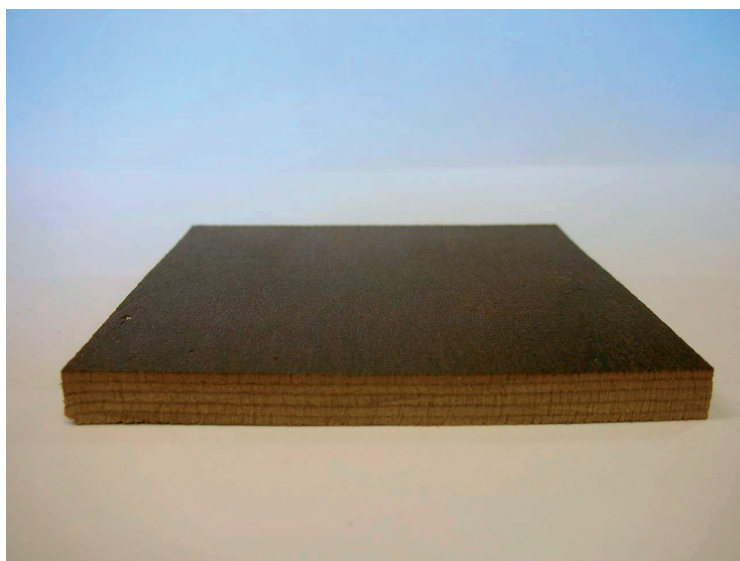


Bonding Veneers Using Only Heat and Pressure

Focus on Bending and Shear Strength



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LICENTIATE THESIS

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ABSTRACT

A compact laminated board can be obtained by pressing layers of veneers with no other material or substance placed between them. The process does not require the use of steam pretreatment, surface activation methods, a gastight press, friction or adhesives. It strictly involves the heat and the pressure induced by the press in the veneers.

The bonding quality of products manufactured under different pressing parameters is revealed by shear-strength results. The ability of the boards to resist breakage when bent is reflected by their bending-strength results.

The factors influencing these two responses are divided into materials and equipment factors. Wood layer thickness, number of layers, veneer type, initial moisture content, grain direction and species are the material properties referred to on this work.

The levels of temperature, pressure and time leading to the highest bending-strength values when material factors are held constant are investigated. The objective of optimizing the process is reached using response surface methodology for modelling and analysis. The parameter interactions are found to be significant.

Photography, scanning, X-ray densitometry, light microscopy and scanning electron microscopy (SEM) are the methods used to visually analyse the final product. Densification and darkening are two of the effects observed. The intensity of structural changes varies depending on the pressing parameters used.

Keywords: wood, high temperature, pressure, veneer, self-bonding, beech, optimization, bending strength, shear strength, microscopy

PREFACE

The work presented in this thesis was carried out at Luleå University of Technology, Division of Wood Science and Technology in Skellefteå. The project was financed by Luleå University of Technology. Substantial financial support offered by Swedbank through Sparbankstifstelsen is gratefully acknowledged, together with Innovationbron funding and Nordea resestipendium.

This thesis was a product of a pleasant working environment rather than a product of my own. It wasn't always easy to be 3000 km away from my home, to create a new home, to live in a society with so different rules. But this is part of the challenge of studying and I still feel a student. I learn the most when facing new situations and destiny keeps on offering me this chance.

It's great to work in a place where people have very profound knowledge in so different areas and they are ready to share it with you anytime you ask for help!

Margosha, I terribly enjoy learning from you, from wood anatomy to surrounding nature! Svenerik, thank you for the influence you had in my career. I'm honoured we worked together. Anna P, Lena, Margit you are smart and strong, you've been a strong example for me! Thank you Janne Nyström, Mats Ekevad, Jonas Danvind and especially Olov Karlsson! You had the patience to describe for me over and over every chemical reaction and to listen to me everytime I ran up to your office with a new article saying : "this is the one, this makes me understand all about self-bonding chemistry!" It was never the case. That is why I thank Ulla Westeramark for her precious hints.

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I'm glad I got to work with you lately, I admire your "toughness", never comprising, working hard and seriously, "the Västerbotten father figure" in my professional life.

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Finally I thank Lucian for being more understanding than ever these last months.

Skellefteå, in November 2008

LIST OF PAPERS

Paper I Cristescu C (2006)

Bonding of laminated veneers with heat and pressure only

Proceedings of the 2nd international conference on environmentally-compatible forest products “Ecowood” Porto, Portugal

Paper II Cristescu C, Lindgren O, Vännman K, Grönlund A (2008)

Autoadhesion of Beech (*Fagus sylvatica* L.) Veneers – Pressing Parameters and Bending Strength

Submitted to Wood Science and Technology

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1 INTRODUCTION

Pressing thin layers of wood at temperatures above 200°C for a sufficiently long time can form strong bonds, not only physical bonds, but chemical bonds as well. The same phenomenon was found for thicker pieces of wood—which require an extremely long pressing time—and for wooden flakes and chips pressed at high temperature to form high-density panels. This thesis will present only results on veneer bonding using heat and pressure.

The technology described in the thesis differentiates itself from the other wood bonding technologies through the following characteristics: no adhesives, no friction, no steaming before pressing and no surface activation treatments are used. Producing a wooden laminated board with heat and pressure refers to simply pressing veneers in a hot plate press.

The product realization is analysed through the relations found between the effect produced by changing the level of some input factors to the output responses. More exactly, the bonding process is screened and optimized. The factors influences are quantified in relation to their effect on two mechanical properties: shear strength and bending strength. These mechanical properties are strongly connected to the bonding quality, so their ability of product characterization is high.

The input variables were grouped into raw material properties and pressing parameters because they relate to two different entities: one characterizes the material to be used and the other characterizes the machine.

1.1 OBJECTIVES

The objective of this thesis was to study how the process parameters affect the properties of laminated boards manufactured using heat and pressure only.

It is not an easy task, since the self-bonding process is a complex chemicophysical phenomenon involving a balance of energies, a balance of chemical reactions, of compounds transformed into other compounds, of physical and chemical bonds being broken and others being newly formed. Particular attention was given to process optimizations. Thus, finding the most suitable pressing parameters (considered as factors) with regard to bending strength (as response) can be considered the main purpose of this thesis.

1.2 LIMITATIONS

The product obtained is new and in many ways different from any other wooden engineered material created and produced before. The differences consist not only in the absence of glue or that the product is denser than, for example, plywood or Laminated Veneer Lumber. There is a difference all the way up to its final and most detailed chemical structure, since the temperature during pressing is higher than in any other technology for producing a composite product. Obviously, there is no such a document as an officially accepted standard norm to test and certify this new product. Therefore, the European Standard EN 310 and EN 314, created for wood-based panel, plywood and Laminated Veneer Lumber, were used in all experiments.

The size of the specimens studied is relatively small (135 x 135 mm). The product properties described throughout the thesis refer to these dimensions. Whether or not to extend the laboratory-sized sample results to larger-sized boards remains a question to be answered after a detailed comparison between their strength values.

The discussions are based on experiments made with a small laboratory press (type Fjellman 2023). The press had a plate area of 140x140 mm and the test boards produced had a thickness between 5 and 8 mm. Therefore it was

impossible to fulfil the length/thickness ratio according to EN 310 in the bending test.

The thesis focuses on the lamination resulting from hot pressing on beech veneers, and most of the experiments results and discussions concern beech veneers. Studies on other other species are rather limited.

The pressing-parameter levels as well as the response values (mechanical properties) characterize the use of veneer from beech (*Fagus sylvatica* L.). Other species require different pressing parameters, and the products obtained have different properties.

The shear- and bending-strength tests results discussed in Papers I and II are connected to the material characteristics used in those experiments. A different moisture content or thickness might lead to a different bending strength. The process optimization part covered only 2.2 mm thick rotary-cut beech veneer, with a moisture content of 9%.

The tests involving raw material influence had only an exploratory character. The steps between some of the levels discussed (for example moisture content) should have been smaller and more symmetrical, but this was not the main objective of this thesis.

1.3 THESIS STRUCTURE

The structure of the thesis is based on the scheme in Fig 1.1.

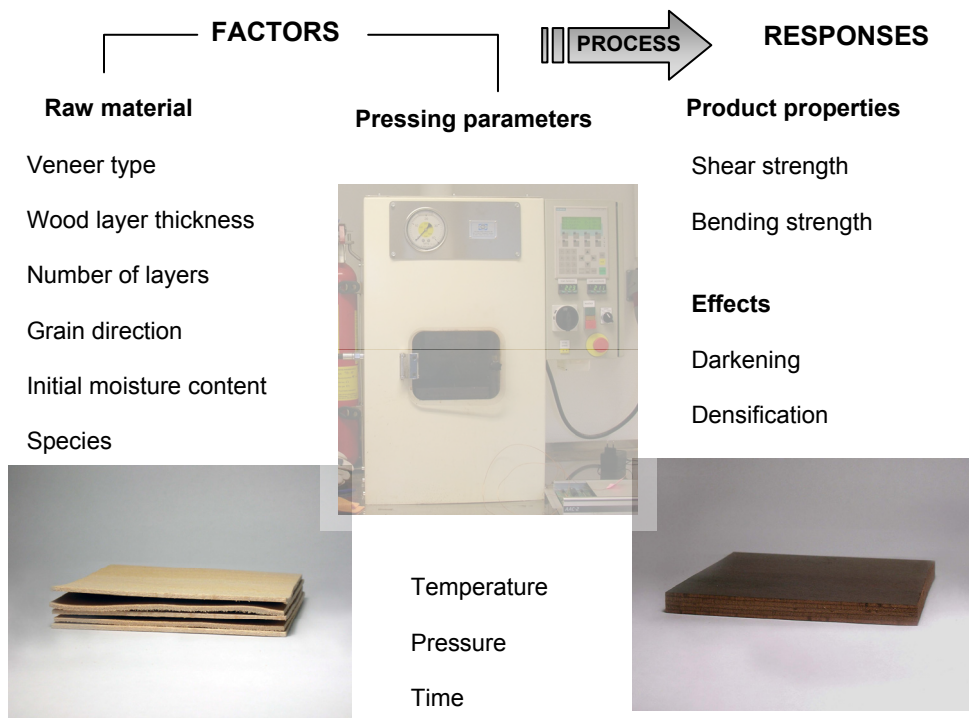


Fig 1.1 Thesis structure scheme

The outline follows the steps of the bonding process. First, the veneers are arranged in a package. Their properties represent one group of factors. The package is pressed using certain parameters; the levels chosen depend on the raw material, but at the same time influence the responses. In the end, a product is obtained, if the factors from the second column matched those of the first column. The quality of the product can be evaluated by measuring its strength. Finding the maximum value leads to process optimization.

Chapter 2 summarizes the achievements in the field of wood autoadhesion. It emphasizes some characteristics of wood structure essential for understanding the bonding process. It also refers to some theoretical assumptions regarding the reasons bonding takes place.

Chapter 3 looks at the first column in Fig. 1.1 and describes more or less the initial screening studies of different parameters, focusing on the importance of the material in the process. Part of these results have not been published before.

Chapter 4 focuses on optimizing the pressing parameter combination for beech veneers. It describes in detail the influence of parameters interactions and reveals what temperature, pressure and time should be used to obtain a product with high bending strength. This subject was discussed in Paper II.

Chapter 5 refers to the product visualization from macroscopic to microscopic scale and is based on the information presented in Paper I.

The conclusions (as an overview) are related to future work, so chapter 6 presents the connection between past and future.

BONDING VENEERS USING ONLY HEAT AND PRESSURE

2 BACKGROUND

Bonding veneers in a simple hot-plate press using no other means than high temperature and pressure is a new process (Cristescu *et al.*2007). Nevertheless, there are similarities with other methods. A review of these methods is presented first, and a brief description of the raw material anatomy follows. Finally, possible theoretical explanations of the bonding phenomenon are discussed in connection with wood behaviour when subjected to high heat and pressure.

2.1 BONDING VENEERS WITH HEAT AND PRESSURE—STATE OF THE ART

A review of the most closely related methods follows the scheme in Fig. 2.1

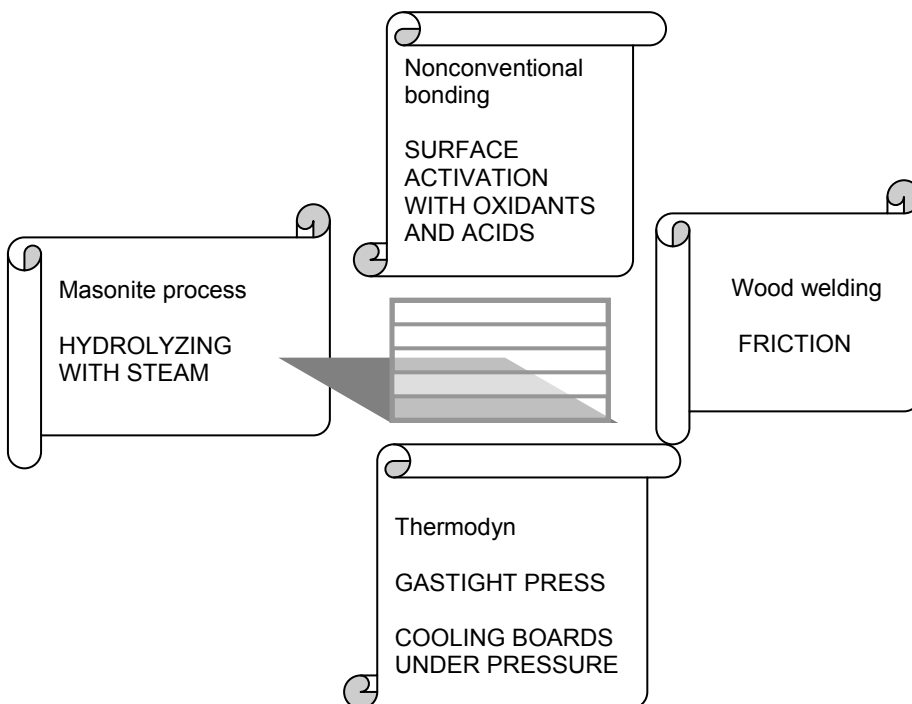


Fig. 2.1 Techniques related to bonding veneers using only heat and pressure together with their characteristic process steps.

It will be shown that although all the methods above are related to the technology discussed throughout the thesis, each of them differs from using only heat and pressure through a certain process step.

The capacity of wood to self-bond was discovered by William H. Mason in 1924. He developed the idea of pressing steam-exploded wooden chips. Steam was the key process in his technology. In his first patent, Mason (1925) reports that under proper conditions of heat, steam and pressure, lignin supplies the cementitious or welding effect, particularly towards the surfaces when the product is dried. Later on, his company tried to apply the hardboard manufacturing technology to plywood production (Boehm 1945). The step of hydrolyzing pretreatment was transferred to the veneer bonding method as well, since Mason was convinced that lignin provided the bonding material, and lignin needs to be activated by steam. In the end of the process, cooling while under pressure was considered compulsory. Unlike the hardboard made out of chips, the plywood product was not industrialized, and no reference was found of mass production according to the Masonite method.

Many other technologies for producing pressed wooden composites started to be experimented from 1930. One of them—strongly connected to the method described in this thesis—was developed in a cement production company. Thus, the idea was supported by experiences from cement manufacturing. The authors (Irvine and Frederick 1936) describe a method of self-bonding wood particles in a sealed, gastight press, since the vapours and gases formed during the first stage of pressing were considered responsible for the bonding phenomenon. A few years later, this idea of gastight moulding was brought to Europe to be industrialized in Germany under the name of the Thermodyn process (Kollmann 1975). Like Irvine and Frederick, the authors (Runkel and Jost 1948) thought that the volatile products (condensable and permanent gases) formed during a first stage of gastight pressing would further hydrolyze the wood constituents, resulting in a decomposition of the lignin-carbohydrate compound. Beech veneer is mentioned as a suitable material. This method was not successfully industrialized.

Kollmann (1975) explains the impediments to the industrialization of the Thermodyn process: a completely gastight construction, the need for recooling after hot pressing, and the utilization of natural binding constituents of the wood.

Starting with the 1970s, revolutionary technologies involving the wood autoadhesion principle gained interest. They were named “nonconventional bonding technologies”. According to Zavarin (1984), this denomination includes many different methods of bonding, radically different from the conventional phenol-formaldehyde and urea-formaldehyde and related methods. Zavarin states that the progress of these methods is handicapped by insufficient knowledge of the chemical composition of wood surfaces as well as of the chemical process involved in bonding. Usually this group of methods is based on surface activation (acid or oxidant activators) prior to pressing. One recent successful example of using such a method in board manufacturing is bonding by oxidative treatment (Westermarck and Karlsson 2005).

With regard to solid wood joining, probably the most exciting technology is frictional wood welding. The method was discovered and patented in Germany in 1996 (Suthoff *et al.* 1996). The Swiss Federal Institute of Technology in Lausanne (EPFL) started to analyse and develop this method, focusing on circular welding (Gliniortz *et al.* 2001). At the same time, in the Higher Technical Schools of Wood, Biel, Switzerland, another team began studying the linear welding movement (Gfeller 2003) together with ENSTIB-LERMAB, University of Nancy 1, Epinal, France. Wooden dowel welding based on rotational frictional movement is studied by ENSTIB-LERMAB, University of Nancy 1, France (Kanazawa *et al.* 2005). The three methods mentioned involve the same frictional process, but the opinions differ considerably regarding the temperatures reached during the bonding process. One team reports a maximum interfacial temperature during welding of 440°C (Stamm 2005), while another team reports that the average of the maximum temperatures in the centre of the wood welding line is 200°C (Ganne-Chedeville *et al.* 2005). The third team states that temperature during rotation dowel-welding

never goes above 183°C (Omrani *et al.* 2008). Nevertheless, there is a common conclusion: beech samples give the best results.

2.2 MICROSCOPIC AND CHEMICAL STRUCTURE OF THE RAW MATERIAL

To understand what causes two pieces of veneer to adhere without an intermediate product, it is important to look at the raw material structure from the macroscopic to the microscopic level. A view of a beech (*Fagus sylvatica* L) veneer sample of 2-mm thickness is presented in Fig. 2.2. The images were taken with an Olympus B061 microscope.

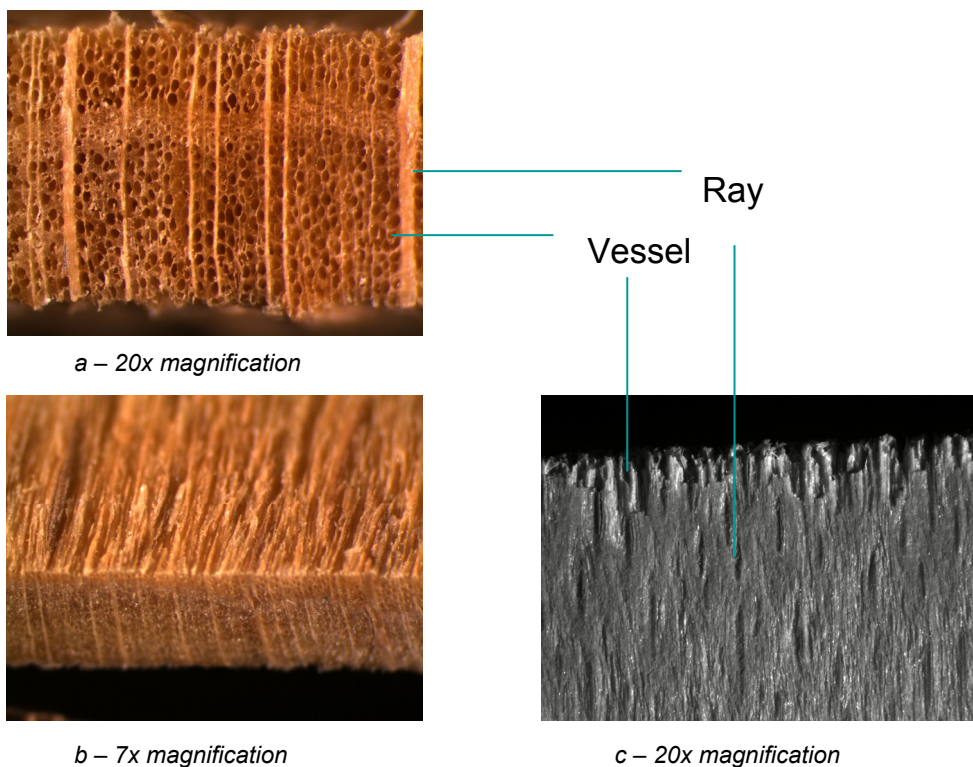


Fig 2.2 Microscopic view of a beech veneer: a, cross-sectional view; b, sample tilted 45° with tangential section on top; c, tangential view.

Fig. 2.2 a shows the even distribution of crosscut vessels (also called pores) along the annual ring. These pores are all similar in size. Beech microstructure is relatively homogenous. Vessels are composed of thin-walled and rather short (0.3–0.6 mm) and wide (30–130 μm) elements that are placed one on top of the other to form a long tube (Sjöström 1981). Vessel cell walls seem to be the main participants in the bonding process (see Fig. 2.2 b and c).

By contrast, the cell walls of the rays (parenchyma cells) structured perpendicularly to the contact surface, do not seem to actually participate in the bonding process. Rays have thick and very strong walls that help them to keep themselves together during the pressing process, despite such a high temperature and pressure treatment. Their role might be the one of channels through which water vapours and gases formed during thermal decomposition are transported out to the veneer surface.

On the molecular level, things are complex, since it is difficult to separate and describe the contribution of a certain type of polymer to a process. It is well known that the main chemical components of wood are cellulose, the hemicelluloses and lignin. When studying the other bonding mechanisms presented in this chapter, researchers often tried to find the explanation in changes of these polymers only. One of the reasons for which lignin was generally considered responsible was the simple fact that defibration (the separation of wood and other plant material into fibres or fibre bundles by mechanical or/and chemical means) involved removing the lignin. However, in a defibration process, a large part of the hemicelluloses is also removed and, more important, the bonds between these polymer groups are attacked. A representation of the bonds between the main wood chemical components is presented in Fig. 2.3.

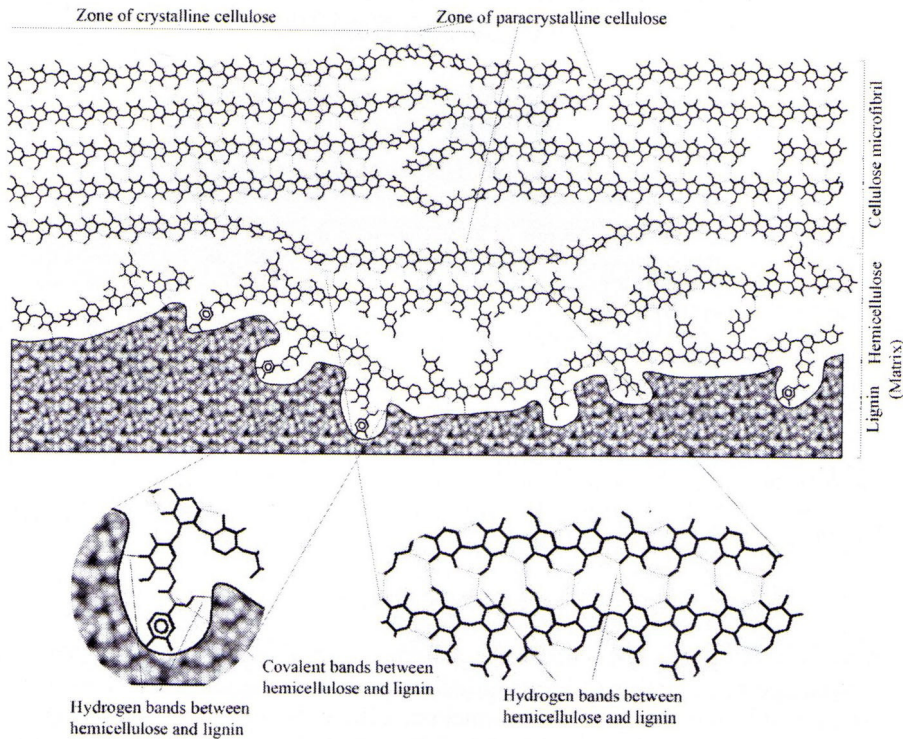


Fig. 2.3 Composite molecular model of a longitudinal section from a dried wood cell wall (Heger 2004).

A laminated product such as the one under discussion here is not flexible (as will be shown) and in many respects does not behave like a solid wood piece of the same size. This means that it under no circumstances regains the same chemical structure. The product has a new structure in which, probably, new bonds and new molecules were formed, leading to different characteristics.

When analysing the chemical changes caused by heat and pressure, research needs to be done on the intramolecular as well as the intermolecular bonds. A good example is the study by Heger (2004) that emphasizes the importance of chemical bonds between the three wood polymers during the thermo-hydro-mechanical processes.

2.3 THEORETICAL EXPLANATIONS OF THE BONDING PHENOMENON

Any autoadhesion phenomenon involving materials other than wood might constitute a path toward the answer to the question “What makes it self-bond?” One example is the stickiness phenomenon encountered during dehydration in the food industry. Roos and Alves-Filho (2005) brought into discussion the stickiness region existing between a glass transition and a flow region on a temperature-water chart of a dehydration process (see Fig. 2.4).

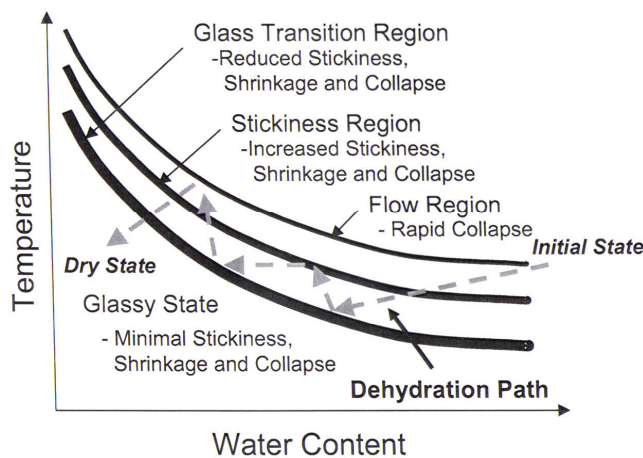


Fig 2.4 Temperature-water content control in food dehydration, reducing flow above the glass transition but allowing free water removal (after Karel et al. 1994).

They also report that some sugar-containing materials are extremely difficult to dehydrate, as the solids tend to adhere on drier surfaces and cake inside other manufacturing and processing equipment (similar to the adhesion of wood chips to the press plate during production of composite boards).

Wood might act in a similar manner (passing a stickiness region) when subjected to a high-temperature and high-pressure treatment. It is very difficult to study wood phase transition since it is formed by several component, each behaving in a

different manner. Unlike food, metal or thermoplastic materials, wood does not undergo thermal flow unless chemically treated (Morita and Sakata 1986). It deforms, it degrades, but wood as a complex material does not pass from solid to liquid state. Solid wood lacks plasticity, and it cannot be softened sufficiently for moulding, melting or dissolving. (Shiraishi 2001).

Nevertheless, wood polymers can undergo a second phase transition from low temperature glass state to the high temperature elastic state, the so-called glass phase transition (Szcześniak *et al.* 2008). This phenomenon is described as polymer softening. Shiraishi (2001) states that an examination on thermal softening of wood as a whole reveals that no thermal softening of wood can be identified similar to that of individual components. Wood softening properties are not formed as the simple summation of individual polymer properties but as a result of a mutual interaction. Although lignin and cellulose are amorphous polymers and much more thermoplastic than cellulose (which is a highly crystalline), their thermal behaviour might be restricted by interactions due to secondary intermolecular bonding with cellulose.

Shiraishi mentions that wood shows thermal softening only at temperature above 200°C because the thermoplasticity of wood is governed by cellulose, which softens at temperatures above 200°C in dry state. This might be one theoretical reason for the need to use temperatures above 200°C to obtain reliable bonding when using only heat and pressure. But the crystalline structure melting point of cellulose is considerably above its decomposition temperature. Thus, no melting of cellulose can occur at temperature that do not cause pyrolysis (Shiraishi 2001).

Pyrolysis is defined as the chemical decomposition of organic materials by heating in the absence of oxygen or any other reagents, except possibly steam. This phenomenon commonly occurs whenever solid organic material is sufficiently heated, e.g. when frying, roasting, baking, toasting. (Even though such processes are carried out in a normal atmosphere, the outer layers of the material keep its interior oxygen-free.)

The literature study revealed that several methods involving different types of equipments are used to investigate changes in wood polymers at high temperature: FTIR (Fourier Transformed Infrared Spectroscopy), DTA (Differential Thermal Analysis), TG (Thermogravimetry) together with DTG (Differential Thermogravimetry) and DSC (Differential Scanning Calorimetry). According to the research field and the final purpose of the study, these methods (especially DSC) were used to analyse either polymer softening (when research team belonged to wood molecular physics or thermo-mechanical pulping area), either polymer decomposition (when the study concerned wood pyrolysis).

Using thermogravimetric analysis in both inert (nitrogen) and oxidizing (air) atmosphere, Órfao *et al.* (1999) showed that the rates of cellulose decomposition become measurable at 225°C, xylan starts to decompose at 160°C while lignin starts decomposing at 110°C. Concerning wood samples, the same source shows that pine degradation occurs at 140°C while eucalypt starts reacting at 155°C. The evolution of lignocellulosic material (pine and eucalypt wood) degradation at a heating rate of 5K/min is presented in Fig. 2.5.

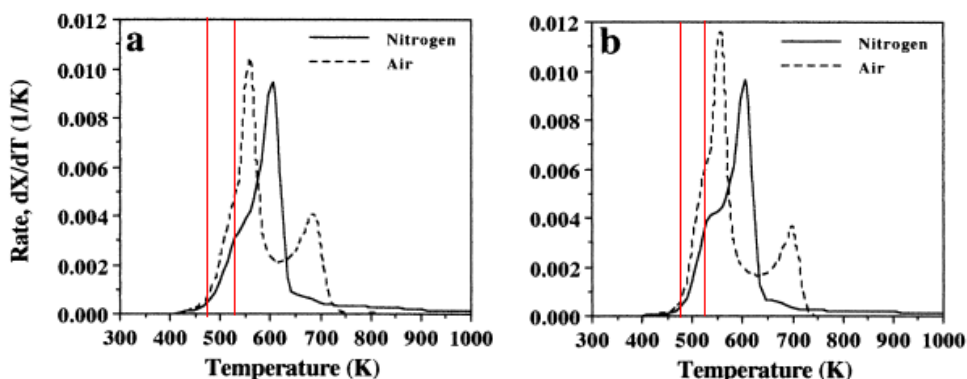


Fig. 2.5 Experimental DTG curves of pine (a) and eucalyptus (b) wood in nitrogen and air under linear temperature programming (5K/min) (Órfao *et al.* 1999).

The red lines mark the minimum and maximum levels of temperature used when bonding veneers with heat and pressure only. Note how the shape of the curves described an important level of degradation at temperature even lower than 250°C. The degradation in air occurs faster than in nitrogen. This might be an explanation for a more darkening of the bonding areas within the hot-pressed laminated product, another assumption than the ones shown in Paper II.

A recent study by Yang *et al.* (2007) compares results obtained with different analysis methods. The DSC curves show the energy consumption property during wood component pyrolysis (see Fig. 2.6). The assumption that the charring process is highly exothermal whereas volatilization is endothermal is speculated by Yang.

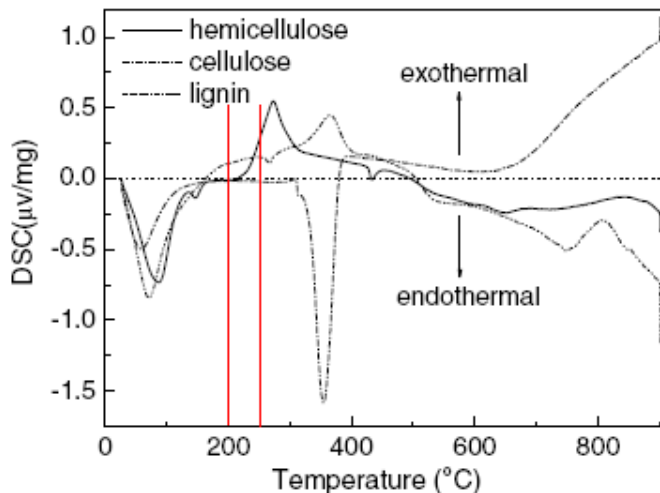


Fig.2.6 DSC curves of hemicellulose, cellulose and lignin pyrolysis, heating at a constant rate of 10°C/min. (Yang *et al.* 2007)

One can conclude that, when heating wood at a constant rate, even at temperature above 200°C and below 250°C, a charring phenomenon develops. The wood components showing to generate solid residues at these temperature levels are hemicellulose and lignin. The charring phenomenon might be involved in wood bonding mechanism when only heat and pressure are used.

The main components (cellulose, hemicelluloses, lignin) were shown to degrade at different intervals when heated because they have such different chemical structures, which is natural since they fulfil different tasks within the wood cell.

A team from Kyoto University, Japan (Shiraishi 2001) performed an intensive study to obtain adhesives from liquefied wood, on the basis that this is a very reactive product. In general terms, wood liquefaction is a type of thermochemical conversion of the material in a reducing environment and severe conditions of conversion for the purpose of obtaining oil. In Shirashi's terms, liquefaction means transforming the material (in this case wood) from solid to liquid state under milder treatment conditions. His studies of lignin liquefaction reveal that at temperatures from 200°C to 250°C, in the presence of phenol without a catalyst, lignin liquefaction occurs rapidly through homolysis (chemical bond dissociation of a neutral molecule generating two free radicals). Homolytic cleavages that occur yield several kinds of radical compounds. As the result, various compounds are produced through reaction among these radical species. Acetic acid can greatly promote the homolysis reaction without altering the reaction mechanism (Shiraishi 2001).

Sundqvist *et al.* (2006) showed that the higher the temperature used during birch heat treatment, the more acetic acid is emitted from the wood itself. It is well known that above 200°C hemicelluloses release acetic acid (Weiland and Guyonnet 2003). Acetic acid also has the role of catalyst in promoting radical formation and coupling in lignin and consequently in the self-bonding phenomenon. The description of the wood liquefaction mechanism mentioned above confirms the explanation proposed by Westermarck *et al.* (1995) and Westermarck and Karlsson (2003): it is the radical coupling rather than the softening of compounds that plays an important role in the autoadhesive process of wood and wood fibres.

It may be concluded that wood self-bonding is a phenomenon related to an initial thermal conversion stage when—determined by environmental conditions—a degradation of the unstable parts of wood polymers occurs, leading to new bonds and the formation of new compounds.

All the techniques mentioned for analysing the thermal behaviour of wood and of wood polymers should be used in a complementary way to reveal the changes occurring during hot-pressing. These tests should investigate parameter levels similar to the ones used when bonding veneers using only heat and pressure. The pressure applied for board manufacturing should be considered as part of the thermal analysis conditions.

3 INITIAL SCREENING TESTS OF MATERIAL PARAMETERS

3.1 INTRODUCTION

The purpose of this chapter is to look at the influence on the final product of the following material properties: wood layer thickness, number of layers, veneer type, initial moisture content, grain direction and species. The chapter is divided into two parts. The first part reports observations made on beech veneers concerning the influence of most of the properties mentioned. The second part is focused on the behaviour under heat and pressure of other species than beech. Most of the results presented have not been published before. A table collecting all results when other species than beech were used can be found in Appendix 1.

3.2 INFLUENCE OF BEECH VENEER PROPERTIES ON THE PRODUCT

3.2.1 Materials and methods

The material used was beech veneer. The veneer sheets used for pressing had a surface measuring 140 x 140 mm. The material parameters had the following levels :

- thicknesses: 0.66 mm, 1.5 mm, 2 mm and 2.2 mm,
- veneer production method: turned and sliced,
- moisture content: dried (to app. 4% and 9% moisture content) and soaked in water

Veneers were pressed in a Fjellmann 2032 laboratory press. Often the pressing parameters differed for each observation because the intention was to obtain a

compact board. No strategy of experimentation was used, the purpose was to explore rather than to optimize.

3.2.2 Results and Discussions

Veneer type

Whether the veneers are rotary-cut or sliced plays an important role. Tests on sliced veneers showed that these do not perform as well as rotary-cut veneer. 2-mm-thick beech veneers cut by both methods were subjected to exactly the same treatment (using the same pressing parameters). The sliced veneers did not bond.

The way cells are formed during tree growth is a model to follow when using this bonding technique. The orthotropic nature of wood is important. When trying to achieve wood self-bonding, it is advantageous to respect the way wood built itself (see Fig. 3.1).

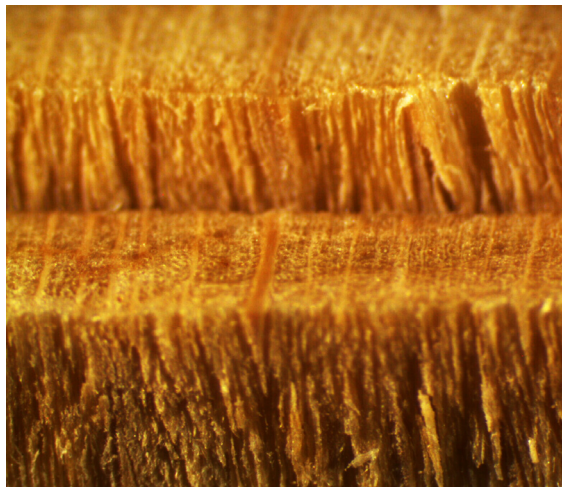


Fig. 3.1 Microscopic view (7x magnification) of two slices of rotary-cut veneer

Rotary-cut veneers have a more homogeneous surface, which helps the process, since it is easier to entangle two even, similar surfaces than surfaces with disruptions, such as those obtained by radial or semiradial slicing.

Thickness of composing layers

The results of the tests showed that 2.2 mm is the thickness that gave the highest level of both shear and bending strength. 2 mm was an acceptable thickness as well. 1.5 mm gave poor results. A package made from beech veneer of only 0.66 mm could not be successfully bonded; one of the reasons might be the need for a sufficient number of cell layers in the bonding area. This explanation is based on the Scanning Electron Microscopy images discussed in Paper I.

It was concluded that in the case of beech, a thickness greater than 2.2 mm can be translated in real terms as a rigid material, and rigidity is unfavourable to the bonding process. The layer has to be flexible. However, it becomes fragile when the thickness is too low.

Number of layers

Dried wood is known to be a good insulator because of its composite structure and the presence of wood cell lumens filled with air. The fewer layers used, the less time is necessary to bond. There is a direct dependency between the number of layers and the time. As it will be discussed in the following chapter, it might be that the temperature within the board (at the contact level) determines the bonding performance. Obviously, when less material is to be heated, the ideal bonding temperature will be reached in shorter time.

Results of the tests showed there is no linear relationship between the number of layers and the pressing time required. When there are few layers to bond, (up to 5) a time of only 40 s/layer is needed. When the number of layers increases, (for example, to 15, the maximum tried so far) 1000 s were used to achieve the same internal temperature in the middle of the board at the end of pressing. To find the

relation between the number of layers and the pressing parameters, an empirical model can be obtained from an experiment involving different numbers of layers.

A complex theoretical model to involve coefficients describing the heat-transfer speed according to the number of layers at certain moments during pressing could also be an option. Nevertheless, a heat transfer model should also take into account the chemical phenomena taking place above 110°C, such as polymer decomposition and, further on, formation of new compounds. These phenomena are very dependent on heat as well.

Initial moisture content

When veneers were soaked in water before pressing, the time necessary to bond was longer because the initial period was spent evaporating the water from the layers prior to starting the chemical degradation phase necessary for bonding. The pressing process turns a wet veneer with a moisture content well above saturation point into a lighter coloured product than a 9%-moisture-content veneer when they were both pressed using similar pressing parameters.

Taking into account the hypothesis “less water, quicker and better bonding”, one might be tempted to conclude that the lower moisture content the better. Surprisingly, a moisture content of around 4% to 5 % obtained after a predrying of the raw material lead to no good results, giving an indication that a certain number of water molecules have a well defined role in the bonding process.

Grain direction

Veneers arranged with parallel grain (Fig. 3.2a) bond better than cross-grain layered veneers (3.2b)



Fig 3.2. Longitudinal section of a – parallel-grained , b – cross-grained board; microscopical view (7x magnification)

The explanation is given in chapter 5, where Scanning Electron Microscopy images with both arrangements are presented. It is, again, wood anatomy and the tendency to re-find the original wood microstructure that is responsible for this different behaviour.

As seen in Paper I, the shear strength of a parallel-grained product can reach 5 MPa. The same pressing parameters applied on a cross-grained board result in a shear strength up to only 1.5 MPa.

3.3 USING OTHER SPECIES THAN BEECH

It was a pure coincidence that beech veneers were the first veneers used. This raw material behaved the best and gave the best mechanical-property results, but all the other species tested did bond. It is important to emphasize again that the other species veneers revealed self-bonding abilities if the pressing parameters were adapted to the veneer properties, especially to the density.

Oak (*Quercus robur* L.) boards were sensitive to pressure. Many of the boards came out of the press with splits along the fibre, possibly because the pressure used was too high. Oak cell morphology does not help this process. There is a big difference between earlywood and latewood vessel diameters (pores in cross section). This non-homogenous structure, which actually classifies it as a ring-porous wood, diminishes the self-bonding ability.

Pine (*Pinus sylvestris* L.) boards had not only an attractive texture, but they were also well bonded. The 1.5-mm pine veneers were pressed under similar pressing parameters as the beech veneers.

Sugi (*Cryptomeria japonica*) is a low-density species. The 2-mm veneers ordered and received from Japan were quite fragile, indicating from the beginning low likelihood of bonding. Nevertheless, they behaved well and formed compact boards when a pressure of 2.8 MPa was used.

Larch (*Larix decidua*) is a species with potential for the technology discussed. 5-layers veneer packed were pressed with only 2 MPa because of the layer thickness: only 0.83. The results were satisfactory.

Spruce (*Picea abies*) veneers tested were 3 mm thickness. It is a too high thickness for obtaining a board. Thus, most of the trials were not successful. More tests with 2 mm thick layers are required.

Other species tested were birch (*Betula pendula*), poplar (*Populus Tremula*) and ash (*Fraxinus excelsior*). When choosing the pressing parameters, the principle “a lower density requires a lower pressure” was used.

One important observation is that none of the boards other than beech remained compact after soaking in water. The explanation might be that either the three pressing parameters chosen were not the optimal ones for the other species, or

that beech cell walls have structural and chemical properties that are more suitable to this self-bonding technique, as it was shown in the background.

3.4 Conclusions

It is likely that all wooden species respond well to the technique of bonding with heat and pressure. The problem is to find the right material parameters and the corresponding pressing parameters.

Six properties of the raw material were discussed. The experience showed that any characteristic of the raw material should be considered a factor that influences the bonding process, from macroscopic to microscopic and chemical level.

The profile of the raw material giving the best performances is: beech veneers, 2.2 mm thick, rotary cut, 9% moisture content, packed with parallel grain direction.

4 BENDING AND SHEAR STRENGTH TESTS

4.1 INTRODUCTION

This chapter discusses the process optimization using three pressing parameters as factors and bending strength as response. The levels of temperature, pressure and time leading to the highest bending strength values are presented. The results are mainly the ones presented in Paper I for shear strength and in Paper II for bending strength.

4.2 MATERIALS

The material used for the experiments in Papers I and II was beech veneers, 2 mm and 2.2 mm thickness, with a moisture content of around 9%. The veneer surfaces were 135 x 135 mm for the first paper and 140 x 110 mm for the second.

4.3 METHODS

Board manufacturing

Levels for temperature, pressure and time were set on the electronic display of the Fjellman laboratory hot-plate press. All the three pressing parameters were kept at a constant level during one testing. The 5 layers of veneers were overlapped with parallel grain or cross grain, depending on the product desired: an LVL-like or a plywood-like board. Only the tests described in Paper I involved both types of grain arrangements. Paper II refers only to parallel grain direction of the composing layers. The veneer package was then pressed until the set-time was achieved, after which the press plates automatically returned to the initial position. The boards were then taken out from the press and allowed to cool.

Experimental design

The shear strength tests shown in Paper I, whose character was rather introductory, did not follow a rigorous design. The parameters were chosen based on accumulated experience and according to a simple criterion: the samples should be resistant to water immersion. If a certain combination was proved to result in delamination caused by wetting, that combination was no longer taken into account. The bonding parameters' ranges were 80°C–300°C, 4–5.5 MPa, 80–450 s. Each set of samples consisted of 10 veneer packages.

By way of contrast, the experiment presented in Paper II followed a response surface design, since the aim was bonding process optimization (finding the best parameter combination). The parameters used are shown in Table 4.1

Table 4.1: Parameters, their actual and coded levels used during experiment:

Parameters	Low (-1)	Medium (0)	High (1)
1. Temperature of the press plates (°C)	200	225	250
2. Pressure (MPa)	4	5	6
3. Pressing time (s)	240	300	360

A central composite – design for three factors was chosen, a face-centred cube, since the intention was to fit a second-order model. Although according to Montgomery (2005) rotatability is important for the second model to provide good predictions throughout the region of interest, a face-centered cubic design was chosen because the region of interest was considered as cuboidal.

Each of the points represented in Fig. 4.1 is a parameter combination

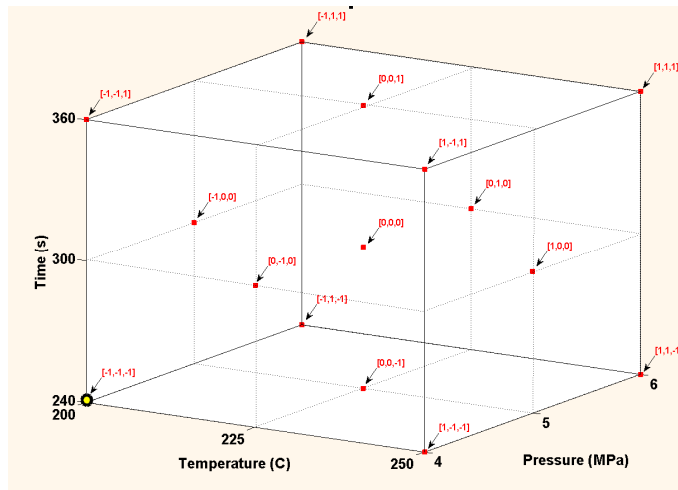


Fig. 4.1 Representation of parameter combinations used when pressing.

The test using the central point combination (0,0,0 standing for 225°C, 4 MPa, 300 s) was performed 10 times extra because it is important to see how different the results are when using the same parameter levels (it help estimating the experimental error). The combination points seen on the cube were performed only 2 times each. That gave a total of $10 + 2 \times 15 = 40$.

It is important to mention that the order in which the pressings and strength tests were performed was random, generated by Minitab software.

Shear strength testing

In Paper I, shear tests were performed according to EN – 314 (1993) using a Hounsfield testing machine. Samples were immersed in water for 8 hours and then dried. One of the problems encountered was making the saw cut stop exactly half way through sample thickness. Previous experience showed that the slightest deviation from the middle of the sample induced important error in the results.

Bending strength testing

The bending sample dimensioning from EN – 310 (1993) could have been easily respected if the press plates had been bigger. The problem encountered, and its solution, are presented in Paper II. Another simple solution would have been to press fewer veneers than 5 when manufacturing the board. Thus the sample thickness would have decreased and it would have been possible to get closer to a sample length of $(20 \times \text{nominal thickness} + 50 \text{ mm})$, as EN – 310 (1993) requires.

4.4 RESULTS

Shear strength tests results

The results of shear strength tests reported in Paper I are presented in Table 4.2 for the parallel-grained veneers (LVL-like board) and in Table 4.3 for cross-grained veneer (plywood-like boards)

Table 4.2: Pressing parameters and corresponding shear-strength values for laminated veneers with parallel grain direction under high heat and pressure

Sample set nr	Temperature (°C)	Pressure (MPa)	Time (s)	Shear strength (MPa)
1	250	5.5	180	4.89
2	240	5	240	3.58
3	300	5	80	2.01
4	250	4.5	240	5.85
5	265	4	150	2.57
6	240	5	360	5.78

Table 4.3: Pressing parameters and corresponding shear-strength values for laminated veneers with cross-grain direction

Sample set nr	Temperature (°C)	Pressure (MPa)	Time (s)	Shear strength (MPa)
1	250	5	300	1.01
2	260	5	240	1.23
3	250	5.5	420	1.9
4	240	5	480	1.56

Bending test results

The results of all 40 bending strength measurements are presented in Appendix 2. This collection of data is not to be found in Paper II, but it represents the basis for all further calculations and discussions within the paper. From all 40 observations, the evolution of 9 samples during the bending load was compared (see Fig. 4.2)

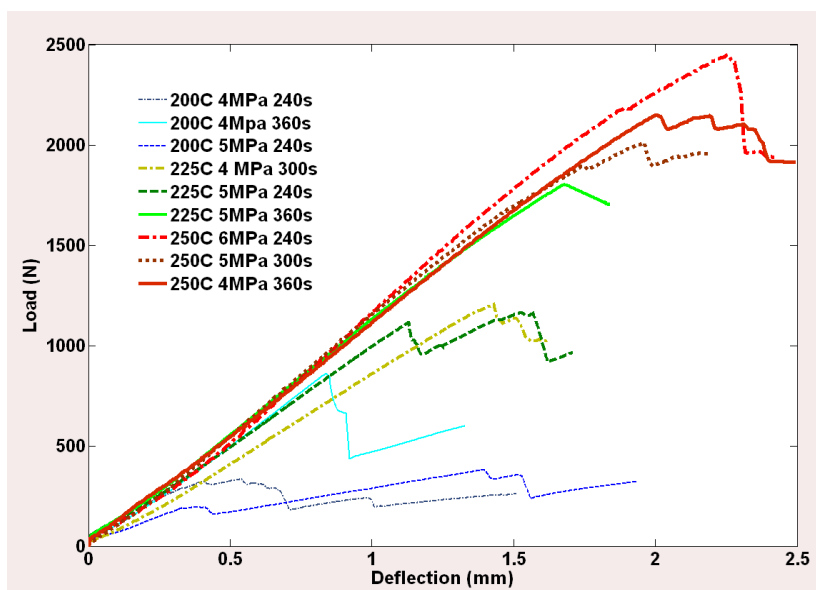


Fig. 4.2 Load deflection curve for 9 samples of laminated boards.

4.5 DISCUSSION

Observations on shear-strength testing

With regard to the shear strength tests, it was interesting to discover that a parallel arrangement of the composing veneers gives higher values and could be considered an advantage. A detailed discussion on this topic follows in the next subchapter. The results showed an interdependence of the 3 pressing parameters, but no model was searched for at that time.

The way the parameters were chosen was both intuitive and empirical. For example, in Table 1, a high level of temperature is shown, 300°C. The time used for that run was only 80 seconds. Why exactly 80 seconds? Because the experience of the manufacturer was that a certain smell revealing the emission of a certain combination of gases is related to the moment when the pressing should stop. A model is obviously called for in order to be able not only to predict the results but also to know the right parameter combination. Such a model was intended in Paper II, but the range of temperature parameters did not go as far as 300°C because the pressure and the time chosen for that design were too high for a good result when pressing at 300°C. There would have been a continuous risk of explosion. Pressing for more than 80 s at 300°C could have given exquisite results, but would have ruined the equipment.

Bending-strength data analysis

Working according to a well-established design, as in Paper II, proved to be a better choice. Trying to find such a model gives not only the satisfaction of finding an equation to fit the technological process, but also provides the opportunity to study the influence—if there is any—of each factor separately —and, most of all, the influence of the interactions between them. The interactions had already drawn a lot of attention in Paper I.

The model obtained with the help of Minitab software is discussed in detail in Paper II. The path to obtaining this model was not a smooth one, because of the combinations that had very low results. These extreme points were included in the design for the sake of showing where “the edge” might be, what combinations of parameters do not offer a reliable product. The impact of their presence in the set of data was too high, distorting the shape of the final equation.

Taking out the low extremes (200°C, 4 MPa, 240 s) from the data set to be analysed solved the problem, but it also implied that the model is not to be used to predict results for that point. But, as mentioned, it was of no interest to predict around these extreme points; on the contrary, it was important to be able to predict what pressing parameters should be used to obtain the highest results possible.

The model obtained was:

$$\begin{aligned}\hat{y} = & 158.074 + 73.394 \cdot x_1 - 1,212 \cdot x_2 + 16.520 \cdot x_3 - 40.484 \cdot x_1^2 \\ & - 15.444x_3^2 - 33.086 \cdot x_1 \cdot x_2 - 46.801 \cdot x_1 \cdot x_3 - 57.081 \cdot x_2 \cdot x_3\end{aligned}\quad (1)$$

Where x_1 = press plates temperature

x_2 = pressure

x_3 = time

$\hat{y}$ = bending strength

One can notice that it is a quadratic model, but the square of pressure (x_2^2) is not present. It was excluded because its P-value was high. This means that the possibility that this parameter has a high influence on the model is low.

According to the statistical analysis, all three pressing parameters influence the bending strength in a rather complicated way, which is caught by the interactions.

Model prediction capacity is quite high, as it can be seen in Fig. 4.3.

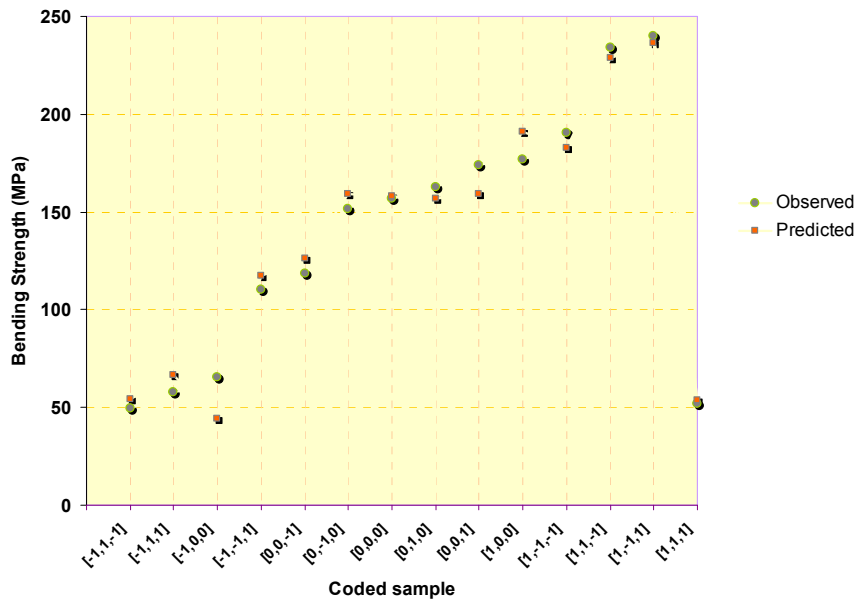


Fig.4.3 Comparison between the predicted values and the averages of the observed values.

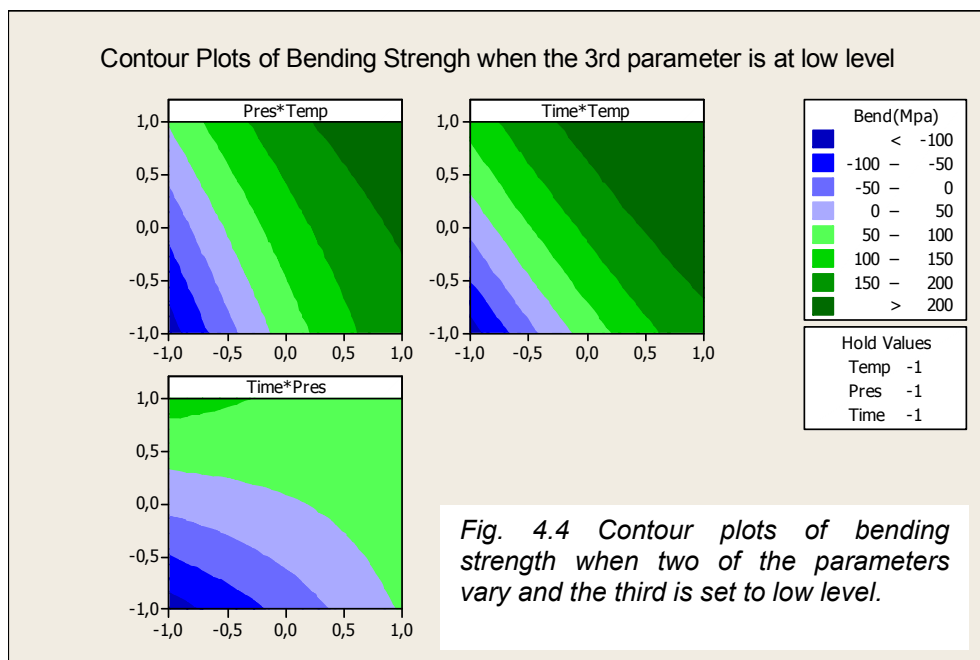
The intention was to obtain a response surface graph capable of showing the participation of all three parameters. This would have meant having four axes—one for each parameter, and the result as well. Neither Minitab nor Matlab could offer such a facility. It was then decided to graphically represent the data in two different ways, but using the same technique: keeping one of the variables constant.

Optimization – Minitab contour plots

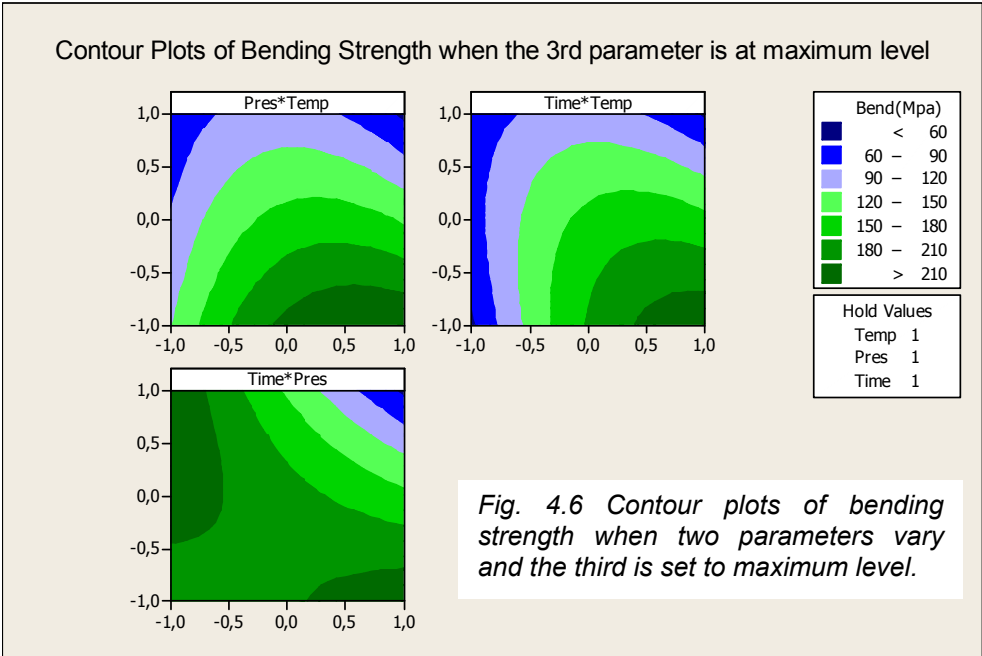
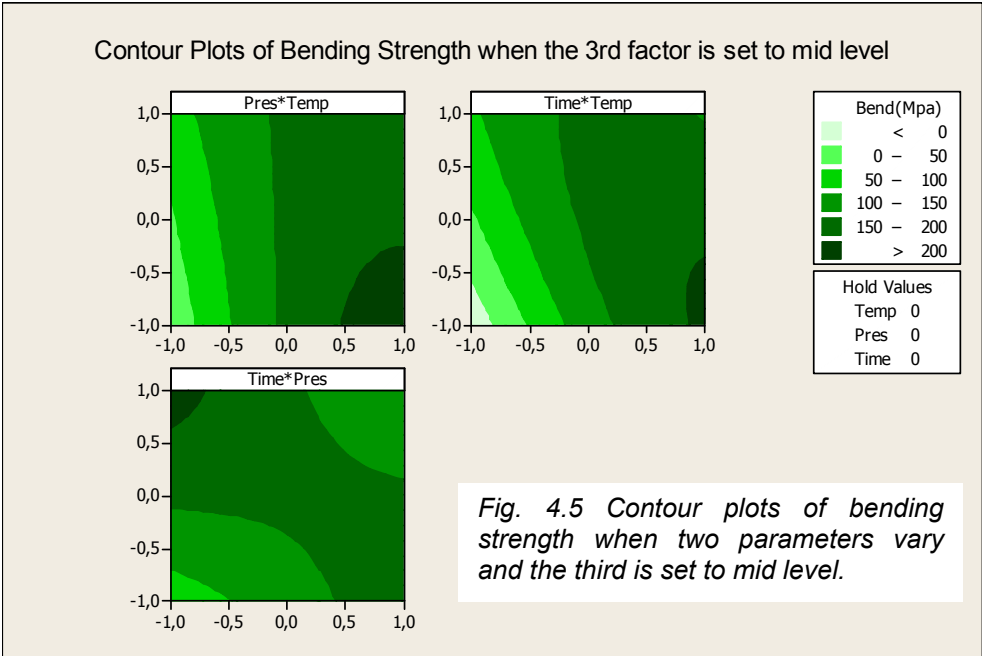
In Minitab, it is possible to obtain contour plots. They help in locating the optimum with reasonable precision (Montgomery 2005).

In Fig. 4.4, Fig. 4.5 and Fig.4.6, the dark green areas correspond to the parameter combinations leading to the higher value of bending strength. For each graph, one of the parameters is constant, while the other two vary, thus showing the

contribution of their interaction to the model. Time*Pressure contour plots (Fig. 4.4c, Fig. 4.5c, Fig. 4.6c) have a different and interesting aspect. Fig. 4.4c shows that a value above 100 MPa cannot be obtained if the temperature is kept at low level. Keeping the time at a low level requires the use of a high pressure and temperature. In the same way, if the pressure is kept at low level only a long time and high temperature can lead to a bending strength above 200 MPa.



On all nine contour plots presented (Fig.4.4, Fig.4.5, Fig 4.6), the first term of the plot title refers to the parameter levels presented on X-axes while the second term of the plot refers to Y-axes. The values on the plots (between -1 and 1) are the coded values, the actual values that they stand for are presented in Table 4.1 (page 26).



The contour plots can be useful if, for example, a certain value of one of the parameters is imposed (e.g., a certain press can use 5 MPa as pressure). One can look at Fig. 4.5 and observe that in order to obtain the maximum value of bending strength, the time level should be 240 s and the temperature level should be 250°C.

Influence of pressing parameters – Matlab plots

Minitab software did not offer the opportunity for three-dimensional visualization. The data were thus imported into Matlab software. Facing the same problem as mentioned before, “how to express four variables on three axes”, it was decided to show the shape of the equation when only one of the factors had the levels showed in Table 4.1. Fig. 4.7 looks at the shape of the surfaces when the temperature is 200°C, 225°C and 250°C.

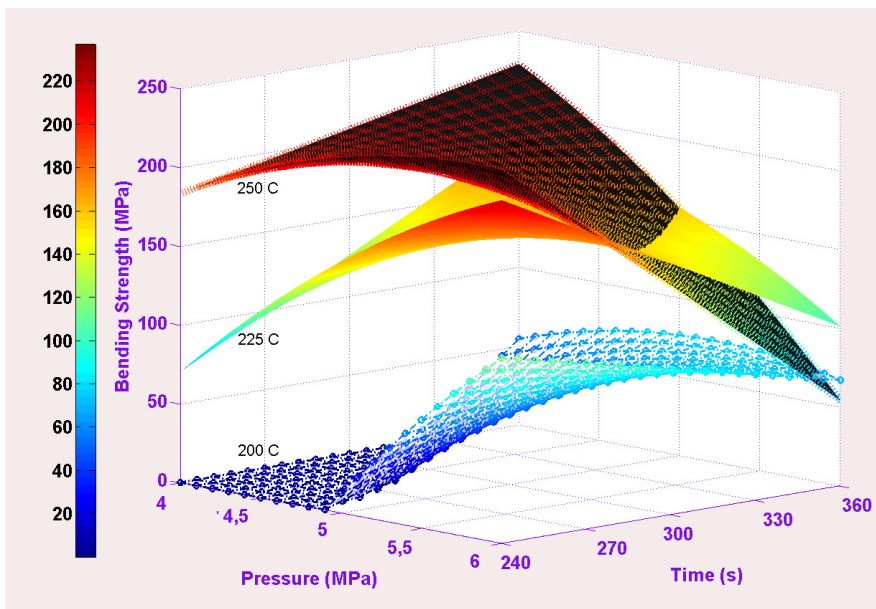


Fig. 4.7 Model representation in Matlab when pressing temperature is set to three constant levels.

For the highest level of temperature, a fall in bending-strength value can be seen if the pressure and the pressing time are also set at the highest level.

Fig. 4.8 and Fig. 4.9 show the influence of pressure and of pressing time. In both plots, when the constant factor is set to low and mid value, the shape of the surfaces tends to have an ascendant character. This means that the more the factor value used increases, the higher will be the product bending strength. This tendency dramatically changes when using the highest level for the factors. At a certain point there is a decline in response value that is actually related to the observation that the sample explodes when too much energy is used in the process.

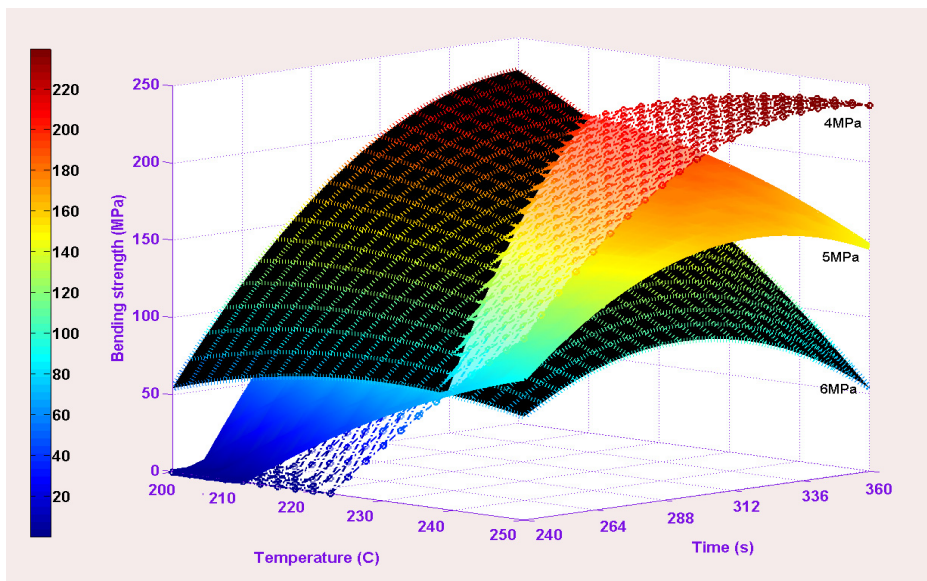


Fig. 4.8 Model representation in Matlab when pressure is set to constant levels.

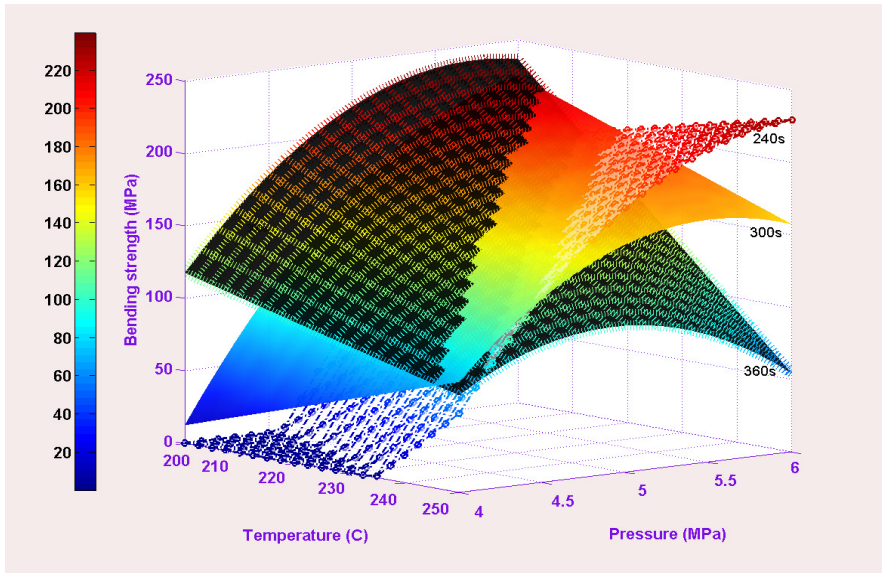


Fig. 4.9 Model representation in Matlab when pressing time is set to constant levels.

4.6 CONCLUSIONS

Shear- and bending-strength results of the final product are dependent on the pressing parameters used during manufacturing.

Shear-strength values for boards with layers arranged with parallel grain are higher than the values for cross-laminated boards.

Three parameters were analysed, all related to the equipment used, a hot-plate press. The statistical surface design analysis revealed that all three parameters influence bending strength in a rather complicated way, which is caught by the interactions. The fact that the interaction between parameters is so strong might imply the need for another parameter to be taken into account. The new parameter

should be able to correlate the effect of the others already analysed. Internal temperature within board, measured between the composing layers, might be the right factor to choose for further analysis. Actually, it would characterize not only the pressing parameters, but also the raw material properties. Their importance is also considerable they affect the shear and bending strength. This subject will be discussed in the next chapter.

Using an experimental design proved to be a very helpful tool. It is not only an organized way of planning how to manufacture the sample and to perform the tests, but it also of great benefit to analysis of the data. The goal of finding which parameter combination produced the optimum bending strength value was achieved.

With regard to bending strength, the highest values can be reached by either combining a high pressure (6 MPa) with a short pressing time (240 s) or a low pressure (4 MPa) with a longer pressing time (360 s). The plate temperature should be above 225°C. The explanation for this lies in the chemical changes that must take place in the wood structure in order to create a good bond.

5 OPTICAL ANALYSIS OF THE PRODUCT

5.1 INTRODUCTION

The focus of this chapter is solely on the final product, using images acquired. The questions to be answered are:

- How important are the changes in appearance during the transition from veneer package to laminated boards? Could they been seen as an advantage?
- Is there a variation of density within the product?
- Has the process affected the ultrastructure of the cell wall?
- The bonding area—what does it look like?

Part of the information can be found in Paper I. The light microscopy images have not been published before. The five methods were arranged in ascending order according to the increasing level of magnification used.

5.2 MATERIALS AND METHODS

Beech veneer laminated boards manufactured according to the process described in the previous chapter were chosen for this series of tests. The pressing parameters used are presented when presenting the equipment used for each method.

Photography

A Nikon Coolpix S1 digital camera was used to observe the modification in colour before and after applying the bonding technology, as well as to assess the differences between samples pressed with different parameter combinations. The samples used (laminated product) were made from veneer of beech with red heartwood of only 1,5 mm thickness, pressed in 5 layers at 260°C and 5 MPa for 180 s. The differences in colour were optically quantified; no measurement instrument was used.

Scanning

A Hewlett-Packard Scanjet 4470c scanner was used to record images of 19 samples. The samples were cut from boards manufactured under different pressing parameter combinations that are written on each sample (see Fig.5). The results were subsequently used to evaluate the differences in colour and thickness of the samples. No colour measurement instrument was used, just human visual perception.

X-ray microdensitometry

The observations were run at the Swedish Agricultural University of Science in Umeå. The equipment consisted of a scanner especially made for microdensitometry imaging by Cox Analytical Systems AB. A Cu X-ray tube was used. The distance from the tube to the sample was set to 25 mm. The exposure conditions were 35 kV, 55 mA and 35 ms. 2 samples of 2 mm thickness (see Figures 1 and 2) and 7% moisture content were scanned. The calibration of the density was done with a cellulose acetate stick.

Light microscopy

The microscope used was an Olympus B061. The cross section of the samples was analysed. In order to obtain a clean image, the surface was first slightly wetted and then firmly cut with a very sharp blade.

Scanning electron microscopy

A JEOL 5200 SEM apparatus was used. Two samples were prepared for this test, one from an LVL-like product and one from a plywood-like product. For the parallel-grain-direction sample, the bonding parameters were 250°C, 5 MPa and 180 s, while for the perpendicular-grain-direction sample, the bonding parameters were 260°C, 5 MPa and 240 s. The preparation consisted of cutting small pieces

with 2 mm thickness and using a microtome to get a smooth and clean observation surface. Then the samples were placed on small metal stubs, fixed with carbon paste and sputtered with gold in a Denton Desk II sputter unit. The images taken and presented in this paper had a magnification of from 35x to 200x.

5.3 RESULTS AND DISCUSSION

Overcoming beech red heartwood defect

The discussion is based on two images taken with a Nikon Coolpix digital camera of one and the same veneer sample placed at the top of the veneer package. Fig 5.1a shows the veneer before pressing. Fig.5.2b shows the same veneer after pressing, when it became the top surface of a 5 layer board..



Fig. 5.1 Beech veneer with red heartwood before and after pressing.

The actual purpose of this test was to demonstrate that beech with red heartwood might be a suitable material for lamination using heat and pressure. The uses of beech with red heartwood are avoided, mainly for aesthetic reasons. Its presence severely reduces the timber quality (Knoke 2003). Such a difference in the colour of one and the same object as presented in Fig. 5.1a can be disturbing for the customer. However, this problem can be overcome if the treatment of bonding using only heat and pressure is used. Fig. 5.1b shows the same piece of veneer after being subjected to the bonding treatment. The difference between the area of the veneer with and the area without red heartwood has disappeared, as can be seen in Fig. 5.1b. Thus, the darkening effect of the high temperature of the press plate (260°C) combined with a high pressure can have a positive effect.

Changes in colour and thickness

A simple and very accessible tool for recording the appearance of several samples under the same conditions of light is a scanner. Generally, it is defined as an electronic device that generates a digital representation of a document for data input to a computer. In this case, the “documents” were the laminated slices arranged one under the other.

As it can be seen in Fig. 5.2, the higher the values of the pressing parameters, the darker the product colour became. The final density of the product was affected in the same way, and this can be visually quantified by comparing sample thicknesses.

This phenomenon needs further research. According to Fig.5.2, there is a relation between of energy used during the pressing process, the level of colouring and the final thickness. This relation would be interesting to explore.



Fig. 5.2 Scanned image of 19 samples. The numbers stand for the pressing parameters used to manufacture them (plate temperature, pressure, time)

Density variation within board

It was expected that the boards would become denser during the hot pressing process. It is the simultaneous effect of pressure—pushing out the water and the air from the lumina—and heat—decomposing the components of the wood cell layers—that affects the entire structure. The x-ray microdensitometry images show that density is not uniform within the board.

The bonding areas are denser than the rest of the sample. The radial parenchyma cells (rays) show a higher density than the other types of constituent cells (see Fig. 5.3).

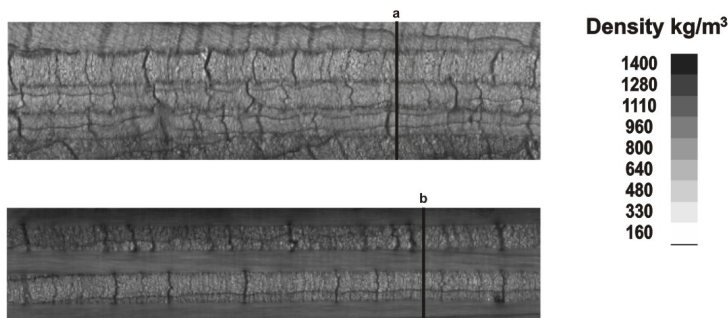


Fig. 5.3 X-ray Microdensitometry images of laminated veneers; greyscale value; a, layers with parallel grain direction; b, layers with perpendicular grain direction.

The density profiles (Fig. 5.4) helped in calculating the approximate value of the minimum and the maximum level of density.

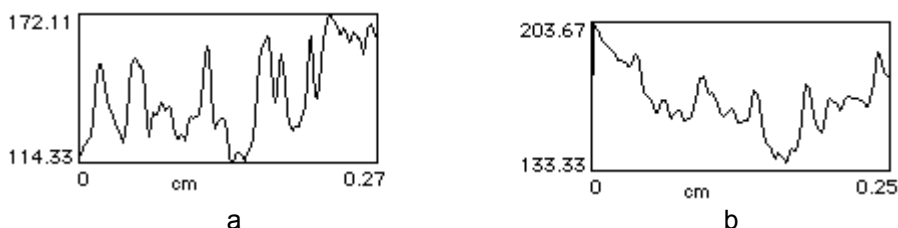


Fig. 5.4 Density profiles of laminated veneers: a, layers with parallel grain direction; b, layers with perpendicular grain direction.

Considering 255 the maximum level, corresponding to 1400 kg/m^3 , and 0 as 0 kg/m^3 , a calculation of the peaks in sample density was performed by interpolation. It was found that the corresponding value of 172.11 is app. 944 kg/m^3 , which represent the maximum level of density for the first sample. In the same manner, 203.67 corresponds to 1118 kg/m^3 , representing the maximum density reached across the second sample.

The explanation for the higher density in the bonding line, from the point of view of this method, consists of an accumulation of substances in those contact areas.. Their nature is difficult to describe, and it actually represents the key to the bonding process. Therefore a deeper look into the bonding areas was required.

Ultrastructural changes

Light microscopy revealed the transformations of the material structure. Figure 5.5 shows raw material images. Note the straight shape of the rays and the almost round shape of the pores.

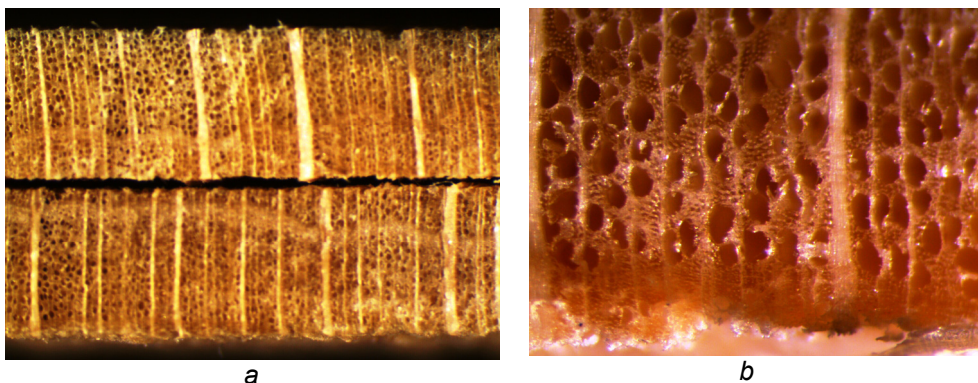
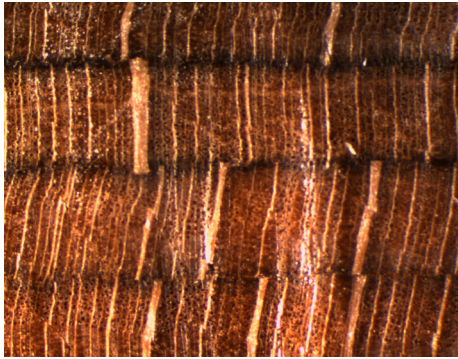
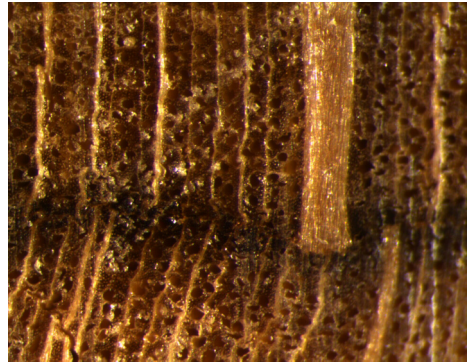


Fig.5.5 Microscopic view of (a) cross-section of two veneers ready to be pressed. Detail of a cross-section: (a)two veneers ,7x magnification; (b) 50x magnification.

Pressing at 200°C with 6 MPa for 360 s affects the shape of the pores and, to a certain extent, the structure of the rays. However, the rays seem to have remained relatively straight, as seen in Fig. 5.6.



a-7x magnification



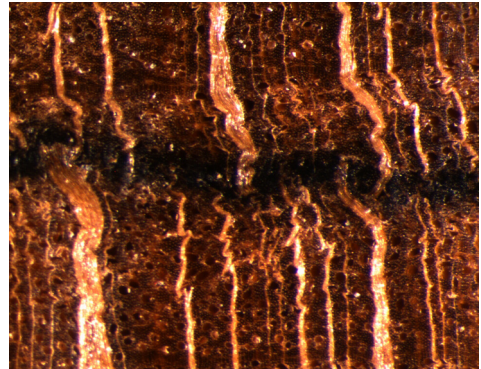
b-30x magnification

Fig. 5.6 Microscopic view of a cross section from board pressed at 200 °C, 6 MPa, 360 s.

From Fig. 5.7a and Fig. 5.7b, showing a sample pressed at 250 °C, 4 MPa, 360 s, it can be seen that the pores are compressed, their size diminished considerably. The shape of the rays is wavy, not straight as before, but they do not seem to have broken.



a-7x magnification



b-30x magnification

Fig. 5.7 Microscopic view of a cross section ; sample pressed at 250 °C, 4 MPa, 360 s

A comparison between 30 x magnified images of the two boards (Fig. 5.6b and Fig.5.7b) reveals that the bonding area is thicker and more visually obvious for the sample pressed at 250°C, 4 MPa, 360 s. More of the raw material seems to have

been strongly degraded and is involved in the bonding area. There is no sign of collapse, especially concerning the rays.

The bonding area – differences between parallel and crossed laminates

A useful method to analyse the bonding area proved to be Scanning Electron Microscopy. A detailed discussion on several micrographs can be found in Paper I. It was important to clarify why the samples with parallel grain showed a higher shear strength, which is related to a better internal bonding. The answer is in the comparison between Fig. 5.8a, with the layers parallel, and Fig. 5.8b, with the layers arranged perpendicularly. In 5.8a, the cells from neighbouring layers are able to entangle, the top of rays from one layer find room in the layer above. In the second case, the gaps between the two layers do not allow the two surfaces to have a proper contact.

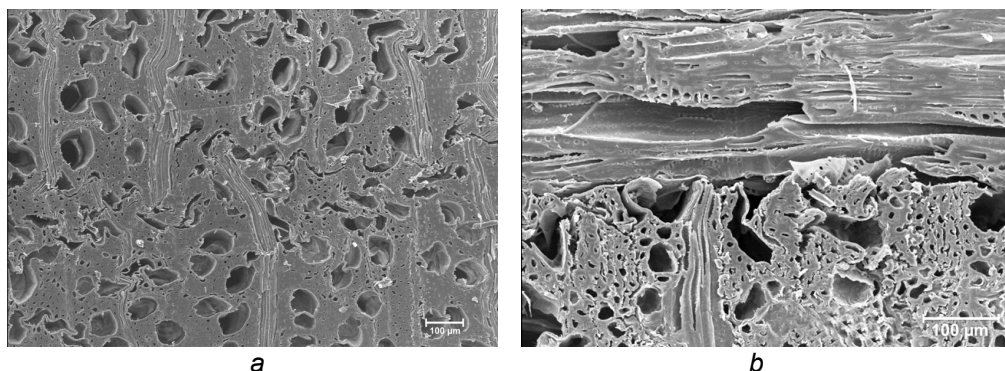


Fig. 5.8 Laminated veneers bonded with parallel (a) and perpendicular (b) grain direction — SEM image of a cross section, 200x magnification.

5.4 CONCLUSIONS

The darkening effect of heat can have positive outcomes when beech veneer with red heartwood is used.

The density of the board is not uniform. The bonding areas are denser, as are with the rays.

The colour of the raw material becomes darker in proportion to the amount of energy used to treat the sample.

The wood cell walls gain plasticity during the pressing process. The surface cells of one layer are thus capable to find room and fit into the adjacent layer. Even the cell walls of the rays, which are pressed in their longitudinal direction, become plasticized by the combined effect of heat and pressure and become wavy.

The bonding area looks like an entanglement of vessels cell walls belonging to the two adjacent layers. It turns darker than the rest of the board and seems to be carbonized. Paper II describes several possible reasons for this. Connections between the darkening phenomenon and some bonding pathways are suggested upon there.

6 FINAL CONCLUSIONS AND FUTURE WORK

The thesis presented the possibility of manufacturing a new laminated product by simply pressing veneers in a hot plate press. The properties of the product are influenced by material factors and interrelated pressing parameters.

The following parameter levels were found to be the most suitable for board manufacturing using only heat and pressure:

- material: rotary-cut beech veneer, 2.2 mm thickness, 9% moisture content
- 5 layers arranged with parallel grain direction
- pressing conditions: either 250°C, 6 MPa, 240 s or 250°C, 4 MPa, 360 s

A parallel-grained board pressed at 250°C, 4 MPa, 360 s reaches bending strengths as high as 244 MPa and a shear strength of 5.76 MPa. Such a board could be considered a representative sample when investigating other aspects: hardness, swelling behaviour, biological deterioration, electrical and acoustical properties, creep behaviour, machining and fire retardancy.

A new parameter needs to be taken into account: the temperature reached within the board. A model describing the relation between this factor and the pressing parameters is required. The internal temperature might also be connected to darkening at the macroscopic level, to densification at the microscopic level and to polymer decomposition at the chemical level.

The question “Why is beech the most suitable species for this technique?” deserves an answer. Nevertheless, investigating the use of other species in this technique is important as well.

Probably the most difficult task will be to find out what types of chemical bonds are formed during hot pressing and what exactly causes boards pressed above a certain level of temperature to become resistant to water soaking.

The assumptions as to the causes of a stronger darkening at the bonding layers need scientific support. Whether the cause is the presence of oxygen at the surface of veneers and not inside the veneer, the deposition of low-molecular soluble compounds during drying or the formation of condensed compounds due to the homolytic cleavage of lignin remains an open subject.

Finding out the chemical reactions behind the phenomenon of bonding veneers using only heat and pressure might help in other areas such as pellet production, chipboard and fibreboard manufacture, pulping, composites based on wood polymers, thermal conversion and heat treatment of wood.

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APPENDIX 1

Initial screening experiments results, parallel-grained layers in all observations

Species	LayerThick- ness(mm)	Number of layers	Plates Temp (°C)	Pressure (MPa)	Pressing time (s)	Bonded	Obs
pine	1.5	3	250	7	180	no	
pine	1.5	3	260	4.2	240	yes	
pine	1.5	3	260	3.12	210	yes	
pine	1.5	3	260	3.12	180	no	
pine	1.5	5	230	4.5	300	yes	
pine	1.5	5	240	4	240	yes	
pine	1.5	5	250	3.5	240	yes	
spruce	3	3	250	3	240	no	
spruce	3	4	250	3.5	240	no	
spruce	3	3	250	3	290	yes	
oak	0.66	3	240	3.36	160	no	
oak	2.5	3	250	4.2	180	no	
oak	2.5	3	235	8.33	180	Yes	split
oak	2.5	3	230	6.34	180	yes	split
oak	2.5	3	245	3	150	yes	
oak	1.66	5	250	3	240	yes	
sugi	2	5	250	5.7	120	no	
sugi	2	5	250	2.8	140	yes	
sugi	2	5	240	2.8	180	yes	
larch	0.83	5	250	2	130	no	
larch	0.83	5	250	2	200	yes	
larch	0.83	5	250	1.75	200	yes	
		5					
ash	1.47	5	250	3.5	200	yes	
ash	1.47	5	250	3.25	240	yes	
ash	1.47	5	250	3	240	yes	
poplar	1.75	6	240	3.5	300	no	
poplar	1.75	6	240	5	360	yes	

APPENDIX 2

Shear and bending test results. The raw material used was rotary-cut beech, 2.2 mm thick, parallel-grained, 5 layers, 9% moisture content. The size of the manufactured boards was 140 x 110 mm. The most fortunate combinations of pressing parameter values are highlighted.

Press plates temperatures (°C)	Pressure (MPa)	Pressing time (s)	Dry shear strength (MPa)	Bending strength (MPa)
250	6	360	0,89	42,19
225	5	300	3,28	114
200	4	360	2,07	107,7
200	6	360	2,91	73,55
225	5	300	3,49	135,2
225	6	300	4,2	153,5
225	5	300	5,05	178,7
225	5	300	4,42	161,6
200	6	240	1,12	28,08
250	4	240	4,08	188,4
225	5	360	4,97	172,5
225	5	300	4,08	166,5
250	5	300	4,55	151,5
250	6	240	4,86	239
200	5	300	2,5	62,74
250	5	300	4,55	202,6
225	5	300	2,93	140,4
225	6	300	4,03	172,5
200	5	300	2,5	68,2
225	4	300	3,72	161,6
225	5	300	3,81	178,2
200	6	360	2,76	42,19
250	4	240	4,12	192
200	4	240	1,3	39,3
200	6	240	2,01	70,5
225	5	360	4,8	175,3
250	4	360	5,76	244,2
225	5	300	3,31	156,7
250	6	240	5,12	228,9
200	4	240	2,1	36,53
225	5	240	3,27	120,2
225	5	240	3,4	117,2
225	4	300	2,68	141,6
200	4	360	1,83	112,5
250	4	360	5,8	235,6
225	5	300	5,19	206,9
225	5	300	3,8	144,7
250	6	360	1,7	61,9
225	5	300	3,93	145,7
225	5	300	4,2	156,7

Bonding Laminated Veneers with Heat and Pressure Only

Proceedings of the 2nd international conference on environmentally-compatible forest products
“Ecowood” Porto, Portugal, 2006

Paper I

BONDING OF LAMINATED VENEERS WITH HEAT AND PRESSURE ONLY

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SUMMARY

A new engineered wood product was obtained by pressing several layers of veneers at high temperatures. Experiments with different pressing parameters were performed in a laboratory hot plate press on *Fagus Sylvatica*. Shear strengths test were run according to EN 314-1 and EN 314-2 "Plywood – Bonding qualities". Samples were analysed with a digital camera to observe the change in colour and with a X-ray microdensitometry scanner to determine the density variation and profiles. Scanning Electron Microscopy (SEM) was used to study the modification of wood cell structure close to and inside the bonding zones. The results showed that the bonding properties of the laminates are good, better when veneers have the same grain direction. Beech veneer with red heartwood can improve its appearance if bonded with the technology presented.

INTRODUCTION

Bonding veneers has always been done with the help of adhesives or surface treatments, resulting in products like plywood and LVL. No references were found about flat sheets of veneer being bonded as laminates with no activating substances, pre-treatment or adhesives. On the other hand, the technology of wood welding known from the work of Gfeller *et al.* 2002 and Stamm *et al.* 2005) implies, by its definition, a friction phenomenon responsible for the success of this method. The question: “is friction compulsory or not when bonding solid wood with no adhesives” was consequently raised. In order to answer this question tests of pressing solid wood at high temperature have been carried out at Luleå University of Technology. The samples initially used were from 15 to 30 mm thick. The observations and the results led to the idea of decreasing the thickness of the samples to improve the bonding properties. Therefore veneer sheets were used at the same pressing parameters and two new wooden products were obtained: LVL and plywood containing no adhesives. This paper presents the bonding technology as well as images and properties of the products.

MATERIALS AND METHODS

Bonding of veneers

Rotary-cut European beech (*Fagus sylvatica* L.) veneer showing moisture content of 9% and 2 mm thickness was sawn into of 135 mm x 135 mm sheets. A Fjellman laboratory hot plate press of small dimensions was used for pressing. The plates of the press (140mm x 140 mm) were heated up to the pressing temperature previously set. 5 layers of veneers were mounted on top of each other inside the press. The temperature and the pressure were maintained constant during the pressing. Several combinations of

parameters were used. The pressing parameters initially had a large variability. The condition of resistance to water immersion for 24 h (pre-treatment imposed by EN-314) excluded some of the parameter combinations.

Finally the bonding parameters ranges were: 80°C - 300°C, 4 - 5.5 MPa, 80 – 450 s (see Table 1 below). After the pressing time finished the laminated products were removed from the press and in cooled at room temperature. In one series of experiments the veneers were all arranged in the same grain direction and a product similar to LVL was obtained. In the second series of experiments the direction of the grain is perpendicular as in plywood. Each set of samples presented in table 1 contained a number of 10 samples. Finally shear tests were performed according to (EN – 314 , 1993) using a Hounsfield testing machine.

Photography

A Nikon coolpix S1 digital camera was used to observe the modification in colour before and after applying the bonding technology. The samples used in this test were different from the ones used for shear strength test. Here they were made from veneer of beech with red heartwood of only 1,5 mm thickness , pressed under heat in 5 layers, at 260°C and 5 Mpa for 180 s.

X-ray microdensitometry

The observations were run at Swedish Agricultural University of Science in Umeå. The equipment consisted of a scanner especially made for microdensitometry imaging by Cox Analytical Systems AB. A Cu X-ray tube was used. The distance from the tube to the sample was set to 25 mm. The exposure conditions were 35 kV, 55 mA and 35 ms.

2 samples of 2 mm thickness (see figure 1 and 2) and 7 % moisture content were scanned. The calibration of the density was done with a cellulose acetate stick.

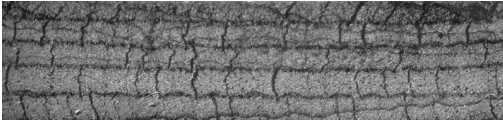


Figure 1: Cross face of LVL-like sample used

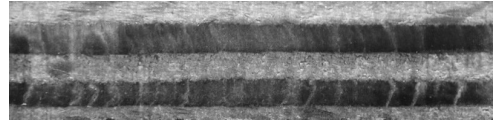


Figure 2: Cross face of plywood-like sample

Scanning electron microscopy

A JEOL 5200 SEM equipment was used. 2 samples were prepared for this test, one from a LVL-like product and one from a plywood-like product. For the parallel grain direction-sample the bonding parameters were 250°C, 5 MPa and 180 s, while for the perpendicular grain direction the bonding parameters were 260°C, 5 MPa and 240 s. The preparation consisted in cutting small pieces with 2 mm thickness and using a microtome to get a smooth and clean observation surface. Then the samples were placed on small metal stubs, fixed with carbon paste and sputtered with gold in a Denton Desk II sputter unit. The images taken and presented in this paper had a magnification from 35 x to 200 x.

RESULTS AND DISCUSSION

Bonding behaviour for beech laminated veneers - parallel grain direction

The results of the shear strength tests are presented in table 1.

Table 1: Pressing parameters and corresponding shear strengths values for laminated veneers with parallel grain direction under high heat and pressure

Sample set nr	Temperature (°C)	Pressure (Mpa)	Time (s)	Shear strength (Mpa)
1	250	5.5	180	4.89
2	240	5	240	3.58
3	300	5	80	2.01
4	250	4.5	240	5.85
5	265	4	150	2.57
6	240	5	360	5.78

One can notice the good bonding behaviour of this product, taking into account that the tests fulfilled the requirements of pre-treatment of samples for outdoor conditions, meaning that boiling of samples, drying and re-boiling was performed before applying the test shear forces.

The interdependence between the 3 bonding parameters is obvious, but no clear linear relation was found between them. Still, when keeping a certain pressure value constant, e.g. 5 MPa, a product with similar bonding properties can be obtained using either a long pressing time at a lower temperature, or using a higher temperature in a short time.

It was important to find out that a press plate temperature of 300°C can be used for pressing, showing good bonding results.(see sample set 3 in Table 1). The laboratory press capacities did not allow the use of higher temperatures. It would have been interesting to see the behaviour of beech veneers at 420°C and above, Wood is believed to reach such a high value during circular wood welding (Stamm *et al*, 2005). But one must take into account the time necessary for wood to reach the temperature of the press plates during pressing

Bonding behaviour for beech laminated veneers - cross grain direction

The results of the bonding properties tests are presented in table 2 :

Table 2: Pressing parameters and corresponding shear strengths values for laminated veneers with cross grain direction

Sample set nr	Temperature (°C)	Pressure (Mpa)	Time (s)	Shear strength (Mpa)
1	250	5	300	1.01
2	260	5	240	1.23
3	250	5,5	420	1.9
4	240	5	480	1.56

The shear strengths values show that the bonding properties of plywood-like product are lower than those of the LVL product. An explanation of this behaviour will be offered by SEM imaging (see figure 12). Although the value are in average around 3 times smaller than in same grain direction samples, the resistance to water immersion and boiling of these cross- layered samples is still good. Nevertheless the results in Table 2 showed

values comparable with shear strengths for plywood obtained with conventional bonding methods.

It is important to notice that the time required for plywood-like products to bond is considerably higher. This means that if 2 samples – one in parallel the other in perpendicular grain direction - are pressed in the same conditions, it is possible to obtain a well bonded LVL product but not a bonded plywood.

Colour change

As a general characteristic, the laminated veneers suffer an important change in colour due to such a high temperature necessary for bonding.

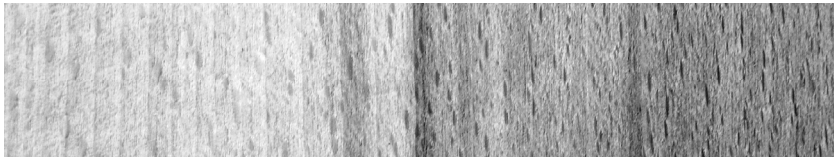


Fig. 3 Beech veneer with red heartwood before pressing

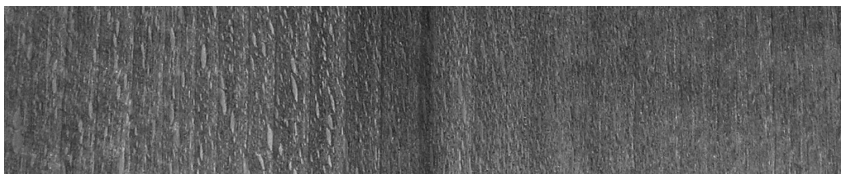


Fig. 4 Beech veneer with red heartwood after pressing

The pictures 3 and 4 show that pressing at high temperature a beech veneer laminate with red heartwood affects the colouration in a positive way by diminishing considerably the contrast between the normal colour and the red heartwood colour of the veneer

Density variation

The images obtained with the Cox Analytical Systems AB woodscanner gave a good description of the density variation. One can notice that the densification is higher at the bonding lines. The rays (radial parenchyma cells) also achieve a high density level.

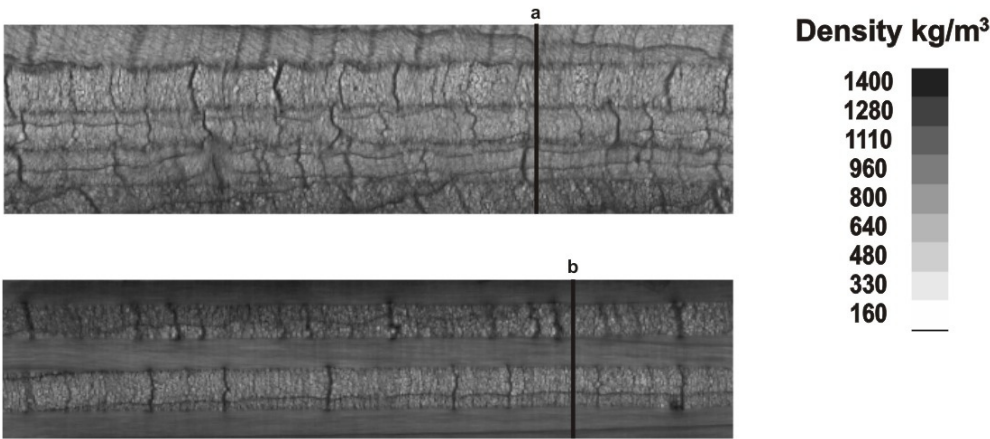


Figure 5: X-ray Microdensitometry images of laminated veneers; a – layers with parallel grain direction ; b-layers with perpendicular grain direction

A more explicit view of the density variation is shown by the density profiles below. The 5 laminates are clearly distinguished between the high density peaks of the bonding lines and the surfaces of the samples. (see figure 6)

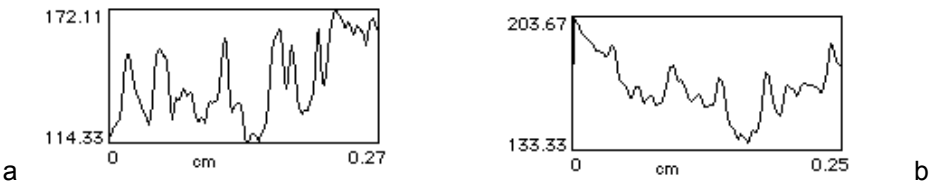


Figure 6 Density profiles of laminated veneers ; a – layers with parallel grain direction
b-layers with perpendicular grain direction

Information about minimum and maximum density values is also given. In this case the first sample has a maximum density value around 944 kg/m^3 and the other one around 1118 kg/m^3 . The 2 samples scanned had been produced with different parameters; the second one was pressed with 60 s and 10°C larger time and temperature respectively. This could be the explanation for the difference in maximum density values.

Scanning electron micrographs

SEM proved to be an excellent tool to observe the four bonding layers, as well as the rest of the product. The cells are very compressed and the bonding areas can be observed clearly as thick light vertical lines. Perpendicular to the 4 bonding lines one can notice the rays on the beech veneer layers in figure 7.

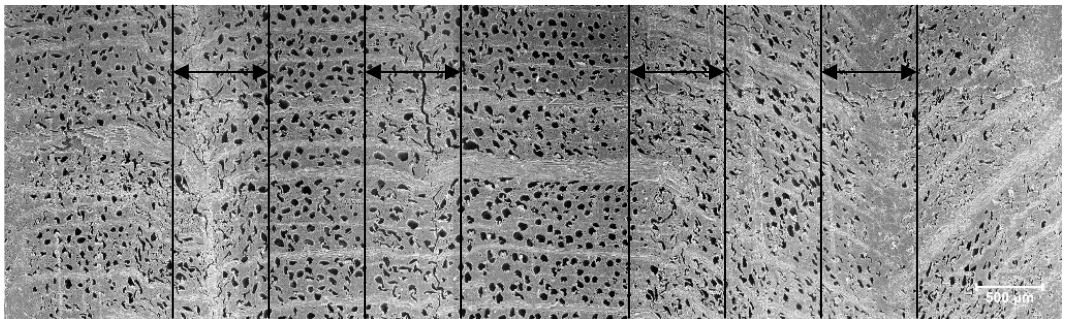


Figure 7: Laminated veneers bonded in parallel grain direction - scanning electron microscopy image of a cross section , 35 X magnification. Bonding zones are marked with arrows between straight lines.

A more detailed view is shown in figure 8 below. The rays of the 2 neighbouring layers meet at the bonding line. This SEM image presents an actual failure start (the right side) of the bonding area and it was especially chosen for observing the distinction between the layers. This bonding line is irregular and curved, showing that during compression, the cells at the surface of one of the veneer layers tried to find room to match into the other layer.

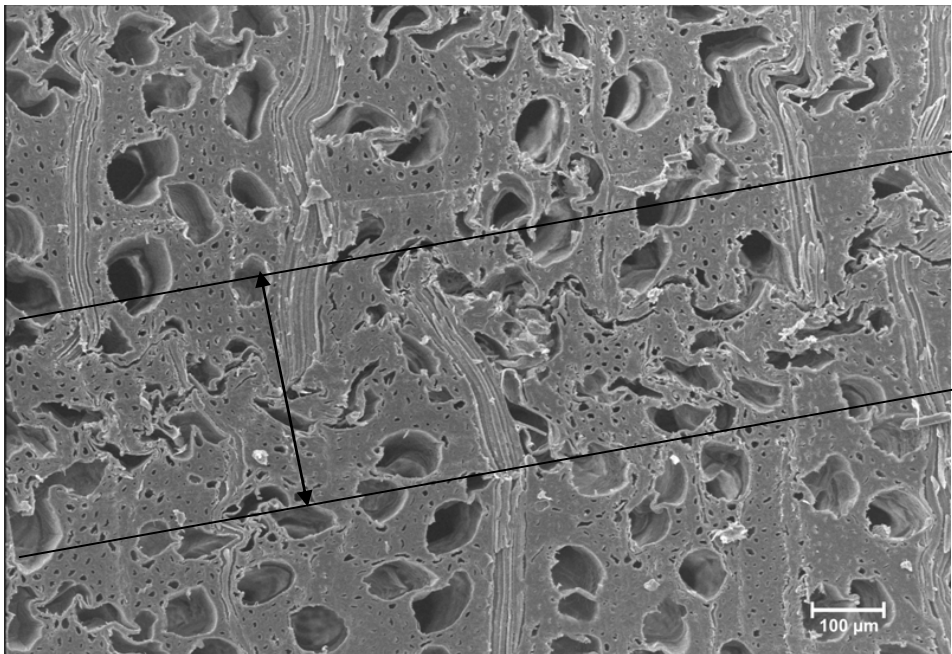


Figure 8: Laminated veneers bonded in parallel grain direction – SEM image of a cross section, 200 X magnification. The bonding line is situated between the straight lines

A clear view of the bonding layer is presented in figure 9. The cells are so compressed that the lumina of some fibres has almost completely disappeared. It is remarkable that they had not crashed, but somehow they were flexible and capable of combining and joining. The high temperature and the pressure must have been the factors responsible to obtain such a capacity of moulding of the cell walls.

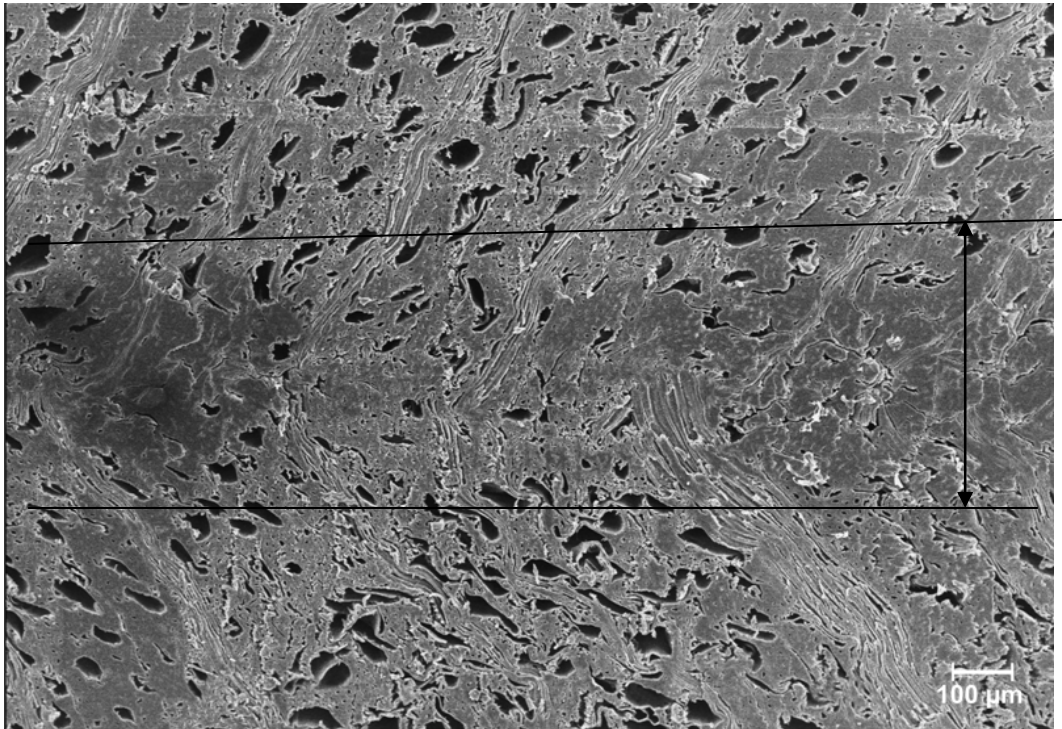


Figure 9: Laminated veneers bonded in parallel grain direction - scanning electron microscopy image of a cross section , 75 X magnification. The bonding zone is marked with arrows between straight lines.

The thickness of the bonding line has an average of 400 µm and can achieve even 600 µm in some areas. It means that the veneer layers must provide a big number of cell layers, enough material to be heated, plasticized, melted and cooled, capable of forming a strong bonding. This need of a big number of wood cells for the bonding layer can also be the explanations for not being able to bond veneers thinner than 1 mm. For example the attempt of bonding 0.65 mm thick beech veneer with different pressing parameter failed.

In a view of a longitudinal section it's difficult to distinguish the 5 layers, it looks like a compact material (see figure 10).

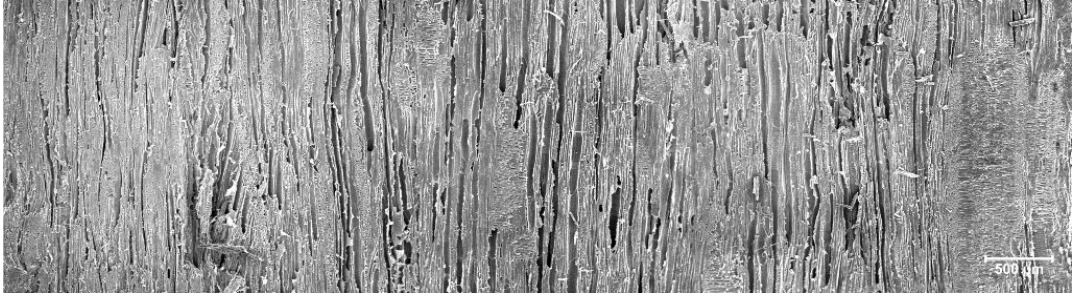


Figure 10: Laminated veneers bonded with parallel grain direction - scanning electron microscopy image of a longitudinal section, 35 x magnification

In the case of cross-positioned veneers one can notice an interesting phenomenon: thin fracture inside the inner layer (cross-section in figure 11). These fractures provoked by the compression forces during pressing could be an explanation for a weaker shear strength resistance. As mentioned, the average shear strengths resistance of cross grain laminates is 3 times weaker than parallel grain laminates.

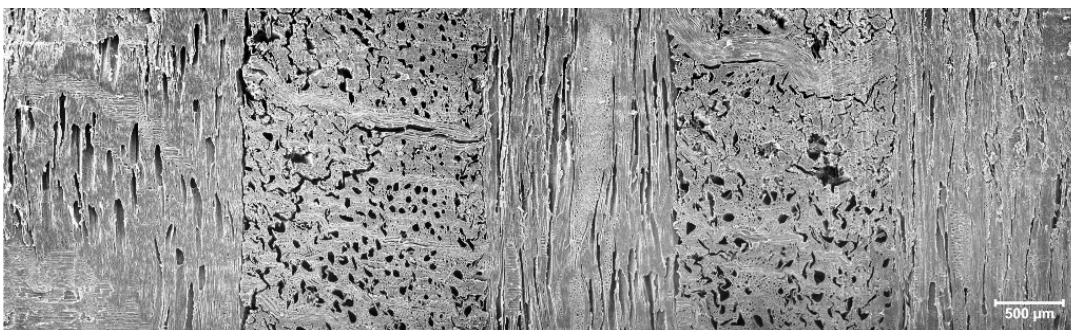


Figure 11: Laminated veneers bonded with perpendicular grain direction - scanning electron microscopy image of a cross section, 35 x magnification

In picture 12 the incapacity of the cells to entangle is even more obvious. Since the structure of wood is so orthotropic and the differences of the physical and mechanical cell properties are so high on the 3 wood directions, it is understandable why a parallel grain direction in a wooden laminate offers better bonding properties. Therefore a different orientation of the grain seems to be, in this case, a disadvantage.

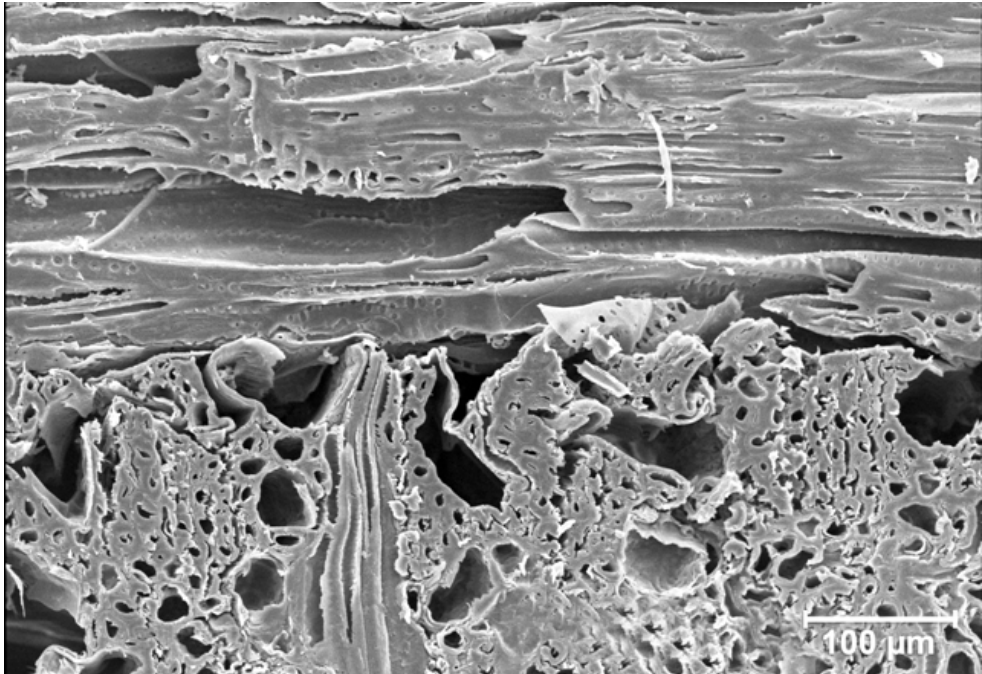


Figure 12: Laminated veneers bonded with perpendicular grain direction - scanning electron microscopy image of a cross section, 200 x magnification

CONCLUSIONS

The tests and observations showed that it is possible to obtain laminated products with no other extra-material or substance present. No adhesives and no chemical pre-treatment for surface activation are required. Furthermore, no mechanical vibration is induced in

order to produce friction between contact surfaces, as in wood welding. This adhesion phenomenon is strictly due to high temperature and high pressure.

The bonding properties tests revealed that parallel grain layered products have a better performance than cross grain layered composites. The X-ray densitometry images showed that the bonding zones proved to be denser than the rest of the laminate. This result was confirmed by the observations taken with a SEM. These images also explained why there has to be a veneer thickness bigger than 1 mm in order to achieve a strong adhesion: there is a need of sufficient number of wood cell layers capable of transforming themselves into gluing material.

Other wood species have also been tested (oak and pine), but beech showed better bonding properties and therefore it was the chosen in this investigation.

The following factors influence the bonding process: the thickness of the veneer layer, the temperature during pressing, the pressure, the pressing time, the number of layers and the species involved. These variables are all correlated to each other and an optimal combination of the process parameters is being studied. Physical and other mechanical properties of the laminate as well as the chemical reactions responsible for the bonding phenomenon are subject to further research.

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**Autoadhesion of Beech (*Fagus sylvatica* L.)
Veneers – Pressing Parameters and Bending
Strength**

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Paper II

Autoadhesion of Beech (*Fagus sylvatica* L.) Veneers – Pressing Parameters and Bending Strength

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Abstract

Autoadhesion of 2.2-mm, rotary-cut beech veneers is possible under high temperature and pressure conditions, without using any chemical activator, copolymerizing agent or steam pretreatment. This study uses the response surface method for experimental design to analyse the combination of 3 pressing factors: temperature, pressure and pressing time. 200°C, 225°C, 250°C levels are combined with 4, 5, 6 MPa for 240, 300, 360 s in 15 different ways. The 40 samples obtained were subjected to bending-strength tests, and results were analysed in Minitab software to obtain an empirical model. Samples pressed at 250°C, 4 MPa, 360 s show the best result: 235 MPa bending strength. Press-plate temperature of 200°C proves to be insufficient for a good fusion. In contrast, sample explosion caused by excessive energy input is reached at 250°C, 6 MPa, 360 s.

Keywords: beech, veneer, autoadhesion, high temperature, high pressure, time

Introduction

Obtaining a laminated product by simply pressing veneer under high temperature is a technology developed at Luleå University of Technology, Skellefteå Campus, Sweden (Cristescu et al. 2007). It does not involve the use of any pretreatment such as steaming or chemical activation. The only participants in this fusion process are rotary-cut veneers and an unsealed conventional hot-plate press. The product was inspired by the recent

technologies of autoadhesion: solid wood welding (Suthoff et al. 1996; Gliniortz et al. 2001; Ganne-Chedeville et al. 2006; Stamm et al. 2005) and oxidation of chips before pressing to form boards (Westermarck and Karlsson 2003; Widsten et al 2003, Pantze et al. 2006). Another information source was the high temperature effect on wood (Pantze et al. 2005; Sundqvist et al. 2006; Johansson 2006), related to the stickiness phenomenon of partially dehydrated food (Roos and Alves-Filho 2005).

The references found on autoadhesion without surface activation or modification (no chemical compound used prior to pressing) describe methods that were discovered as result of chipboard and fibreboard manufacturing research. They both differ from the one presented in this paper. The Thermodyn method (Runkel and Jost 1948) used a gas-tight press and a recooling phase after hot-pressing (Kollmann 1975). Masonite's plywood production method (Boehm 1951) required steaming in an autoclave prior to pressing, and cooling under pressure was necessary as well. None of these sources offered data on bending strength properties for laminated products.

Recently, Okuda and Sato (2007) report an attempt of self-bonding sugi veneers under 150°C–200°C and 3 MPa with no successful results. In contrast, Cristescu (2006) reported good autoadhesion of beech veneers obtained under 4 to 5 MPa and 250°C–300°C press-plate temperature. Clarifying which are the best parameter combinations for fusion of beech veneers is the aim of this study. Press-plate temperature, pressure and pressing time are the factors, bending strength is the response and Minitab software is the tool to analyse their relation. It is of interest to find out which of the parameters is the most influential, the maximum value of bending strength that could be obtained, but also, if there are any limits in using the chosen pressing parameters. An empirical model capable of showing what parameter levels should be used to obtain a certain response is intended as well.

Materials and methods

Experimental design

The three parameters to be analysed in this paper refer to the pressing machine parameters. The previous results (Cristescu 2006) showed that the region of interest is cuboidal. Those results helped in choosing the steps and the limits as well (delamination, fusion, char forming)

The parameters to be used for this experiment are presented in table 1 together with their corresponding levels. Samples will be named according to the coded variables, for example the notation [1, -1, 0] will stand for temperature at 250°C, pressure at 4 MPa, and pressing time 300 s.

Table 1: Parameters, their actual and coded levels used during experiment:

Parameters	Low (-1)	Medium (0)	High (1)
1. Temperature of the press plates (°C)	200	225	250
2. Pressure (MPa)	4	5	6
3. Pressing time (s)	240	300	360

The intention is to fit a second-order model,

$$y = \beta_0 + \sum_{i=1}^k \beta_i \cdot x_i + \sum_{i=1}^k \beta_{ii} \cdot x_i^2 + \sum_{i < j} \beta_{ij} \cdot x_i x_j + \varepsilon \quad (1)$$

Where:

$$x_i = \begin{cases} -1 & \text{when parameter } i \text{ is kept at its low level} \\ 0 & \text{when parameter } i \text{ is kept at its medium level} \\ +1 & \text{when parameter } i \text{ is kept at its high level} \end{cases}$$

y = the response variable = bending strength (Mpa)

β_i = regression coefficients,

ε represents the noise, which is assumed to be normally and independently distributed, with mean zero and constant, but unknown variance.

Central composite designs (CCDs) are considered to be efficient when fitting these models (Montgomery 2005). Although rotatable designs are generally recommended ((Montgomery 2005), a face-centred central composite design (CCD) was chosen for the property of reaching extreme points, which was one of this study's intentions.

The design consists of 15 different combinations of the 3 levels of the 3 parameters, including a centre point, as seen in figure 1. It was decided to perform 2 replicates of each combination of parameter settings, apart from the centre point, where 10 replicates were made in order to improve the error estimation, giving a total number of 40 runs.

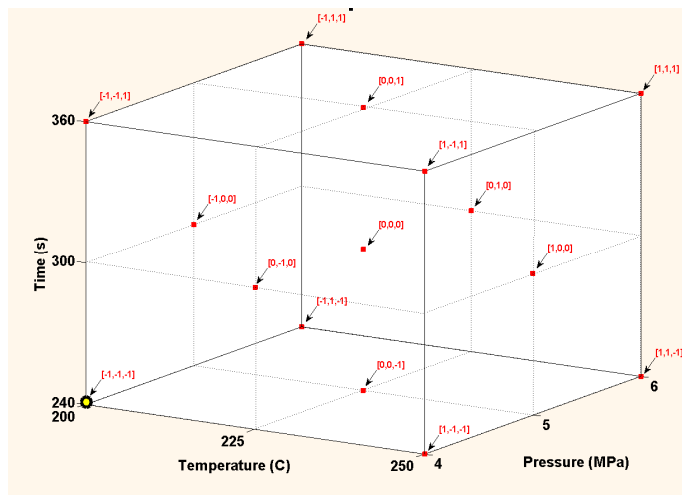


Fig. 1 Representation of parameter combinations used when pressing

The design matrix (the order in which the pressings were performed) was generated by Minitab software using the data from table 1. This order, randomly established by response surface design matrix from Minitab, was followed when pressing the veneers, but also when performing bending tests.

Board manufacturing

The beech (*Fagus sylvatica* L.) veneers used for these tests came from Silvacarpat SA, Romania. They were 2.2-mm rotary-cut veneers normally used for plywood manufacturing, obtained from steamed beech logs harvested during winter 2006 in Brasov, Romania. The logs had been steamed for 48 h prior to cutting, and the veneers obtained were dried at 140°C–160°C to a moisture content of around 10%.

In Skellefteå, the veneers were cut to small-sized sheets before pressing. This small size was imposed by the small Fjellman laboratory, with a press-plate surface of only 140 mm x 140 mm and maximum force 100 kN. Since the highest pressure needed for this test was 6 Mpa, the largest surface that could be pressed was calculated to be 140 mm x 110 mm. The veneer's thickness was 2.2 mm and the moisture content before pressing around 9%.

The pressing process started by overlapping five veneers with parallel grain in the hot-plate press. All three process parameters were digitally set on the small Fjellman 2032 laboratory press. The pressing time was counted from the first second that the force of the plate reached the targeted value. The temperature and pressure levels were continuously surveyed on the press display. A fan connected to a pipe carried away the volatile compounds formed during pressing. When the set time was reached, the pressure was released. The boards were then taken out from the press and allowed to cool outside.

Bending test

From each laminated board manufactured, a sample was cut and then conditioned. All the boards showed around 4% moisture content after conditioning. Board thickness differed according to the parameter used. The more energy that was applied (higher temperature and pressure), the lower the thickness and the higher the density of the samples. Thus sample thickness differed considerably from the [-1, -1, -1] to the [1, 1, 1] level.

The EN 310 bending test sample dimensioning could not be respected because of the reason presented above concerning the size of the press plates. The length of 150 mm was impossible to obtain with the small laboratory press; thus 130 mm was the common length for all 40 samples.

In an attempt to have a fair response from all samples, and taking into account their moment of inertia as well as the dimensions indicated in EN 310, width varied according to the thickness of the samples, following a formula derived from the Euler-Bernoulli equation:

$$w = \frac{l}{0.12} t^2$$

(2)

where w , l , t , are width, length and thickness of the samples respectively.

For this study, the span was parallel to the grain in all measurements. The span value also varied according to thickness, since a constant span/thickness ratio was intended.

It was desirable that the load act in an equal manner on all samples. Therefore the rate of load was decided after testing with different speeds on similar samples. In the end it was concluded that in order to reach failure within 30 to 90 s, the loading rate had to have values of 1 mm/s for the samples pressed at 200°C and up to 3 mm/s for 250°C samples.

Results and discussions

Bending test sample evaluation

The results of the bending tests varied from a minimum of 28.8 MPa to a maximum of 244.2 MPa. The best results were obtained for samples pressed at 250°C plate temperature.

Figure 2 shows that the laminated product can be considered a new material, in many respects different from the wood it was obtained from. One can notice that the plastic zone of these curves is very small or even absent for some samples. From an elastic state the material goes directly to maximum strength. It is not a plastic material, and this

might be explained by a diminished quantity of water molecules because of evaporation during such a high-temperature treatment. The air within lumina was also removed under high pressure. Furthermore, the entire chemical network of the wood underwent important transformations, including creation of linkages between polymers belonging to adjacent layers.

For example, a change in pressing time from the lowest to the highest level significantly improves bonding strength, as seen in figure 2. It is also obvious that interactions have a great influence, since similar responses are obtained with different combinations of two of the parameters when the third is kept constant.

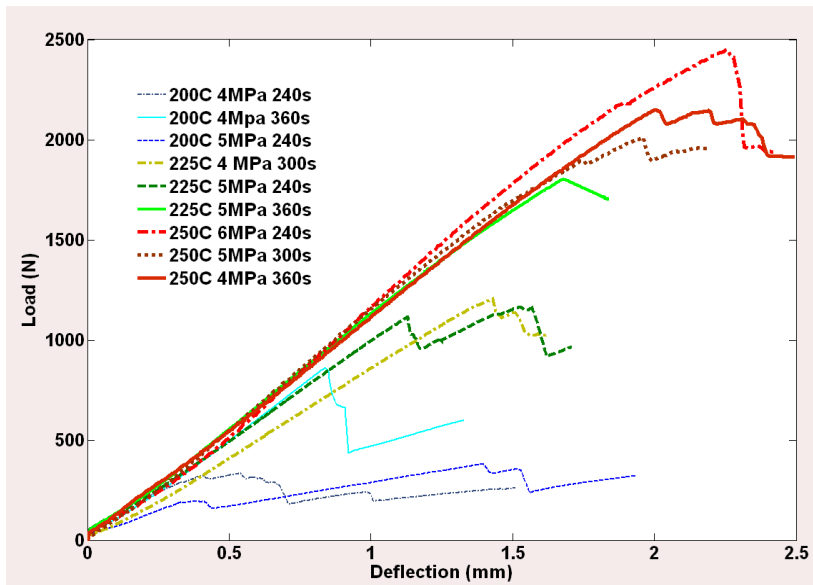


Fig.2 Load deflection curve for 9 samples of laminated boards obtained by veneers fusion

An important observation is that when pressing at maximum levels for all parameters (250°C, 6 MPa, 360 s) the results are extremely low. Both samples pressed at [1,1,1] level suffered a high damage caused by a phenomenon similar to a light inner explosion when opening the press. The layers delaminated because of the excessive internal pressure produced by gases from wood volatilisation during hot-pressing. The colour became extremely dark, and the impression was of charcoal rather than heat-treated wood.

Model analysis

The statistical analysis of the data was done using the coded levels of the parameters. As suggested by Montgomery (2005), a model based on coded levels has the capacity to reveal the real weight of a predictor in a process, regardless of the measurement unity. Minitab was used to estimate the assumed full second-order model. When all the obtained 40 runs were used, the analysis of variance revealed a lack-of-fit for the assumed model. The P- value for Lack-of-fit was less than 0.0005, which means that the model needs to be improved. Instead of trying to fit a more complicated model, an investigation was done to see if the model was unsuitable for any specific parameter setting. By studying the diagnostic measure DFITS, which represents a scaled difference between the fitted values calculated with and without the i th observation, it was found that for the parameter setting [-1, -1, -1] both observations were influential, with DFITS 1.24 and 1.36 respectively. This means that by removing the observations in [-1, -1, -1], the model might be changed considerably. The parameter setting [-1, -1, -1] is extreme in the sense that all three parameters are kept at the lowest setting. Furthermore, this setting implied very low values (36.53 and 39.30) of bending strength. The analysis was redone without the two observations corresponding to the parameter setting [-1, -1, -1]. It turned out that the estimated second order model based on 38 runs showed no significant lack-of-fit (P-value = 0.23). Furthermore, the square of the parameter pressure did not contribute significantly (P-value = 0.72) and was removed from the model. Also, the pressure itself was non-significant (P-value = 0.86). It was kept in the model due to the hierarchy principle, since the interaction between pressure and time as well as between pressure and temperature was highly significant. The estimated coefficients, together with the corresponding estimated standard errors, T-values and P-values for the final model are shown in table 2. This estimated model shows an even larger P value for lack-of-fit (P-value = 0.32), and has $R^2 = 89.8\%$ and $R^2_{adj} = 87.0\%$. As is seen from table 2, the final model proposed for bending strength as response contains all the linear and the interaction terms plus the quadratic terms of temperature and time.

Table 2 The estimated coefficients, together with the corresponding estimated standard errors, T-values and P-values for the proposed model for the bending strength

Term	Coef	SE Coef	T	P-value
Constant	158.074	4.910	32.193	0.000
temp	73.394	5.596	13.116	0.000
pressure	-1.212	5.596	-0.217	0.830
time	16.520	5.596	2.952	0.006
temp*temp	-40.484	8.394	-4.823	0.000
time*time	-15.444	8.394	-1.840	0.076
temp*pressure	-33.086	6.502	-5.089	0.000
temp*time	-46.801	6.502	-7.198	0.000
pressure*time	-57.081	6.502	-8.779	0.000

A residual analysis of the final model shows that the normality assumption is reasonable (see Fig. 3a). When plotting the standardized residuals against the fitted values, a tendency of nonconstant variance can be seen (see Fig. 3b). However, there is no funnel shaped pattern indicating an easy way to improve the model. Figure 3a also reveals that there are three standardized residuals outside the interval $[-2, 2]$, where two of them belong to the centre point and one to the setting $[1, 0, 0]$

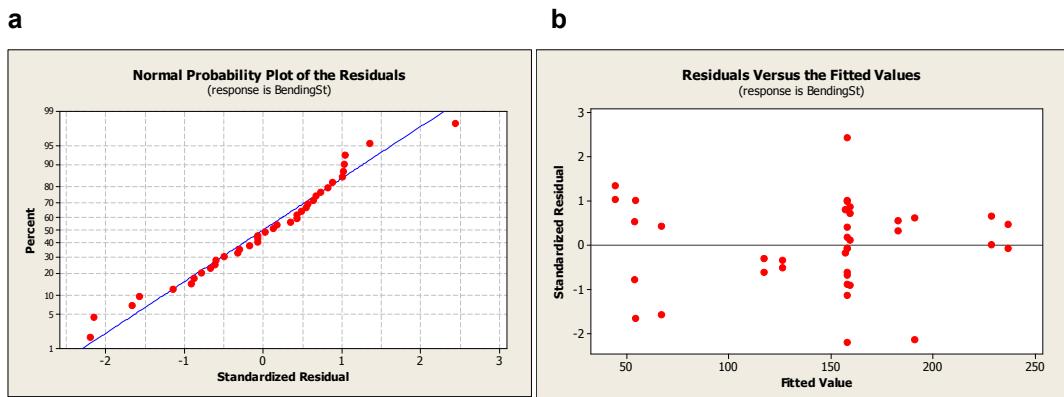


Fig.3 The standardized residuals from the final model plotted in a normal probability plot (a) and against the fitted values (b)

A plot of the predicted values, (fitted values), $\hat{y}$, versus the observed values, y , confirms that the model is acceptable see. In figure 4, the line $\hat{y} = y$ has been added.

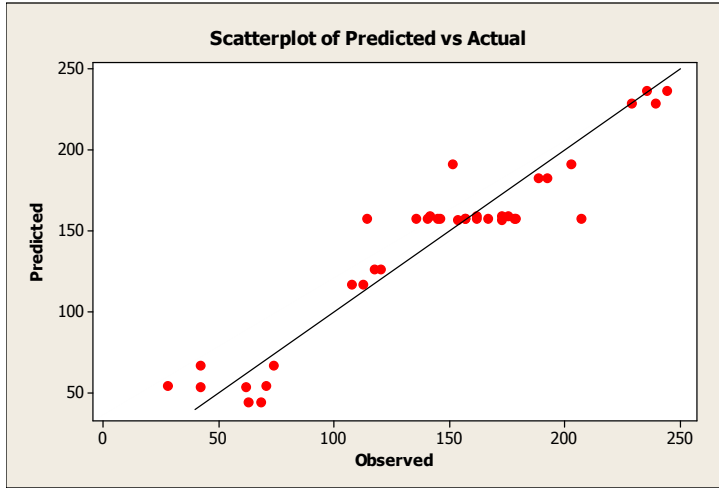


Fig.4 Scatter plot of the predicted values, $\hat{y}$, against observed values, y , of the bending strength.

The line shows $\hat{y} = y$

The results can be regarded as satisfactory. Hence the estimated regression model, in the coded levels x_i of the parameters, and using the coefficient in table 2, is:

$$y = 158.074 + 73.394 \cdot x_1 - 1.212 \cdot x_2 + 16.520 \cdot x_3 - 40.484 \cdot x_1^2 - 15.444 \cdot x_3^2 - 33.086 \cdot x_1 \cdot x_2 - 46.801 \cdot x_1 \cdot x_3 - 57.081 \cdot x_2 \cdot x_3 \quad (3)$$

The estimated model is derived without the observations in the setting $[-1, -1, -1]$ and hence is not valid for this case. For the setting $[-1, -1, -1]$, the model in (3) will predict the negative value -123.5 , which, of course, is unrealistic. One way to handle this problem is to say that whenever the model in (3) gives a negative value, the corresponding predicted value is set to 0. This is how it is handled in the figures below.

The scatterplot that compares the average of the observed values and the predicted values for each of the parameter settings shows how similar these groups of values are. It is remarkable how well the model covers especially the interval 225–250°C, and that it predicts the descending trajectory towards the extreme [1, 1, 1] (Fig.5).

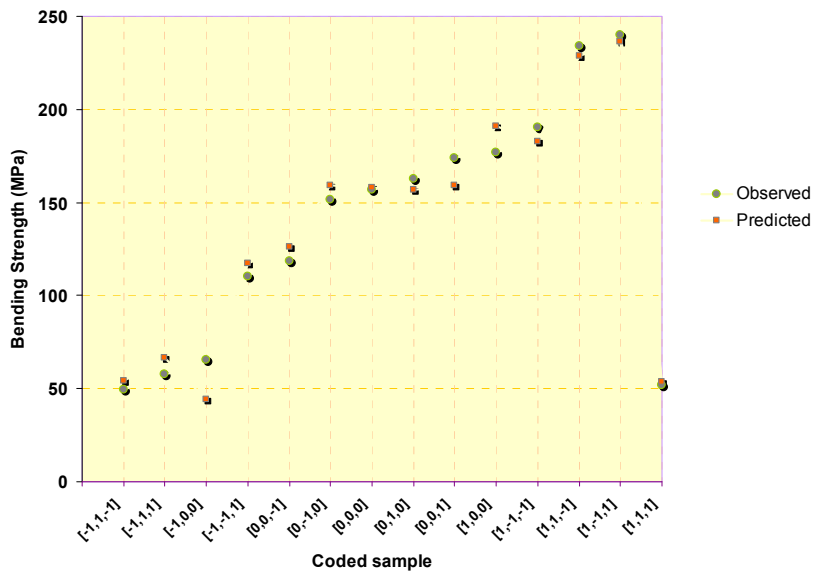


Fig.5 Comparison between the predicted values and the averages of the observed values

Visualising the estimated model

The estimated model in (3) was used in Matlab to visualise how, for a fixed level of a parameter, the other two parameters affect the responses. When interpreting the estimated model in (3), one should keep in mind that it is an estimate of the average performance of the response for given values of the parameter levels. The twists of the surfaces seen in figure 6 below reflect how influential the interactions are.

The best responses are shown in the red-coloured area in figure 6. This optimum area can be reached by combining either a high pressure (6 MPa) with a short pressing time (240 s) or a low pressure (4 MPa) for a longer pressing time (360 s).

When temperature is maintained constant at 200°C, there are small chances to obtain a strength higher than 100 MPa (Fig. 6a) even under a high level of pressure (6 MPa for 360 s). The medium level for temperature can give a good bending strength, above 150 MPa, if at the same time pressure and time are kept at suitable levels.

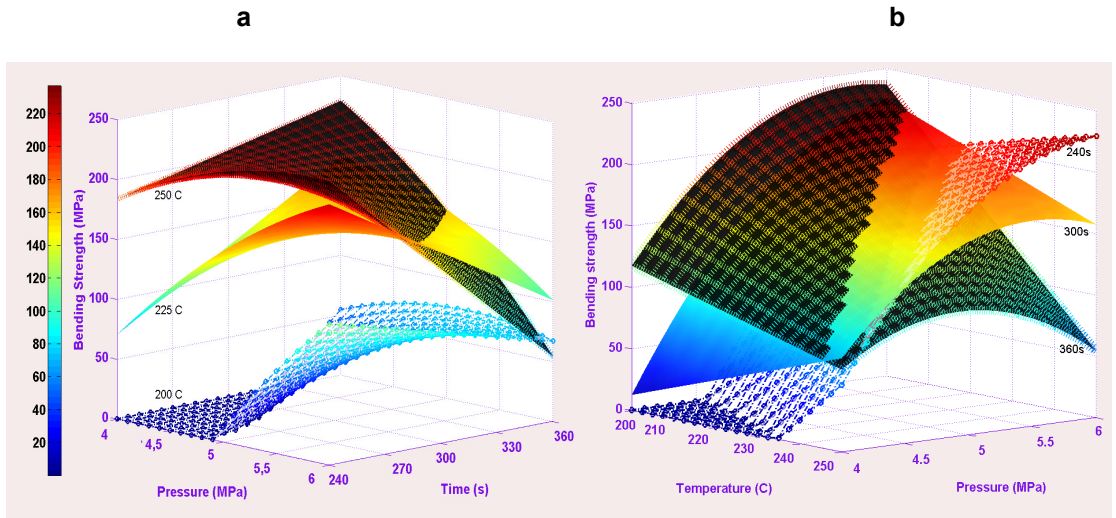


Fig.6 Bending strength behaviour for fixed levels of temperature (a) and time (b)

The plate temperature should be above 225°C when pressure is at least 4 MPa and time at least 240 s, and the explanation is in the chemical changes that must take place in the wood's structure in order to create a good autoadhesion.

According to the derived estimated model, it was found that when the temperature of the press-plates is 250°C, the pressure is 4 Mpa, and the pressing time is 360 s, the expected bending strength is between 209 and 265 units, with 95% confidence.

This strong interaction between time and pressure seen in Fig. 6a and demonstrated by the similar shapes of the surfaces in Fig. 6b might be expressed through a new parameter: the inner temperature within sample. But this parameter could also involve the plate temperature as well. The surfaces of the sample together with the exterior veneers will reach the plate temperature very soon after being in contact. On the contrary, the inner layer is prevented from heating up by the outer layers, which will behave more like

an insulator, especially after water evaporation. When the pressure is higher, the outer layers will be more compressed, their thickness will be reduced, thus allowing the heat to be transferred faster to the inner layer. At the same time, it is obvious that the higher the plate temperature, the shorter the time to heat up the inner layer within the veneer pack. It was actually shown that pressing as briefly as 80 seconds is enough to produce a good adhesion if the plate temperature is 300°C (Cristescu 2006).

Failure behaviour

The 40 samples did not behave in the same manner with regard to the type of failure. Fig. 7 shows two samples, one with the lowest and the other with the highest response.

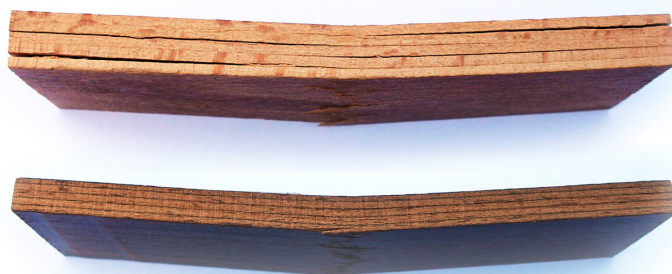


Fig. 7 Images of a sample pressed at 200°C, 4 MPa, 240 s (*top*) and 250°C, 6 MPa, 240 s (*bottom*)

At a molecular level, the arrangement of the entire network within the sample is disturbed during load application. The molecules forming polymer chains are pushed, which implies movements and attempts at refinding a place in the network. The three-point bending tests develop vertical shear stresses, compression at the top and tension at the bottom of samples, plus horizontal shear stress as well (Bodig and Jayne 1982).

The type of failure is caused by the lowest strength of the sample to one of the mentioned stresses, which is actually related to the microscopical structure of wood and the chemical bonds between molecules.

The [-1, -1, -1] sample (Fig. 7 top) had the lowest resistance to horizontal shear stress because of the weak bonding between the molecules of the adjacent veneer layers. This

low temperature- pressure-time combination was not strong enough to produce the necessary chemical changes and

at the same time to bring the surface cells belonging to neighbour layers close enough together.

By way of contrast, the [1, -1, 1] sample (Fig. 7 bottom) shows a failure caused by simple tension combined with cross-grain tension. Most of the well-bonded samples (with a maximum bending force exceeding 2000 N) exhibit this combined type of rupture. This occurs in the outer layer.

An important observation is that veneers developed dark lines at the bonding areas, as seen in Fig. 7. A visual assessment reveals that this colouring within samples at veneer surfaces shows the same intensity as the exterior sample surface colour (that had been in direct contact with the press plates) and the same intensity as the wood rays' colour after compression. This browning effect is a sure sign of higher density, as reported before (Cristescu 2006) and might actually be the key to the bonding phenomenon. Some explanations for the dark lines found in literature are:

- the low-molecular soluble compounds' migration during solid wood drying (McGinnes and Rosen 1984; Theander et al. 1993; Terziev et al. 1995). Beech contains a large amount of low molecular sugar units (Zula 2003).
- caramelisation of water-soluble content—especially sugar units—carried out towards surfaces by vapours—considered the reason for wood particle adhesion to the press plate during the semi-dry fibreboard production (Elbert and Dorkhova 1983)
- in the food industry, browning is directly related to the amount of hydroxymethylfurfural formed by degradation of monosaccharides during the baking of cookies (Ameur et al. 2006)
- in paper industry, heat-induced colouring of bleached chemical pulp is connected to the decay of the unstable polysaccharide chain such as hemicelluloses to low molecular carbohydrate compounds that subsequently undergo dehydration and condensation reactions to form coloured products (Beyer et al. 2006)
- Shafizadeh (1984) proved that heating wood below 300°C leads to reduction in the degree of polymerisation by bond scission, elimination of water, formation of free radicals, carbonyl, carboxyl groups and finally production of highly reactive carbonaceous char. Inari et al. (2007) shows that char formation begins for beech at temperature above

200°C, and degradation of beech holocellulose does not result solely in volatile byproducts, but in newly formed carbonaceous materials as well. The creation of new carbon-carbon bonds during pyrolysis of lignin has also been reported (Scholze et al. 2001)

- during wood thermal treatment the lignin-polysaccharide complex is cleaved by organic acids released from hemicelluloses and secondary lignin-carbohydrate linkages are formed (Košíková et al. 1999)

- high temperature produces homolytic cleavages in lignin (Kawamoto et al. 2008). The free radicals might undergo coupling reactions leading to chromophore formation (Westermarck et al. 1994)

The relation between high temperature, darkening and wood fusion needs further research.

Conclusions

Autoadhesion between 5 layers of 2.2-mm-thick beech veneer under high pressure and temperature treatment resulted in laminated products with high bending strength performance.

In order to obtain a model not showing lack-of-fit, it was necessary to exclude the observations for the setting [-1, -1, -1]. However, as the results show, the setting [-1, -1, -1] is not close to the region that will give a maximum bending strength, and hence this action does not affect the conclusions. Removing a design point will, on the other hand, affect the statistical properties somewhat negatively. In future experimentation, a rotatable CCD design will be chosen instead in order to avoid problems with extreme settings like [-1, -1, -1].

The statistical surface design analysis revealed that all three parameters influence bending strength in a rather complicated way, which is caught by the interactions. Samples responded differently because of the different fusion quality. A temperature of 200°C is not sufficient for a good product, although it seems that veneers are bonded. At 225°C plate temperature, bending strength improves considerably, especially if pressing time is longer. 250°C induces the highest strength, if combined properly with pressure

and temperature.. A new parameter, internal temperature within board, will be studied in future experiments. Further research on the relation between the strong darkening of the bonding areas and the fusion phenomenon is required. The effect of high temperature on generating and coupling free radicals deserves attention as well.

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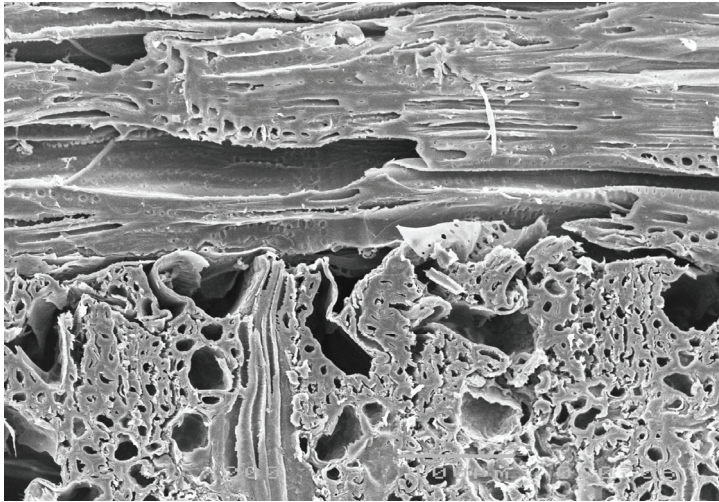
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... an academic step halfway to the
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about the subject ...

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