norway spruce (*Picea abies* L. Karst.) tree. The selected specimens were specifically chosen to span a wide range of oil uptake levels (between 8 and 125 % of wood dry weight). The dimensions of each specimen were 50*25*15 mm (l*h*w).

NIR spectra were acquired using a spectrometer equipped with a fibre optic probe, placed < 1 mm from the surface perpendicular to the longitudinal direction at three points along the radial direction (from the pith towards the bark) for each specimen. Each spectrum consisted of an average spectrum from 32 scans at each measurement point, and the three spectra from each sample were summed. The spectral data, together with the densities of linseed oil obtained for each sample, were imported into multivariate analysis software, and the correlations between linseed oil densities and NIR absorbance were analyzed by Partial Least Squares (PLS) regression with unit variance scaling. Nine PLS models were created, each excluding one of the nine samples. The linseed oil densities were then predicted using the PLS model for each sample from which the sample had been excluded, and regression coefficients between the observed and predicted linseed oil densities were calculated. The overall regression equation, including data from all samples, was $0.97x+0.016$, with a regression coefficient (r^2) of 0.89 and a standard error of 0.056.

The relatively good fit of the regression equation clearly indicates that there is scope to develop an industrial method for predicting oil uptake levels based on the investigated Near-Infrared Spectroscopy approach.

Discussion

Materials and methods

Since wood can be generally regarded as an anisotropic, heterogeneous and hygroscopic material, numerous interacting factors have to be considered by researchers investigating its material properties. Furthermore, silvicultural practices also influence its properties, affecting various characteristics such as the relative proportions of different types of tissues, and the abundance of knots (Vestøl & Høibo, 2001). There are no reasons to believe that there are any principal differences regarding impregnation possibilities between sound knots and other types of xylem wood. However, knotwood is strongly associated with irregularities regarding fibre orientation and furthermore, also with reaction wood. A brief description of relevant tissue types includes the following: Sapwood is generally referred to as the part of the xylem that has an ability to transport water. Juvenile wood is being formed in a certain number of rings from the pith, and the period of juvenile wood formation and the properties of juvenile wood is probably under strong genetic control (Kyrkjeeide, 1990).

For these reasons, stem wood was divided into several types of tissues (i.e. juvenile and mature sap- and heart-wood), and only clearwood was used in the studies underlying this thesis. In addition, relevant physical and chemical characteristics were monitored throughout all experiments by standard methods that are routinely used in wood science.

Samples and specimens examined in the studies described in Papers I-V were impregnated using an apparatus designed to allow high resolution process settings. A complete description of the non-commercial process settings used for the studies described in Papers II, IV and V includes: a 5 minute pre-acclimatisation phase, a 15 minute pressure phase (1 MPa with an oil temperature of 100°C) and a 20 minute pro-acclimatisation phase with samples still in the oil; however, without any pressure applied. Commercial process settings used for the studies described in Papers I and III included also a final vacuum phase. Furthermore, treatment times might very well differ between the commercial process schedule and the non-commercial schedule presented above. The linseed oil derivative “Linogard” was used as impregnant in all the studies described in Papers I-V. No problems related to the impregnation process were considered to occur from the use of this apparatus; impregnation treatments were always replicated at least twice in all of the studies presented in Papers I-V, to allow for statistically rigorous analysis of the results.

Estimates of oil uptake levels were based on weight measurements taken before and after extraction in the studies described in Papers I, III and V, while calculations of global oil uptake were based on weight measurements before and after impregnation for the studies in Papers II and IV. Calculations of the wood’s moisture content (MC), resin content and density were based on measurements made on sub-samples adjacent to the samples and specimens used for impregnation in the studies presented in Papers I-V. The properties of the raw materials have not generally been described in such detail in previous studies related to impregnation

(Mathias *et al., op. cit.*; Li, Furuno & Katoh, 2001; Van Eckeveld, Homan & Militz,. *op. cit.*; Obataya, 2005; Hyvönen, Piltonen & Niinimäki, 2006). However, there are also some good examples of studies in which more thoroughly characterised wood raw materials were used (Banks, *op. cit.*; Olesen, 1977; Vinden, *op. cit.*; Militz, 1993; Militz & Homan, 1993). The results of these studies collectively indicate that the permeability of the wood is closely related to the material properties mentioned above, implying that it is important to closely monitor raw material properties in samples used for impregnation studies. Such monitoring not only improves the resolution of the analysis and validity of any conclusions drawn, but also facilitates comparisons of results from different studies and the design of future experiments.

For the microscopic studies of oil uptake and dispersal presented in Paper I samples were mounted on a tray and exposed to x-rays in a Woodtrax instrument. The minimum density, mean earlywood density, mean latewood density, and maximum density within annual rings were determined for each sample from the Woodtrax images. The oil contents were obtained, as percentages of wood dry weight, from the Woodtrax data. The oil was extracted from the wood in a two-step process as described above, and the x-ray measurements were then repeated. The uptake was quantified by comparing the density values of each half-slice examined pre- and post-extraction of the oil, after correcting for the resin contents of each annual ring, determined as described above. This procedure allowed the retention of the oil to be studied in much greater detail than in many other investigations of impregnation with hydrophobic oils (Van Eckeveld, Homan & Militz*, op. cit.*; Hyvönen, Piltonen & Niinimäki, *op. cit.*).

Scanning electron microscopy analyses were carried out on the samples chosen for x-ray analyses to evaluate the range of the oil uptake levels using a CamScan S4-80DV electron microscope. Three consecutive 6 x 6 x 5 mm specimens were taken from one end of each 30 mm-sample and sputter-coated with gold to allow SEM examination of the wood from the surface through to the centre of sample.

Internal cracks were measured with a ruler and the naked eye in the study presented in Paper III. In addition, the flexural modulus and bending strength of wood specimens after impregnation were measured in longitudinal direction with loads applied both radially and tangentially to them. The outer dimensions of specimens were measured with a digital caliper to calculate shrinkage and swelling coefficients as well as treatment effects in the study reported in Paper IV. These analytical techniques were selected because they are all routinely used in wood analyses (Moore *et al*., 1984; Malmquist, Esping & Nilsson, 1988; Obataya, *op. cit.*).

The variable ‘water-filled porosity’ has not previously been used in wood science and technology. However, it is widely known and applied in soil science, for instance by Barton *et al.* (1999). The water-filled porosity of samples examined by macroscopic and microscopic analyses was calculated as follows. First, the porosity (P) was determined from the average density values obtained from the macroscopic or microscopic analyses and the mean value for cell wall density (1500 kg m^{-3}) published by Dinwoodie (2000). The water-filled porosity in the sample was then calculated as: available water volume in one m^3 of wood/porosity (P) in one m^3 of wood multiplied by 100. The available water volume in m^3 was calculated as: (mean density value * initial moisture content) * (1-0.3), where 0.3 is assumed to be the fibre saturation point (30% moisture content).

NIR spectra were acquired using a NIR spectrometer with a fibre optic probe, placed < 1 mm from the surface perpendicular to the longitudinal direction at three points along the radial direction (from the pith towards the bark) for each specimen. Each spectrum consisted of an average spectrum from 32 scans at each measurement point. The spectra were recorded at room temperature (ca. 20 °C) for every second wavelength from 400 nm to 2498 nm (1050 wavelengths), and the reflectance spectra (r) were transformed into absorbance values (a) from the equation: $a = \log(r^{-1})$. The three spectra from each sample were summed, and the resulting spectral data, together with the densities of linseed oil for each sample, were imported into Simca-P version 3.01 software (Umetri AB, Umeå, Sweden) for multivariate analysis. The literature concerning measurements of oil within wood is sparse, but the selected method seemed at least to be appropriate for initial evaluations of the potential utility of NIR procedures. NIR techniques have been applied in several other published analyses of wood properties (Hoffmeyer & Pederson, 1995; Axrup, Markides & Nilsson, 2000; Thygesen & Lundqvist, 2000a; Thygesen & Lundqvist, 2000b; Gindl *et al.,* 2001; Hauksson *et al.,* 2001; Schimleck, Evans & Ilic, 2001; Thumm & Meder, 2001; Schimleck, Evans & Matheson, 2002; Hodge & Woodbridge, 2004; Kelly *et al.,* 2004) and their results collectively indicate that they could be highly suitable for such purposes.

Oil uptake during impregnation

It is important to identify and study in detail factors that affect oil uptake in Norway spruce wood in practice, since the obvious ultimate aim for a future commercial hydrophobic oil impregnation process is to introduce desired levels of oil into the wood structure. Furthermore, this should preferably be done in a predictable way.

The data presented in Papers I-V indicate that it is possible to fully impregnate Norway spruce wood with linseed oil in the Linotech process. This is, of course, an important result because wood from Norway spruce is considered to be a refractory material. The studies presented in Papers I, II and IV also show that levels of oil uptake differ between different tissue types subjected to the same impregnation protocol. Generally, uptake was higher in sapwood than in heartwood, higher in mature than in juvenile wood (Paper II), and generally higher in earlywood than in latewood (Paper I). In Paper I the variable water-filled porosity was shown to be the single most important factor governing oil uptake, although it has not previously been used as an explanatory factor in studies regarding wood science and technology. The clear effect of water-filled porosity also suggests that the oil might penetrate the wood as a component of an oil-in-water emulsion when the moisture content (MC) of the wood exceeds the fibre saturation point (Paper I); which also, at least partly, explains why the investigated method seems to allow better penetration in spruce wood than water based impregnation methods does. The possible interaction between oil and water is further supported by the conclusion that the process also facilitates drying (Paper I, II & IV); an interaction should be fully possible since the water is removed from the wood and the oil inserted through the same porous volume under relatively high temperature and pressure conditions.

The positive correlation between oil uptake and water-filled porosity was further investigated in the study presented in Paper II. It was concluded that it is possible to derive regression equations that can provide reasonably accurate predictions of oil uptake levels from data on physical characteristics of the wood. The results indicate that the regression coefficients are higher for sapwood tissues than for heartwood tissues. However, factors measured at a macroscopic level were used in this study, which were clearly more suitable for sapwood than for heartwood tissues, so this difference between tissue types would probably be diminished if more explanatory factors were considered in future studies.

Crack development due to impregnation

Abundance and size of macroscopic cracks within any produced material clearly affects its potential uses, since they may negatively affect both its mechanical and aesthetic properties, and in worst case scenarios make it unusable for some purposes. The studies presented in Paper III conclude that no surface cracks occurred in any of the 40 sapwood samples examined in them, and surface cracks occurred only occasionally in heartwood samples. However, internal radial cracks occurred in both sapwood and heartwood samples. The radial orientation of the internal cracks implies that they originated from a drying process. There was a clear negative correlation between initial water-filled porosity and internal crack development. The lower incidence of cracks found in samples with relatively high initial water-filled porosity might be due to the cell walls being more elastic in such samples than in samples with lower water-filled porosity and/or heartwood tissue types. Furthermore, more cracks developed within heartwood than in sapwood. Literature concerning cracks due to impregnation processes is sparse, but Malmquist, Esping & Nilsson (*op. cit.*) report that surface crack development is positively correlated to the temperatures used in the treatment process. There seem to be differences in crack development between traditional water-based processes and hydrophobic oil impregnation processes. Various raw material properties and features of the processes applied are probably interactively responsible for the observed differences. The development of both surface and internal cracks seems to be positively associated with low initial moisture contents, but high temperatures seem to have stronger negative effects in water-based impregnation processes.

Resulting material properties

Static bending properties are highly important for material that may be used in load-bearing constructions. Therefore, we investigated the static bending properties of the impregnated material (Paper III), and concluded that the water-filled porosity before impregnation did not have any effect on the studied mechanical properties. However, the range of flexural moduli was lower for impregnated sapwood specimens than for reference specimens. The flexural moduli of impregnated specimens were not in any way higher than those of reference specimens, indicating that the negative effects of any structural damage that the impregnation process may have caused were compensated for by hydraulic effects (cf. Megnis *et al., op. cit.*).

Dimensional instability often poses serious problems for wood products in various potential applications (Frühwald, 2006), so the effects of impregnation on dimensional stability were evaluated in the study presented in Paper IV. It was concluded that the treatments eliminated much of the variation in swelling and shrinking coefficients observed between untreated tissues. This means that the treatments applied in this experiment diminished much of the natural variability within the wood. Furthermore, indications that cell wall bulking might have occurred were also found (Paper IV).

Effects of the applied hydrophobic oil impregnation treatment on durability were not evaluated in the studies presented in this dissertation, mainly because of the long time span, often several years, required to measure effects on durability with any certainty in field tests. However, the positive effects on dimensional stability, originating from reduced water uptake, reported in Paper IV indicate that impregnation should also increase durability.

Use of Near Infrared Spectroscopy for evaluating uptake levels

Paper V presents results from a study with limited scope; and the relatively small number of specimens (nine) used makes it difficult to draw detailed general conclusions from it. However, the results indicate that there is scope to develop reliable, accurate evaluation methods based on NIR techniques, since it was readily possible to calculate regression equations that predicted oil uptake levels with satisfactory accuracy. Furthermore, the deviations between observed and predicted values of global oil uptake did not seem to be correlated with the uptake levels, which is encouraging since it indicates that NIRS analysis can be applied to samples with a wide range of retention levels. Measurements were done in a step-wise manner, but it was concluded that continuous scans of surfaces of the specimens would provide better predictions, due to inhomogeneities in uptake levels (Paper I). Therefore, in future studies specimens should be used for which the pre-impregnation history and characteristics are known as thoroughly as possible, and calculations should be based on oil uptake data obtained from both macroscopic/global and microscopic levels to maximise the value of the results for both industrialists and researchers.

Silviculture and wood impregnation

The development of silvicultural management practices that facilitate the production of raw materials with specified material properties requires further investigations on how desired properties develop within trees. Current uncertainties are exemplified by the statement by Pape (2001) that reported correlations between wood density and its vertical location in the stem are inconsistent, since both positive and negative correlations have been found (cf. Nylinder, 1953; Nylinder & Hägglund, 1954; Johansson & Persson, 1997). Silvicultural practices have a profound impact on the development of individual trees, and their genotypes also (of course) affect variables such as wood density within trees (Klem, 1957; Flaete & Kucera, 1999). In addition, climatic factors (notably those that determine the length of the growth season) also affect these variables. For example, a later ending of the growing season leads to a higher latewood percentage (Pape, *op. cit.*). This might be one reason why differences between individual trees are generally much greater than differences between provenances (Persson & Persson, 1997). Therefore, more research is clearly needed on the relationships between diverse factors affecting variables within trees that are less strongly related than has traditionally been recognised. Ståhl & Karlmats (1995) argue that there is probably no casual relationship between ring width and wood density. However, they are both related to factors such as annual weather conditions (*ibid.*). Collectively these conclusions imply that it is essential to develop silvicultural management practices that allow material properties within the stem wood produced to be both monitored and controlled. Identifying the properties desired for the final wood-based material is probably a useful starting point for attempts to identify desirable properties of trees and appropriate silvicultural management practices to promote them. Desired properties within stemwood used for hydrophobic oil impregnation processes are dependent on the desired levels of oil uptake within different tissue types. Thus, a wide range of wood features and processes should ideally be considered, including the development of different types of tissues and knots, moisture and resin contents and density at the macroscopic level, while at the microscopic level fibre lengths and microfibrillar angles will probably be the most important factors to consider. It is however interesting to further investigate possibilities to integrate studies regarding wood and processing processes like the one discussed here also with more detailed anatomical studies of wood (cf. Eriksson, 2005).

Practical implications

The impregnation process

The results presented in Papers I, II & IV clearly indicate that it now is possible to use Norway spruce wood as a raw material in future industrial wood preservation production chains, as well as Scots pine (*Pinus sylvestris* L.) sapwood, which is currently the only type of wood that is generally used in industrial impregnation processes in Sweden. The potential use of wood from another tree species should clearly increase the scope of many procedures both in silvicultural management practices and further within the supply chain.

Wood is dried for various purposes at different stages throughout industrial production chains; and possibilities to achieve high levels of oil uptake after drying should be advantageous for both technical and economic reasons. It seems possible to develop practices that also allow high uptake of oil after initial drying in future industrial production chains. In addition, there seems to be scope to use the impregnation process itself as a drying stage within industrial production chains. Collectively, these findings strongly suggest that it is feasible and desirable to develop production chains in which knowledge regarding both raw material properties and process effects are used to optimise the order and settings of the applied procedures (cf. the discussion of NPD in the *Introduction*).

Predicting oil uptake levels is also clearly important for any production chains in which oil impregnation treatments are applied, so there are good reasons for exploring ways to develop regression equations with greater predictive power. Furthermore, development of measures that allow oil uptake levels in individual components to be predicted would give the industry's production chains much greater flexibility, since the levels of oil retained by different tissues subjected to the same impregnation treatment vary (Papers I and II), and appropriate sorting of raw materials for different potential end-uses would reduce both waste and variability. The study presented in Paper II concluded that there is scope to calculate regression equations that can provide satisfactory predictions of oil uptake levels from data on wood properties, and could be extensively used to optimise supply chains in which raw materials are sorted and directed into appropriate process streams.

To summarize, the clear effect of percentage of water-filled porosity on oil uptake (Papers I and II) together with possibilities to calculate regression equations that can predict oil uptake levels in different tissues (Paper II) may provide a basis for further research aiming to (1) develop a classification system for oil-based impregnation processes, (2) facilitate the production of designed products with known material properties, (3) elucidate ways to identify raw materials that are suitable for oil-based impregnation processes, and (4) develop silvicultural measures that facilitate the production of suitable raw materials for oil-based impregnation processes.

There is a need to develop evaluation processes that meet the needs of effective and flexible production processes; i.e. allow individual components of the wood to be rapidly evaluated. In the batch approach that is generally used today uptake levels of preservatives are calculated from the total volume of wood and total volume of impregnated liquid (obtained by measuring metal concentrations, for preservatives that have known concentrations of metal). Furthermore, samples are taken from the production streams and later evaluated by other institutions in order to achieve clearance from branch organisations to use whatever desired marking they have on products. This approach does not readily allow evaluation at an industrial (i.e. on-site) level.

The availability of methods that allow individual processing units to control the quality of produced components autonomously would greatly facilitate new product development processes. NIR-based methods, such as those described in Paper V, could be developed into on-site evaluation procedures by combining them with close monitoring of the properties of the raw materials used. If well defined and properly sorted raw materials are used, NIR-based evaluation methods could be applied to check that desired oil uptake levels are maintained at both batch and component levels by using randomly selected samples taken from the production streams. Such evaluation methods could reduce both waste and variability within most production processes and facilitate the production of products with thoroughly characterised properties.

Impregnation and material properties

For many potential uses of these materials it is clearly important for there to be few surface cracks. Frequent development of surface cracks in the impregnation process can disqualify produced materials from many potential end-uses; and the results suggest that production chains should consider impregnating their products as closely as possible to the final stage.

However, the occurrence of internal cracks is interesting, since they are believed to originate from a drying process, clearly suggesting that there are interactive effects between the properties of the raw materials and the impregnation process. Overall, the presented results imply that it is important to monitor the raw materials in industrial production chains carefully if the presence of internal cracks is undesirable in the final products, for instance in products that may be planed or used to make joints.

Generally, the findings that the development of internal cracks is related to the initial raw material properties, that the flexural modulus does not change much after impregnation with hydrophobic oil, except that it may become more homogenous in the longitudinal direction after impregnation in sapwood, indicate a need for more research on material properties. Eventually it may be possible to produce a wood-based material with more homogenous static bending properties by using raw materials with known and selected characteristics in an impregnation process that is tailored to the properties of the raw material.

Furthermore, the results imply that it may be necessary to monitor the flexural moduli of the raw materials to produce components with specified flexural moduli. The process did not significantly either enhance or reduce the mean flexural moduli values, and the fact that no negative effects of the process in this respect were found is encouraging for future possible uses of the process. However, it should be stressed that creep characteristics of the impregnated materials have not yet been evaluated, despite their importance in load-bearing constructions.

Overall, the positive results of these studies indicate that it should be possible to control more of the variability in the properties of wood as a raw material, provided that its initial properties are properly monitored and controlled throughout the processing steps. As exemplified in Fig. 1a, b.

Effects of hydrophobic oil impregnation in general, and the Linotech process in particular, on durability have not been evaluated in the studies presented in this dissertation. However, the results highlight possibilities to produce materials with well-characterised, homogenous durability, since it was possible to impregnate all of the tested Norway spruce tissues (Paper I). Impregnated materials had lower water uptake rates than reference specimens in subsequent soaking trials (Paper IV)

Fig. 1a. Schematic illustration to show guidelines for processing chains; mature sapwood (a), juvenile sapwood (b), respectively. Diagrams, from left to right, presents relations between properties in the tissue type (i.e. the raw material), oil uptake in relation to important tissue properties within a given impregnation schedule and ASE within a certain climate and time span (i.e., one important material property) as a result from two different treatments made prior to impregnation with an hydrophobic oil. Figure is based on papers II and IV.

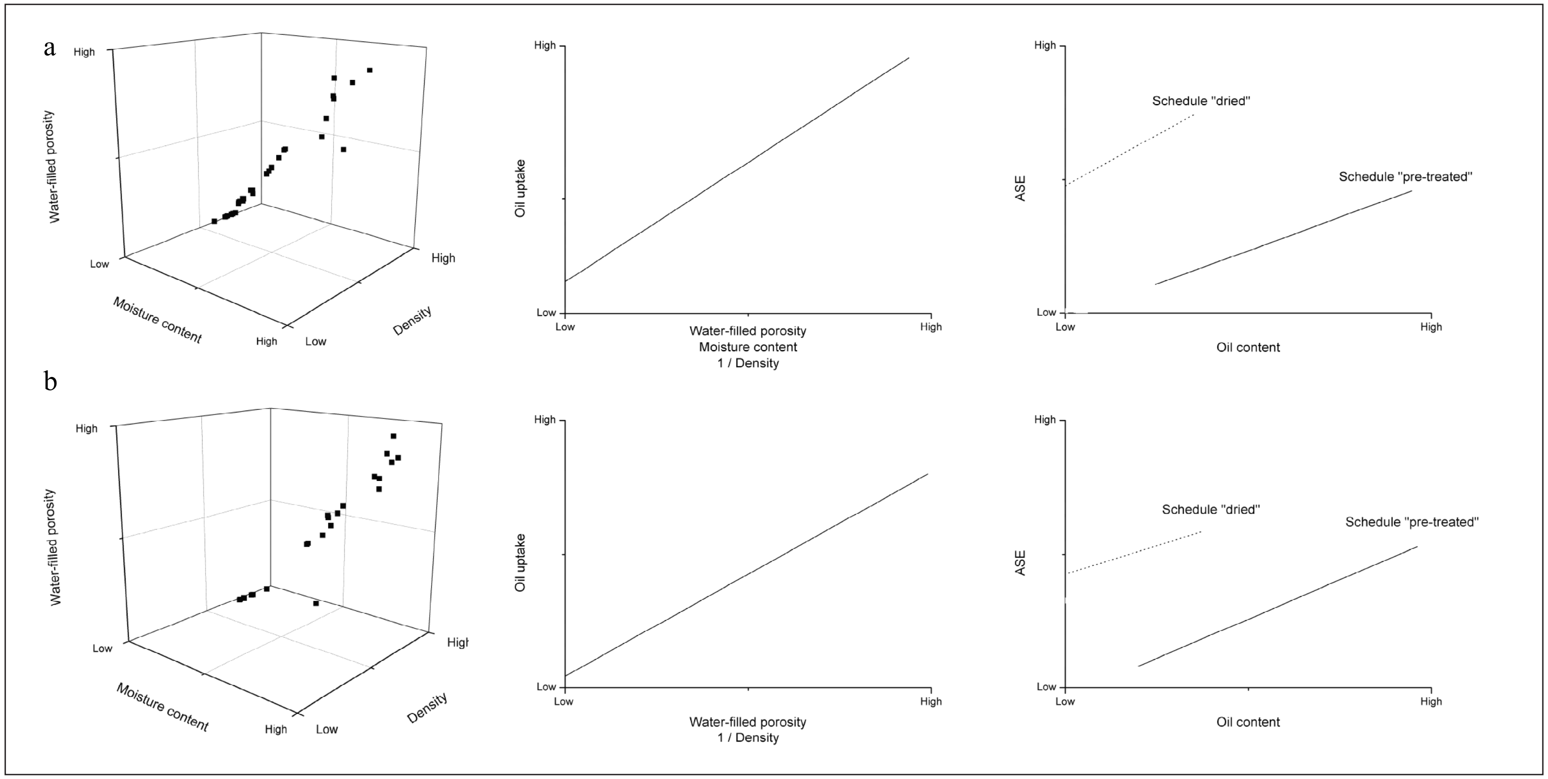
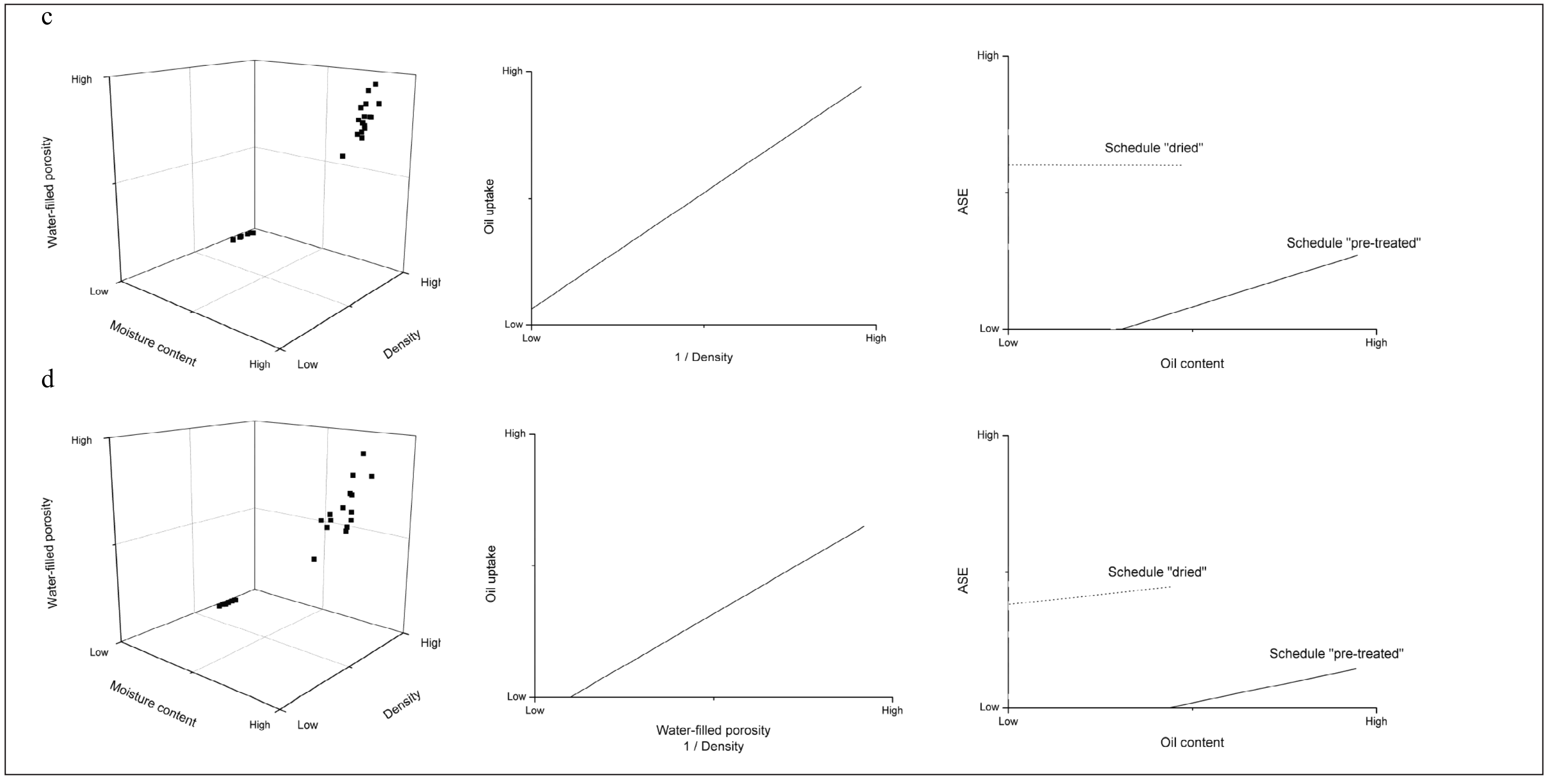


Fig. 1b. Schematic illustration to show guidelines for processing chains; mature heartwood (c) and juvenile heartwood (d), respectively. Diagrams, from left to right, presents relations between properties in the tissue type (i.e. the raw material), oil uptake in relation to important tissue properties within a given impregnation schedule and ASE within a certain climate and time span (i.e., one important material property) as a result from two different treatments made prior to impregnation with an hydrophobic oil. Figure is based on papers II and IV.



and it was possible to predict oil uptake levels after impregnation in Norway spruce wood treated in different pre-treatments (Paper II). Generally, there is a need for new evaluation methods for determining durability after treatment with non-toxic substances (Råberg *et al., op. cit.*). However, methods developed for this purpose must be accurate and allow the effects of silvicultural treatments to be assessed, and adjusted if necessary. If they fail to give valuable information for silvicultural purposes, they will miss opportunities to support the development of new products by the industry.

The results of the presented studies imply that it is possible to produce wood-based materials with homogenous properties and to develop evaluation methods that allow rapid and accurate measurements of relevant material properties. It should therefore also be possible to develop effective new product development procedures, for both wood and wood-based materials within forest industries; i.e. NPD procedures according to established practices in other sectors, as discussed in the introduction to this thesis.

References

Abdurrohim, S., Nurjaman, J. & Hadiane, A. 2003. Preservation of Tisuk (*Hibiscus macrophyllus Roxb.)* and Sukun (*Artocarpus horridus Jarret.)* wood with CCB and boron preservatives by full-cell processes. *Buletin Penelitian Hasil Hutan 21(1),* 83-90.

Alfredsen, G., Eikenes, M., Militz, H. & Solheim, H. 2004. Screening of Chitosan against wood-deteriorating fungi. *Scandinavian Journal of Forest Research 19(5),* 4-13.

Anon. 1994. Durability of wood and woodbased products – Natural durability of solid wood – Part 2: Guide to natural durability and treatability of selected species of importance in Europe. European standard 350-2., *European Committee for Standardization,* 1-35.

Anon. 2003. URL. http://www.patentstorm.us/patents/6610870-fulltext.html (2006-09-13).

Axrup, L., Markides, K. & Nilsson, T. 2000. Using miniature diode array NIR spectrometers for analysing wood chips and bark samples in motion. *Journal of Chemometrics 14(5-6),* 561-572.

Bailey, P.J. & Preston, R.D. 1969. Some aspects of softwood permeability. *Holzforschung 23*, 113-120.

Baines, E.F. & Saur, J.M. 1985. Preservative treatment of Spruce and other refractory wood. *Record of the 1974 Annual Convention of the British Wood Preserving Association Cambridge 25-28 June 1974.* American Wood-Preservers Association.

Banks, W.B. 1970. Some factors affecting the permeability of Scots pine and Norway spruce. *Journal of the Institute of Wood Science 25,* 10-17.

Barton, L., McLay, C.D.A., Schipper, L.A. & Smith, C.T. 1999. Denitrification rates in a wastewater-irrigated forest soil in New Zealand. *Journal of Environmental Quality 28(6)*, 2008-2014.

Baysal, E. & Yalinkilic, M.K. 2005. A new boron impregnation technique of wood by vapour boron of boron acid to reduce leaching boron from wood. *Wood Science and Technology 39(3),* 187-198.

Bolton, A.J. 1988. A reexamination of some deviations from Darcy's Law in coniferous wood. *Wood Science and Technology 22(4),* 311-322.

Boutelje, J. 1983. Preservative Treatment of Spruce – Possibilities and Requirements. Wood Technology Report 22. *Svenska Träforskningsinstitutet STFI-meddelande serie 807.* 1-53 (in Swedish).

Bowyer, J.L. 2000. Wood science in a changing world – Where are we headed?. *Wood Science and Technology 34,* 175-181.

Bramhall, G. 1971. The validity of Darcy's Law in the axial penetration of wood. *Wood Science and Technology 7,* 319-322.

Carbonell, P., Escuredo, A.I.R. & Aleman, J.L.M. 2004. Technology newness and impact of go/ no go criteria on new product success. *Marketing Letters 15 (2-3),* 81-97.

Comstock, G.L. 1965. Longitudinal permeability of green Eastern Hemlock. *Forest Products Journal 15(10), 441*-449.

Dhamodaran, T.K. & Gnanaharan, R. 2006. An economical treatment schedule for boron impregnation of Eucalyptus grandis wood: commercial trial of partially dried material. *Holz als Roh- und Werkstoff 64(1),* 80-81.

Diaz, B.G., Luna, M.L., Keil, G.D., Otano, M.E. & Peri, P.L. 2002. Behaviour of Populus nigra cv Italica timber in creosote and CCA preservation treatments. *Ivestigacion Agraria Sistemas y Recursos Forestales 11(2),* 325-338.

Dinwoodie, J.M. 2000 *Timber: Its nature and behaviour*, 2nd edn. E&FN SPON. 257 pp.

Eikenes, M., Hietala, A.M., Alfredsen, G., Fossdal, C.G. & Solheim, H. 2005a. Comparison of quantitative real-time PCR, chitin and ergosterol assays for monitoring colonization of *Trametes versicolor* in birch wood. *Holzforschung 59,* 568-573.

Eikenes, M., Alfredsen, G., Christensen, B.E., Militz, H. & Solheim, H. 2005b. Comparison of chitosans with different molecular weights as possible wood preservatives. *Journal of Wood Science 51,* 387-394.

Eriksson, D. 2005. *Scots pine wood: an anatomical structure in the tree and an engineering material in industry.* Swedish University of Agricultural Sciences, Department of Silviculture, reports, no. 65. Licentiate dissertation.

Flate, PO. & Kucera, B. 1999. *Virkesegenskaper till mellomeuropeiska og norske granprovenienser plantet i östfold.* NISK rapport fra skogsforskningen 1/99 (in Norwegian)

Frühwald, E. 2006. Improvement of shape stability by high-temperature treatment of Norway spruce effects of drying at 120 °C with and without restraint on twist. *Holz als Roh- und Werkstoff 64,* 24-29.

Furuno, T., Lin, L., Katoh, S. & Lin, L. 2003. Leachability, decay and termite resistance of wood treated with metaborates. *Journal of Wood Science 49(4),* 344-348.

Gindl, W., Teischinger, A., Schwanninger, M. & Hinterstoisser, B. 2001. The relationship between near infrared spectra of radial wood surfaces and wood mechanical properties. *Journal of Near Infrared Spectroscopy 9(4),* 255-261.

Hansmann, C., Gindl, W., Wimmer, R. & Teischinger, A. 2002. Permeability of wood – a review. *Wood Research Drevarsky Vyskum 47(4),* 1-16.

Hart, S., Erik, H., Tzokas N. & Commandeur, H. 2003. Industrial Companies' Evaluation criteria in new product development gates. *Journal of Product Innovation Management 20(1),* 22-36.

Hauksson, J.B., Bergqvist, G., Bergsten, U., Sjöström, M. & Edlund, U. 2001. Prediction of basic wood properties for Norway spruce, interpretation of near infrared spectroscopy data using partial least squares regression. *Wood Science and Technology 35(6),* 475-485.

Hodge, G.R. & Woodbridge, W.C. 2004. Use of near infrared spectroscopy to predict lignin content in tropical and sub-tropical pines. *Journal of Near Infrared Spectroscopy 12,* 381-390.

Hoffmeyer, P. & Pederson, J. 1995. Evaluation of density and strength of Norway spruce wood by near infrared reflectance spectroscopy. *Hols als Roh- und Werkstoff 53*, 165-170.

Hovgard, A. & Hansen., E. 2004. Innovativeness in the forest products industry. *Forest Products Journal 54(1),* 26-33.

Humar, M., Bokan, M., Amartey, S.A., Sentjurc, M., Kalan, P. & Pohleven, F. 2004. Fungal bioremediation of copper, chromium and boron treated wood as studied by electron paramagnetic resonance. *International Biodeterioration and Biodegredation 53(1),* 25-32.

Hyvönen, A., Piltonen, P. & Niinimäki, J. 2006. Tall oil/water-emulsions as water repellents for Scots pine sapwood. *Holz als Roh- und Werkstoff 64*, 68-73.

Johansson, K. & Persson, A. 1997. *Wood properties of naturally regenerated and planted Norway spruce (Picea abies* L. Karst.*) on a productive site in southwestern Sweden.* Danish Forest and Landscape Research.

Kakitani, T., Hata, T. & Imamura, Y. 2003. Evaluation of mobility of metal elements in CCA-treated wood for efficient recycling and safe disposal. *Wood Research Kyoto 90,* 21-22.

Kartal, S.N. & Imamura, Y. 2003. Chemical and biological remediation of CCA-treated waste wood. *Wood Research Kyoto 90,* 111-115.

Kifetew, G., Thuvander, F., Berglund, L. & Lindberg, H. 1998. The effect of drying on wood fracture surfaces from specimens loaded in wet condition. *Wood Science and Technology 32(2),* 83-94.

Kelly, S.S., Rials, T.G., Snell, R., Groom, L.H. & Sluiter, A. 2004. Use of near infared spectroscopy to measure the chemical and mechanical properties of solid wood. *Wood Science and Technology 38,* 257-276.

Klem, G.G. 1957. *Kvalitetsundersökelser av norsk och tysk gran.* Meddelser fra det Norske Skogsforsoksvesen.

Kollman, F.F.P. & Coté, W.A. 1984. *Principles of Wood Science and Technology, Vol. 1 Solid Wood,* Springer Verlag Berlin. 535 pp.

Kyrkjeeide, P.A. 1990. A wood quality study of suppressed, intermediate and dominant trees of plantation grown *Picea abies*. *Forest Products Laboratory. USDA Forest Service.* One Gifford Pinchot Drive, Madison, WI 53705-2398. USA.

Laming, P.B. & Welle, B.J.H. 1971. Anomalous tangential pitting in *Picea abies* (L.) Karst. (European spruce). *IAWA Bullentin 4,* 3-10.

Lee, A., W.C., Chen. G., Tainter, F.H. & Chen, G, 2001. Comparative treatability of Moso bamboo and Southern pine with CCA preservative using a commercial schedule. *Bioresource Technology 77(1), 87*-88.

Li, J.Z., Furuno, T. & Katoh, S. 2001. Preparation and properties of acetylated and propionylated wood-silicate composites. *Holzforschung 55:1,* 93-96.

Liese, W. & Bauch, J. 1967. On the anatomical causes of the refractory behaviour of spruce and Douglas fir. *Journal of the Institute of Wood Science 1,* 3-14.

Malmquist, L., Esping, B. & Nilsson, K. 1988. Drying of preservative-treated wood. Investigations of cracks, moisture content, impact strength and efficiency of treatment. Drying of preservative-treated wood. *Work and activities initiated by the Swedish Wood Preservation Institute 16, 1977 – 1987.* 43 – 64.

Mathias, L.J., Lee, S., Wright, J.R. & Warren, S.C. 1991. Improvement of wood properties by impregnation with multifunctional monomers. *Journal of Applied Polymer Sciences 42(1),* 55-67.

Megnis, M., Olsson, T., Varna, J. & Lindberg, H. 2002. Mechanical performance of linseed oil impregnated pine as correlated to the take-up level. *Wood Science and Technology 36(1),* 1-18.

Militz, H. 1993. Enzymatic treatments of Norway spruce roundwood and sawn timber to improve their treatibility with preservatives. *Holz als Roh und Werkstoff 51(5),* 339-346.

Militz, H. & Homan, W.J. 1993. Pretreatment of Norway spruce wood with chemicals to improve permeability. Trials with sawnwood; preservative penetrability and strength properties. *Holz als Roh- und Werkstoff 51(1),* 14-20.

Moore, G.R., Garges, L.D., Blankenhorn, P.R. & Kline, D.E. 1984. Effect of epoxy impregnation on the MOE and MOR of intact and failed yellow-poplar (*Liriodendron tulipifera* L.) sapwood beams. *Wood and Fiber Science 16(4),* 508-517.

Nylinder, P. 1953. *Variations in density of planted spruce.* Report of the Forest Research Institute of Sweden 43. 42 pp.

Nylinder, P. & Hägglund, E. 1954. *The influence of stand and tree properties on yield and quality of sulphite pulp of Swedish spruce (Picea excelsa).* Statens Skogsforskningsinstitut, Stockholm. Meddelande 44(11) (in Swedish).

Nyrén, V. & Back, E. 1960. Characteristics of parencymateous cells and tracheidal ray cells in *Picea abies* Karst. *Svensk papperstidning och Svensk pappersförädlingstidskrift 63(16),* 501-509.

Obataya, E. 2005. Reversible volumetric changes of acetylated wood with after-treatments. *Wood Science and Technology 39*, 472-483.

Olesen, P.O. 1977. Resistance of some common Danish timbers to pressure impregnation (*Picea abies, Picea sitchensis, Abies alba, Abies grandis*). *Holzforshung 31,* 179–184.

Olsson, T., Megnis, M., Varna, J. & Lindberg, H. 2001. Measurement of the uptake of linseed oil in pine by the use of an X-ray microdensitometry technique. *Journal of Wood Science. 47(4),* 275 – 281.

Pape, R. 2001. *Differences in wood properties of Norway spruce between northern and southern Sweden: literature review within the project "The future sawmill for spruce timber".* Rapport Sveriges Lantbruksuniversitet, institutionen för skogshushållning, 15 (in Swedish).

Persson, B. & Persson A. 1997. Variation in stem properties in a IUFRO 1964/1968 Picea abies provenance experiment in southern Sweden. *Silvae Genetica 46(2/3),* 94-101.

Richardsson, B. A. 1993. *Wood Preservation,* second edition. E&FN SPON.

Rowell, R.M. 1996. Physical and mechanical properties of chemically modified wood, Chemical modification of lignocellulosic materials. *USDA Forest Service and University of Wisconsin Madison,* Wisconsin.

Råberg, U., Edlund, M-L., Terziev, N. & Land, C.J. 2005. Testing and evaluation of natural durability of wood in above ground conditions in Europe – an overview. *Journal of Wood Science 51,* 429-440.

Schimleck, L.R., Evans, R. & Ilic, J. 2001. Estimation of Eucalyptus delegatensis wood properties by near infrared spectroscopy. *Canadian Journal of Forest Research 31,* 1671-1675.

Schimleck, L.R., Evans, R. & Matheson, A.C. 2002. Estimation of Pinus radiate D. Don clear wood properties by near-infrared spectroscopy. *Journal of Wood Science 48(2),* 132-137.

Schmidt, E.L. & Westberg, C. 2001. Heartwood temperatures of Red pine pole stock during pentachlorophenol pressure treatment and presence of decay fungi prior to treatment. *Forest Products Journal 51*, 11-12, 41-44.

Schneider, P.F., Morrell, J.J. & Levien, K.L. 2005. Internal pressure development during supercritical fluid impregnation of wood. *Wood and Fiber Science 37(3),* 413-423.

Siau, J.F. 1972. The effects of specimen length and impregnation time upon the retention of oils in wood. *Wood Science 4,* 163-170.

Siau, J.F. 1984. *Transport Processes in Wood.* Springer-Verlag.

Story, V., Saker, J. & Smith, G. 1998. *Characteristics of a successful new product development process for European Automotive Components Suppliers: A quantitative study.* European Marketing Academy Conference, Stockholm.

Ståhl, E.G. & Karlmats, U. 1995. *Yield, wood properties and timber harvest at establishment of seed tree and shelterwood systems.* Studia Forestalia Suecia 197. 15 pp.

Thumm, A. & Meder, R. 2001. Stiffness prediction of radiate pine clearwood test pieces using near infrared spectroscopy. *Journal of Near Infrared Spectroscopy 9(3),* 117-122.

Thuvander, F., Kifetew, G. & Berglund, L.A. 2002. Modelling of cell wall stresses in wood. *Wood Science and Technology 36(3),* 241-254.

Thygesen, L.G. & Lundqvist, S.O. 2000a. NIR measurement of moisture content in wood under unstable temperature conditions. Part 1. Thermal effects in near infrared spectra of wood. *Journal of Near Infrared Spectroscopy 8(3),* 183-189.

Thygesen, L.G. & Lundqvist, S.O. 2000b. NIR measurement of moisture content in wood under unstable temperature conditions. Part 2. Handling temperature fluctuations. *Journal of Near Infrared Spectroscopy 8(3),* 191-199.

Tzokas, N., Hultink, E.J. & Hart, S. 2004. Navigating the new product development process. *Industrial Marketing Management 33,* 619-626.

Usta, I. & Guray, A. 2001. The effect of wood specimen length on the proportional saturation of preservative fluid in Sitka spruce (*Picea sitchensis* (Bong.) Carr.) and Corsican pine (*Pinus nigra var. maritima*). *Turkish Journal of Agriculture and Forestry 25(1),* 1-4.

Van Eckeveld, A., Homan, W.J. & Militz, H. 2001. Increasing the water repellency of Scots pine sapwood by impregnation with undiluted linseed oil, wood oil, coccos oil and tall oil. *Holzforschung und Holzverweltung 53(6),* 113 -115.

Vestøl, G.I. & Høibo, O.A. 2001. Prediction of knot diameter in *Picea abies* (L.) Karst. *Holz als Roh- und Werkstoff 59 (1/2),* 129-136.

Vinden, P. 1984. The effect of raw-material variables on preservative treatment of wood by diffusion processes. *Journal of the Institute of Wood Science 10,* 31-41.

Wardrop, A.B. & Davies, G.W. 1961. Morphological factors relating to the penetration of liquids into wood. *Holzforschung 15(5),* 129–141.

Yamagushi, H. 2005. Silicic acid/boric acid complexes as ecologically friendly wood preservatives. *Forest Products Journal 55(1),* 88-92.

Acknowledgements

I wish to express my sincere gratitude to my supervisors, professors Urban Bergsten and Henrik Lindberg, for their valuable guidance and help during my efforts to better learn, understand and practice science. I also want to thank the Kempe Foundation and Norrskog Research Foundation for financial support; furthermore, I want to thank Linotech AB for providing oil and impregnation equipment. Special thanks to Ulla Nylander, Hans Göran Nilsson, Gunnar Karlsson and all the other at SLU that has helped me with practical issues during the project. Last but not least, I want to thank my wife Kristina, my family and all friends that have supported me during these years.