



**DETERMINING THE RESISTANCE
TO DELAMINATION OF WOOD
ADHESIVES USING A VACUUM
PRESSURE IMPREGNATION
METHOD**

Christopher Mills

Bachelor's Thesis
April 2013
Degree Programme in
Laboratory Science

TAMPEREEN AMMATTIKORKEAKOULU
Tampere University of Applied Sciences

ABSTRACT

Tampereen ammattikorkeakoulu
Tampere University of Applied Sciences
Degree programme in Laboratory Science

MILLS, CHRISTOPHER:

Determining the Resistance to Delamination of Wood Adhesives using a Vacuum Pressure Impregnation Method

Bachelor's thesis 56 pages

April 2013

The thesis was completed for the Research and Development (R&D) laboratory at Kiilto Oy. The purpose of the thesis was to study the delamination of glued solid timber and glulam, glued with moisture cured one component polyurethane adhesives (1K PUR) and an emulsion polymer isocyanate adhesive (EPI) using the new easyQ DLA appliance. Delamination is the failure of the adhesive layer between wood lamellae, used to produce glued solid timber and glued laminated timber (glulam). Delamination tests are accelerated weather exposure tests which aim to simulate the stresses over the lifetime of a glued timber structure.

From July 2013, glued solid timber and glulam produced in Finland has to be CE-marked. This means production must be covered by the harmonized EN 14080 standard, which includes the quality control standard EN 391 as well as the adhesive evaluation standard EN 302-2. Currently, 1K PUR adhesives can be used for glued solid timber and glulam production. In the final draft of the FprEN 14080:2013 standard, EPI adhesives can also be used, though only for service class 1 & 2 applications.

Parallel EN 391 quality control samples, provided by Kontio, were tested in two different drying chambers and four different conditions. The objective was, to evaluate the need for a drying chamber to accompany the delamination appliance, as well as compare results with Kontio. A method following EN 302-2 as closely as possible was developed for the second objective of comparing R&D formulated adhesives.

The testing of parallel quality control samples resulted in higher delamination percentages at Kiilto when compared with tested at Kontio. Of the 36 parallel samples tested, 35 achieved accepted status at Kiilto and 36 at Kontio. Out of the adhesives tested with the modified EN 302-2 method, two samples passed the test while 6 failed. The samples that passed were the short closed assembly time commercial 1K PUR sample and the short closed assembly time sample for an experimental 1K PUR formulation.

The results from comparing quality control samples from Kontio show that drying conditions affect EN 391 results greatly, and thus there is a need for a specific drying chamber. The comparison of adhesives formulated in the laboratory showed the importance of assembly time for good bonding performance and resulted in a possible new method for determining assembly time. In addition, the performance of adhesives could be compared, using instructions prepared for the thesis. The results for EPI were mixed, and require further study on the causes.

Key words: delamination, glued solid timber, glulam, 1K PUR, EPI

TIIVISTELMÄ

Tampereen ammattikorkeakoulu
Laboratorioalan koulutusohjelma

MILLS, CHRISTOPHER:

Puuliimojen delaminointivastuksen määrittäminen käyttäen alipaine – paine kyllästys menetelmää

Opinnäytetyö 56 sivua
Huhtikuu 2013

Opinnäytetyön tavoitteena oli tutkia lamellihirren sekä liimapuun delaminoitumista. Opinnäytetyö tehtiin Kiilto Oy:n tuotekehitysosastolle. Työn tarkoituksena oli uuden easyQ delaminointilaitteen käytön aloittaminen, sekä uusien testausmenetelmäohjeiden kehittäminen. Delaminointilaitteen avulla voidaan simuloida sään aiheuttamaa pitkän ajan kosteusrasitusta rakenteellisissa puutuotteissa. Suomessa heinäkuussa 2013 voimaan tuleva lainsäädäntö edellyttää, että kaikki lamellihirsi- ja liimapuutuotanto on CE-merkittyä. Jotta rakennustuote voidaan CE-merkitä, on siinä käytettyjen liimojen oltava EN standardien mukaisia.

Työn tarkoituksena oli vertailla Kiilto Oy:n ja Kontion laadunvalvonnassa saamia testituloksia ja arvioida saatujen tuloksien perusteella mahdollisen kuivauskaapin tarvetta. Vertailu tehtiin delaminoimalla tuotantokappaleita standardin EN 391 mukaisesti yhteistyössä Kontion kanssa. Lisäksi työssä vertaillaan rinnakkain neljää Kiilto Oy:n valmistamaa liimaa. Kolmea kosteuskovettuvaa polyuretaaniliimaa (1K PUR) ja yhtä kaksi-komponenttista emulsiopolymeeriisosyanaattiliimaa (EPI) vertailtiin menetelmällä, joka pyrkii noudattamaan standardin EN 302-2 laboratorimenetelmää Kiilto Oy:n olosuhteissa.

Rinnakkain tehdyssä laadunvalvonnassa Kontion delaminointitulokset olivat kautta linjan matalampia. Kuivattaessa palat 65 °C:n lämpötilassa ja 9 %:n suhteellisessa kosteudessa erot olivat pienempiä. Kaiken kaikkiaan Kontiolla läpäisivät kaikki palat, ja Kiillolla 35 palaa 36:sta. Vain kaksi kahdeksasta laboratorimenetelmällä tehdyistä näytteistä saavuttivat hyväksyntärajan.

Kiillolla valmistettujen liimojen keskinäisessä vertailussa selvisi, että koontiajalla oli erityisen suuri merkitys 1K PUR-liimojen delaminointitulokseen. EPI-liiman tuloksista samaa ei voitu päätellä. Eri kuivausolosuhteiden tulosten perusteella kuivauskaapin hankinta olisi suositeltavaa. Jatkossa liimojen delaminointivastuksen tutkimista jatketaan easyQ DLA laitteella.

Table of Contents

1	INTRODUCTION.....	5
2	BACKGROUND.....	7
2.1	Adhesives used in the study.....	7
2.1.1	One-component moisture-cured polyurethane adhesives	7
2.1.2	Emulsion polymer isocyanate adhesives.....	10
2.2	Wood composition.....	12
2.2.1	The macro structure of wood lamellae.....	12
2.2.2	Longitudinal tracheid cell structure	13
2.3	The chemical structure of the wood surface	14
2.3.1	Cellulose.....	15
2.3.2	Hemicellulose.....	16
2.3.3	Lignin	16
2.3.4	Extractives.....	16
2.4	Theories of adhesion with adhesives	17
2.5	Glued solid and laminated timber	21
2.5.1	The manufacturing process of glulam.....	22
2.6	Delamination tests of glulam and glued solid timber	24
2.7	Standards and certifications for glulam and glued solid timber	26
2.8	Development of new adhesives	28
3	MATERIALS AND METHODS	29
3.1	EN 391 method B	29
3.1.1	EN 391 method B sample preparation	30
3.1.2	SFS EN 391 method B vacuum-pressure soak	30
3.1.3	Evaluation of EN 391 method B samples	33
3.2	EN 302-2:2004 laboratory method	36
3.2.1	Moisture-cure one component polyurethane adhesive formulation.....	36
3.2.2	Modified SFS EN 302-2:2004 method	37
4	RESULTS.....	42
4.1	Evaluation of delamination.....	42
4.2	SFS EN 391:2001 method B results	43
4.2.1	Results in different drying conditions	43
4.2.2	Comparison with Kontio quality control results	45
4.3	Kiilto test method (modified SFS EN 302-2 test method) results	48
5	CONCLUSIONS AND DISCUSSION.....	51
	REFERENCES.....	54

1 INTRODUCTION

Kiilto Oy is a Finnish chemical company based in Lempäälä, Finland. Kiilto specialises in providing preparation, bonding and surface coating products (Kiilto Oy 2013). This thesis was completed in the research and development laboratory of Kiilto Oy. Research and development is important for Kiilto, which aims to receive 20 % of annual sales for each year from new products. The research and development laboratory at Kiilto focuses on creating tailor-made, environmentally sustainable, efficient solutions for customers.

The background purpose of the thesis is to study the delamination of glued solid timber and glued laminated timber. Kiilto has invested in an easyQ DLA vacuum-pressure delamination appliance, used to simulate stresses created in glued solid and laminated timber exposed to weather over a long period of time. Delamination is the adhesive failure between layers of bonded wood. From 1.7.2013, a CE-mark will become compulsory for building products. For manufacturers to brand their glued solid and laminated timber products as CE - certified, the adhesive bonds must pass delamination quality control standard SFS EN 391 included in the standard SFS EN 14080:2005, which also includes the delamination laboratory method standard SFS EN 302-2 for certifying adhesives.

The first objective of the thesis is to provide usage instructions for the easyQ DLA delamination appliance for assessing quality control specimens produced in a glued solid or laminated timber factory, as well as evaluate the need for a drying chamber to accompany the device. The aim is to use parallel SFS EN 391 method B test samples from the quality control process at Kontio, a glued solid timber factory located in northern Finland. Kontio is one of the largest producers of log cabins, built from glued solid timber, in the world (Kontio, 2013). Kontio kindly agreed to supply quality control samples from their normal production for this objective. The results would provide a starting point for comparing customer results to the new results from Kiilto. The second objective of the thesis is to create a method for comparing the delamination performance of adhesives available or formulated in the research and development laboratory at Kiilto, and use it to compare the performance of three single component moisture curing polyurethane adhesives and one emulsion polymer isocyanate adhesive. The

method is needed to study the effect of different adhesive formulations on delamination test performance for development and certification purposes. For this reason the aim is that the method should follow SFS EN 302-2 as closely as possible.

2 BACKGROUND

Wood as a building material is light, strong and environmentally friendly. However, using solid wood for buildings is not efficient, or economically viable, because of the long time needed to grow large enough trees for large structures. The forestry industry is very efficient in using all the parts of the trees, but adhesives are needed to fully exploit the potential of wood as a construction material. Glued solid or laminated timber when made with a certified adhesive is stronger and much more versatile in shape than solid timber. The chemical composition of wood, however, presents bonding challenges as it is heterogeneous. New wood adhesives are being developed constantly around the world. The main advancements in wood adhesives have been in increasing water resistance, environmentally friendly components, shorter pressing times, easier production and innovative new applications for adhesive products. As safety is important when constructing buildings, there are strict requirements on adhesives used to manufacture glulam, as well as strict quality control requirements to ensure product quality.

2.1 Adhesives used in the study

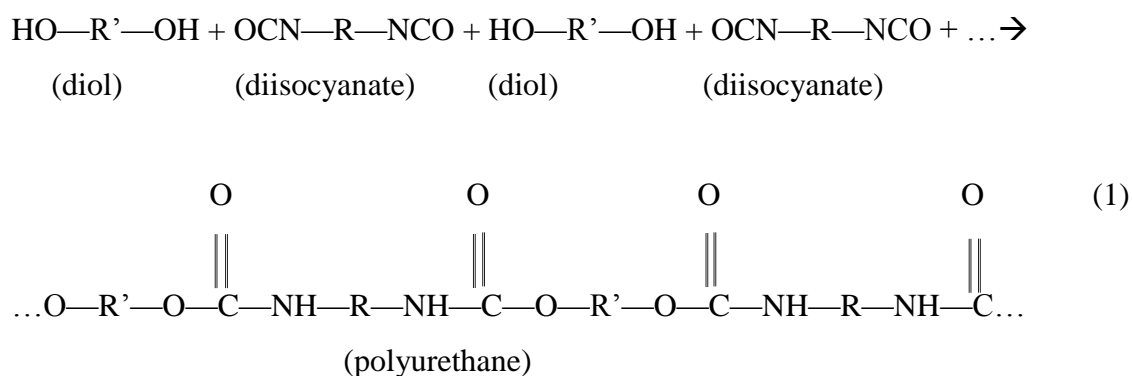
The samples that were used for comparing Kontio's quality control with the easyQ DLA appliance EN 391 method B were bonded with a commercial moisture-cure polyurethane adhesive. The adhesives compared using the created test method were one commercial moisture-cure polyurethane formulation, two experimental polyurethane formulations and one commercial emulsion polymer isocyanate adhesive. The experimental adhesives chosen for the study were formulated in the research and development laboratory from commercially available raw materials.

2.1.1 One-component moisture-cured polyurethane adhesives

One-component moisture-cured polyurethane adhesives (1K PUR) were first used for gluing glulam in 1985. They belong to the polyurethane family. 1K PUR adhesives have since their first use gradually received acceptance for gluing load bearing structures, with the first adhesive receiving certification in the 1994. 1K PUR adhesive ad-

vantages are: no mixing, easy application, low coating weight, no formaldehyde, clear or wood colored weather resistant bond lines. (Wang 2012.)

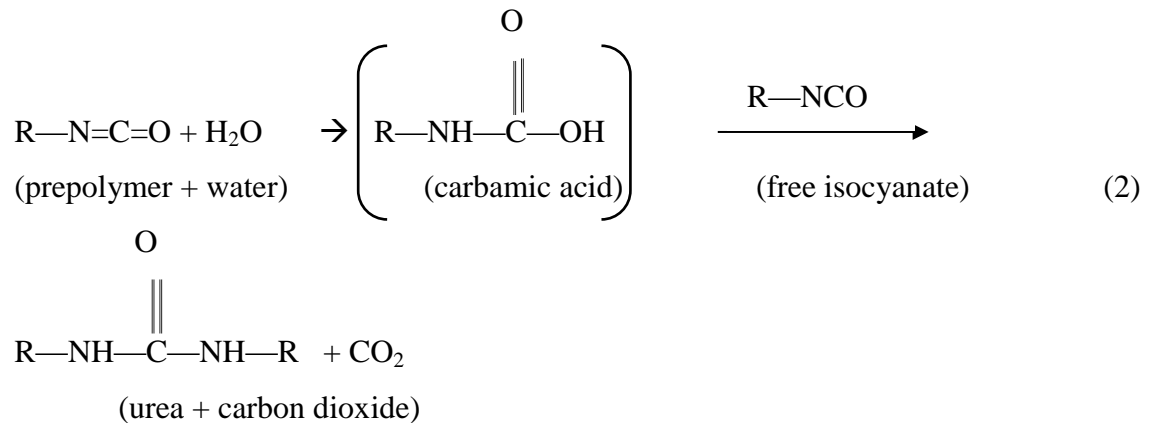
Dr. Otto Bayer, working for I.G. Farben, discovered basic polyurethane chemistry in 1937. The first polyurethane was formed by reacting 1,8-octane diisocyanate with 1,4-butanediol, which formed the ester of a carbamic acid, commonly now known as a urethane or PU or PUR. Polyurethanes are formed when a diisocyanate or polyisocyanate is reacted with a diol or polyol. (Szycher 2012, 1.) An example of polyurethane synthesis is shown in equation (1) from Öertel (1985, 11).



The reaction in equation (1) is called a polyaddition reaction, and the variation of the di- or polyisocyanate (R) and di- or polyol (R') components can form any imaginable number of polyurethane products (Öertel 1985, 11). The Polyols used in polyurethanes are hydroxy functional macro molecules. Polyethers are the most important molecules used, but also polyester or any combination can be used. The molecular weight varies from 250-1000 for highly branched polymers to 1000-8000 for linear or slightly branched polymers. The hydroxyl nature, primary or secondary, as well as its hydroxyl functionality and molecular weight are important in determining the properties of the polymer. (Ulrich 1996, 399.)

The most important component of a moisture cured 1K PUR adhesive is called a NCO-prepolymer. A NCO-prepolymer is formed by the polyaddition reaction described in equation (1), but with using an excess molar amount of the di- or polyisocyanate, to create a polymer with reactive NCO groups at each end of the polymer chain (Öertel 1985, 20). A common di-isocyanate used for prepolymer production is MDI, or methyldiphenyl isocyanate. When a NCO-prepolymer comes into contact with water, usu-

ally from atmospheric humidity or water in the substrate to be bonded, the reaction in equation (2) taken from (Öertel 1985, 8) occurs.



In equation (2) the NCO-prepolymer reacts with water, forming a carbamic acid that is unstable. Carbon dioxide is split, and an amine is formed. The amine then reacts with isocyanate remaining in the NCO-prepolymer, forming a urea. The excess isocyanate in the prepolymer serves to crosslink the polyurea or polyurethane that is formed forming a poly (urea urethane) film. Carbon dioxide also acts to form bubbles, which can evaporate if the film is thin, however, in thick films the carbon dioxide creates a foamy structure. (Öertel 1985, 8-9.) The main morphological feature of a 1K PUR film is the separation into hard and soft segments. The hard segments are formed from the di- and polyisocyanate sections and the soft segments are formed from the polyol sections. (Ren 2010, 19.)

In addition to the prepolymer, 1K PUR adhesives can contain various solvents, fillers, extenders, defoamers and catalysts. These can change the final properties of the adhesive, modifying the viscosity and mechanical properties such as elongation at break of the adhesive. In addition, fillers can be used to improve the heat resistance of the adhesive. A common filler used is calcium carbonate (CaCO_3). In general, unfilled 1K PUR adhesives are used for bonding glulam. Catalysts are used to adjust the curing rate of the adhesive which in turn adjusts the pressing time required for a good adhesive bond. The composition of 1K PUR adhesives greatly affect the bonding behavior and delamination resistance of adhesively bonded members.

2.1.2 Emulsion polymer isocyanate adhesives

Emulsion polymer isocyanate (EPI) adhesives are two-component dispersion and isocyanate adhesive systems. Because the dispersion component is thermoplastic and the isocyanate hardener thermosetting, EPI adhesives have versatile properties. The two components of EPI adhesives are stored separately but mixed just before use. EPI adhesives were first developed in Japan in the 1970's and named aqueous polymer isocyanate adhesives (API), and have spread to Europe and the US over the past 15 years named EPI adhesives. The components are a water based polymer dispersion and an isocyanate hardener. Common polymers used in the dispersion are: polyvinyl alcohol (PVA), polyvinyl acetate (PVAc), ethylene vinyl alcohol (EVA), vinyl acetate-acrylate copolymerized emulsion (VAAC), acrylic-styrene emulsion (AcSt) and styrene-butadiene latex (SBR). In addition to polymers, the dispersion can contain fillers, dispersion agents, extenders, defoamers and anti-fungal agents as well as water. Fillers can be CaCO_3 , wood flour, talc, clays, silica's and shell flours. Fillers affect the viscosity and final properties, including wear on planing tools which is an important woodworking consideration. (Grøstad & Pedersen 2010, 1358-1360.)

The isocyanate portion is usually Polymeric MDI (pMDI). Polymeric MDI is a byproduct of pure MDI production. The chemical structure of MDI is shown in figure 1. MDI consists of two phenyl groups with two reactive isocyanate groups providing a functionality of 2. The MDI in figure 1 is the 4,4' isomer, with the isocyanate attached opposite to the methylene and is also known as pure MDI. Polymeric MDI consists of a mixture of about 50% pure MDI (2,2', 2,4' and 4,4' isomers) and the other 50% is comprised of oligomeric forms of isocyanate with a high functionality of 2,5-2,7. (Ulrich 1996.) Polymeric MDI is a low viscosity liquid and contains about 30% isocyanate (Ren 2010, 14). Because of its high functionality, pMDI is an effective crosslinker. A new type of pMDI is called emulsifiable pMDI (EPMDI). EPMDI is formed by attaching a hydrophilic chain to pMDI, making it water soluble. EPMDI could be particularly useful in EPI adhesives to provide easy and even mixing of the two components which is important for good bond results.

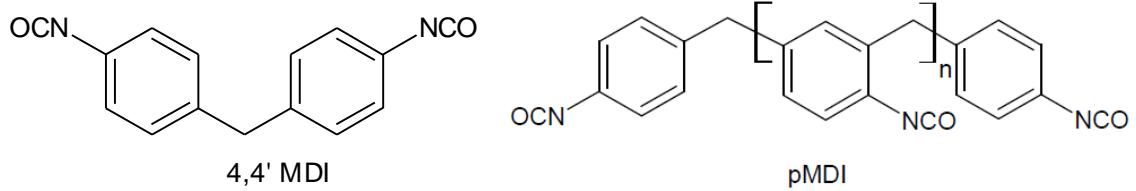


FIGURE 1. The chemical structure of 4,4' MDI and pMDI (Greenberg & Sekizawa 2000 edited)

EPI adhesives differ from 1K PUR adhesives in that they need to be mixed before use. The EPI components are mixed either manually or mechanically, before application on to the surface to be bonded. The mixing ratio for an EPI adhesive is usually 100 parts dispersion and 5-20 parts isocyanate cross-linker. The two components are stored separately, which allows them to have a long shelf-life. The curing of an EPI adhesive occurs gradually, with the isocyanate hardener acting as a cross-linker and an adhesive film of cross-linked polymers coalescing. Several reactions are taking place simultaneously during the curing process. Water is removed from the bond line by evaporation and capillary action into wood. The emulsion is coalescing to form a film of the polymer particles. The isocyanate in the hardener reacts with water, PVA, hydroxyl groups on the wood surface, hydroxyl groups in the emulsion and itself. These reactions form urethanes and biurets. After mixing, the EPI adhesive has a pot life, which is dependent on the components used and the ratio of the mixed adhesive. A good quality EPI bond line is formed when the adhesive is used within its pot life, which can be checked via testing bonded assemblies using the EN 302-2 standard. (Grøstad & Pedersen 2010, 1361-1364.)

EPI adhesives can be cured at low temperatures, which is an advantage. In addition, EPI adhesives have good heat and moisture resistance. The dried bond line is light colored, which is important for aesthetic reasons. The viscosity can be adjusted to be used with contactless finger-jointing machinery, and the curing time is adjustable by variation of the composition. EPI adhesives are currently used in laminated solid wood panels, window frames, veneer boards, plywood, parquet flooring, and glulam and glued solid timber. In addition, EPI adhesives attach well to metals, making it an ideal adhesive system for composite structures. (Grøstad & Pedersen 2010, 1370-1378.)

2.2 Wood composition

Successful bonding of wood lamellae into glulam is dependent not only on the adhesive used, but also on the wood substrate being bonded. Wood in general is composed of wood polymers, extractives and moisture. The wood surface being bonded is polar, anisotropic, hygroscopic and has a heterogeneous surface. Scots pine and spruce that are used for glued solid timber and glulam are both coniferous, softwood wood species. (Ren 2010, 3; Frihart 2005, 216,225; Forest.fi 2012; Rowell 2005, 11.)

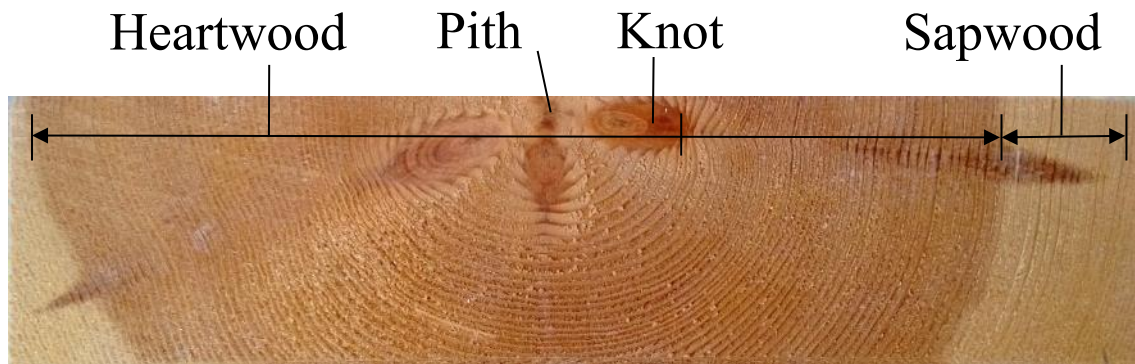
2.2.1 The macro structure of wood lamellae

Wood lamellae are made up of two main zones, sapwood and heartwood. Sapwood is lighter in color, while heartwood is darker. Both sapwood and heartwood are characterized by earlywood and latewood sections, which are known as annual rings, and can be seen as alternating dark and light stripes. Annual rings in Scots pine can be 0,1 mm to 10 mm apart, depending on the speed of growth. Other macroscopic features which can be seen are the pith, resin canals, knots, rays, and reaction wood known as juvenile-, tension and compression wood. The cells of coniferous softwoods are made up of 90-95% longitudinal tracheid cells, and 5-10 % transversal ray parenchyma cells. (Sjöström 1981, 1-20.)

Sapwood is defined as the part of the tree which is metabolically active. Sapwood has several functions that affect its composition. Sapwood provides transport of water and nutrients, it conducts sap and also creates and stores biochemicals such as starch and lipids (Rowell, 2005, 12; Sjöström 1981, 2). Sapwood is generally considered easier to bond than heartwood, as it is more polar and contains fewer extractives than heartwood (Frihart 2005, 225).

Heartwood is formed from sapwood cells into the center of a tree trunk. Heartwood is made up of dead cells, filled with biochemically produced extractives. The extractives give heartwood its typical darker color as can be seen in picture 1. These extractives are produced by sapwood cells in a layer around the heartwood, and deposited for storage. Because of the high extractive content in heartwood compared to softwood (Sjöström 1981, 91), heartwood is less polar and harder to bond (Frihart 2005, 226).

Phenolic compounds in scots pine heartwood give protection against biological attack. In particular decay resistance can be linked to the stilbene content. (Verta et al 2008.)



PICTURE 1. Several macroscopic features of Scots Pine

Earlywood is formed in the spring and summer in the northern hemisphere. Latewood is typical of autumn and winter growth. Earlywood is characterised by thin walls and large cavities in the tracheial cells. Cavities and pits are important for transport between cells. Latewood has thicker cell walls and less cavities, which gives it a higher density and darker color. (Sjöström 1981, 4-8.) The smaller amount of cavities and thicker cell walls of latewood cells makes it harder to bond, because the adhesive cannot penetrate easily to the lumen which is the central part of dead wood cell (Frihart 2005, 227).

Resin canals are empty spaces in wood surrounded by parenchyma cells that produce resins. Pines contain larger resin canals that can be seen, whereas they are smaller in spruce. (Frihart 2005, 21-22). Resin canals or pockets typically contain resins that are difficult to bond and liquify in the high temperatures of the drying process of the delamination standards.

2.2.2 Longitudinal tracheid cell structure

A tracheid cell in Scots Pine and Norway spruce is generally between 1 and 4 mm in length, and 0,02 mm and 0,04 mm in width, which means it can be seen with the naked eye. The finer structure of a tracheid cell is shown in figure 2. The relative composition of cellulose, hemicellulose and lignin in each of the layers labeled in the figure: the middle lamella, primary wall, inner layer, middle layer and outer layer, are all different. The basic structure is similar in each layer. A layer is made up of cellulose microfibrils

providing the backbone, combined with a hemicellulose matrix and surrounded by lignin. The Inner- (S_3), Middle- (S_2) and Outer- (S_1) layers of the secondary wall differ in the orientation of the cellulose microfibrils as well as in thickness. (Missouri S&T 2012; Sjöström 1981, 12; Peura 2007, 9.)

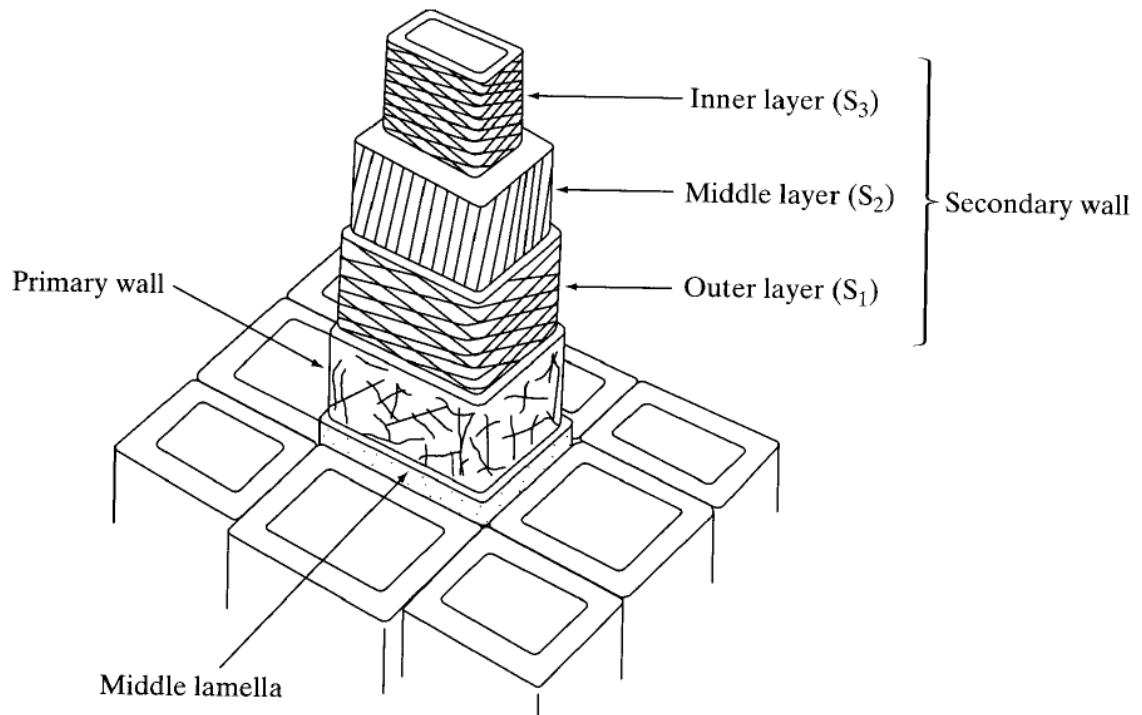


FIGURE 2. The layered structure of a softwood tracheid cell (Missouri S&T 2012)

2.3 The chemical structure of the wood surface

An important factor affecting bond quality in glued laminated timber is the surface of the wood being bonded. The underlying structure of the wood cell is made up of four main polymer types: cellulose, lignin, hemicellulose and extractives. These make up the structure of wood in varying percentages, depending on the species and growth conditions of wood. The distribution of polymers of different types for Scots Pine and Spruce are shown in table 1. In particular, Scots Pine has a higher percentage, 3,5 % of extractives, compared to 2,1 % in Norway spruce. Correspondingly the hemicellulose percentage is lower in Scots Pine, 28,5 %, while Norway spruce has 30,6 %.

TABLE 1. Distribution of the four main compounds in Scots Pine and Spruce (Sjöström 1993, 292)

Type of molecule	Scots Pine	Spruce
Cellulose	40,0 %	39,5 %
Hemicellulose	28,5 %	30,6 %
Lignin	27,7 %	27,5 %
Extractives	3,5 %	2,1 %

2.3.1 Cellulose

Cellulose is a homopolysaccharide made up of repeating units of D-glucose, which are bound together by β -1,4-glycosidic bonds. From the chemical structure in figure 3, it can be seen that cellulose has a large number of polar -OH groups. These polar -OH groups facilitate hydrogen bonding between cellulose chains, and this results in partial crystallization of the cellulose polymers. Cellulose in Norway spruce is 52 ± 3 % crystallized and the remaining portion is amorphous (Peura 2007, 9). Cellulose polymers group together to form fibers that are called microfibrils that are orientated in the growth direction (Ren 2010, 29-30). Cellulose in softwood can form polymers of up to 10,000 repeating units, such as the ones shown in figure 3 (Ragauskas 2013, 1).

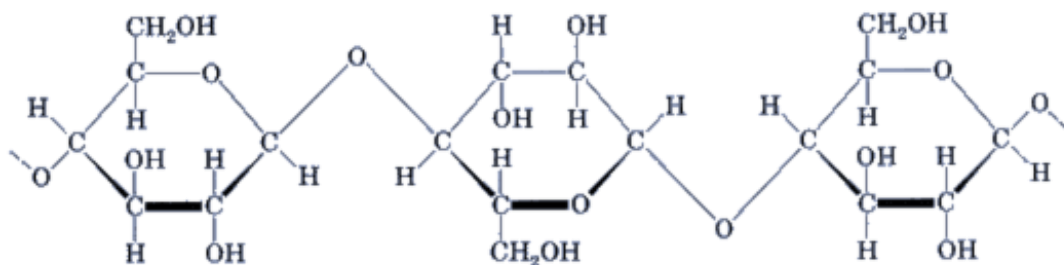


FIGURE 3. The chemical structure of cellulose (Senese 2010)

2.3.2 Hemicellulose

Hemicelluloses are heterogeneous polysaccharides, which are much smaller than cellulose, with a typical chain length of a few hundred units. The two main hemicellulose polymers in Nordic softwoods are galactoglucomannans and arabinoglucuronoxylan. They are amorphous, branched polymers. The main function of hemicellulose is to strengthen the cell wall. Hemicellulose is attracted to cellulose via Van der Waals forces and hydrogen bonding, and can covalently bond to lignin. (Sjöström 1993, 60-64.)

2.3.3 Lignin

Lignin is the substance that binds the cellulose and hemicelluloses together into a cell wall. Lignin has no repeating structure, unlike cellulose and hemicellulose. Lignin in softwoods is mainly made up of polymerization products of coniferyl alcohol with various conformations and functional groups. Lignin is found in large fractions in the middle lamella, which is in between the individual cells. There is a lower fraction of lignin in the secondary wall sections shown in figure 2, but the secondary wall is much thicker, resulting in larger total amounts. Lignin contains various functional groups such as carbonyl, benzyl alcohol, noncyclic benzyl ether, phenolic hydroxyl and methoxyl. (Sjöström 1993, 70-82.)

2.3.4 Extractives

Extractives that are formed in resin canals are called oleoresins. Resin canals are surrounded by parenchyma cells, which produce the oleoresins. Extractive content can differ in different parts of the same tree or species. Extractives can be grouped into aliphatic compounds, terpenes and terpenoids, and phenolics. Aliphatic compounds are comprised of different fats and waxes, an example found in wood is the fatty acid stearic acid. Terpenoids are molecules formed from 2-methylbutadiene that have the basic formula $(C_{10}H_{16})_n$. Phenolic compounds come in many forms. An example of an important phenolic extractive is pinosylvin, a stilbene that is found in pines which reacts readily. The extractive content varies according to location and conditions, and is high-

er in more northerly forests, like the ones from which the samples for this thesis were taken from. (Sjöström 1993, 83-97.)

2.4 Theories of adhesion with adhesives

Adhesion is the interaction of an adhesive surface with a substrate. Covalent, ionic and hydrogen bonds as well as dispersive forces such as the Van der Waals and London forces provide attraction. Dr. Pocius (1997, 118-146) in his book, *Adhesion and Adhesives technology*, provides a summary of six different theories concerning adhesion while noting there is no unified universal theory of adhesion. It is likely that such a theory would include components from all the theories of the summary.

1. The electrostatic theory is that an electronegative material will attract an electropositive material, forming an adhesive bond. This bond is due to the formation in the interface of an electrostatic bilayer, due to the electropositive material donating a charge to the electronegative material. During tests while in a vacuum, it has been shown, that breaking an adhesive bond resulted in electromagnetic discharges, providing support for the theory. In addition measuring with a force meter, a charge was found.
2. In the diffusion adhesion theory, adhesion is caused by the substrates dissolving partially into each other, creating a mixed interphase. According to the theory, adhesive strength is highest when the substrate and adhesive solubility are similar. The theory is particularly useful for describing the adhesion of polymers to each other and themselves. In particular, the diffusion of block copolymers such as polyurethane is partially controlled by the lengths of the blocks.
3. The mechanical interlocking theory of adhesion describes how the surface shape of the substrates facilitates adhesion. The rougher the surface, the more surface area and openings for the adhesive to fill, the stronger the bond because the adhesive must deform to be torn from the interphase. For the adhesive to fill the gaps provided by a surface, it must efficiently fill pores and wet the surface to be bonded.

4. The acid – base theory of adhesion is that acidic – base interactions form part of the adhesive force. A general principle that works in practice is to use acidic adhesives to bond base substrates, and vice versa.
5. The covalent bonding theory of adhesion is based on the strongest known chemical force, the covalent bond. The energy needed to break apart a pair of atoms that share an electronic pair is far in excess of the dispersive attraction forces.
6. In addition to the adhesive – substrate interaction, an important condition known as a weak boundary layer greatly affects adhesion. A weak boundary layer is a thin layer of the substrate or adhesive that has low cohesion. In wood bonding, a layer of wood dust or extractives can cause a weak boundary layer.

When an adhesive is applied onto the surface of wood, in addition to the six theories, other factors affect the adhesive bond quality and performance. In figure 4 a model of adhesively bonded wood is shown. The three main parts are the wood, the interphase between the wood and the adhesive, and the adhesive. The adhesive has to wet, penetrate and spread to the opposite surface (figure 5). The viscosity, molecule size, polarity and solubility of the adhesive affect this process. The general principle applied to the wood adhesive bond, is that the bond must be either the same strength or stronger than the wood it bonds, in all the conditions the product is subjected to (Selby 1975, 3). To meet this requirement, several factors in addition to the adhesion and properties of an adhesive must be taken into account. The type, density and shape of the wood being bonded can cause the internal stresses in the x,y and z directions (figure 4) that lead to delamination. Tensile forces in the normal direction (y) are where the adhesive is weakest (Frihart 2005, 244). The bonding process itself must also be carefully calibrated. It has to be optimum with respect to conditions, bonding pressure and length of time, to prevent over penetration of the adhesive into the wood lamellae, leaving a starved adhesive layer (Frihart 2005, 232).

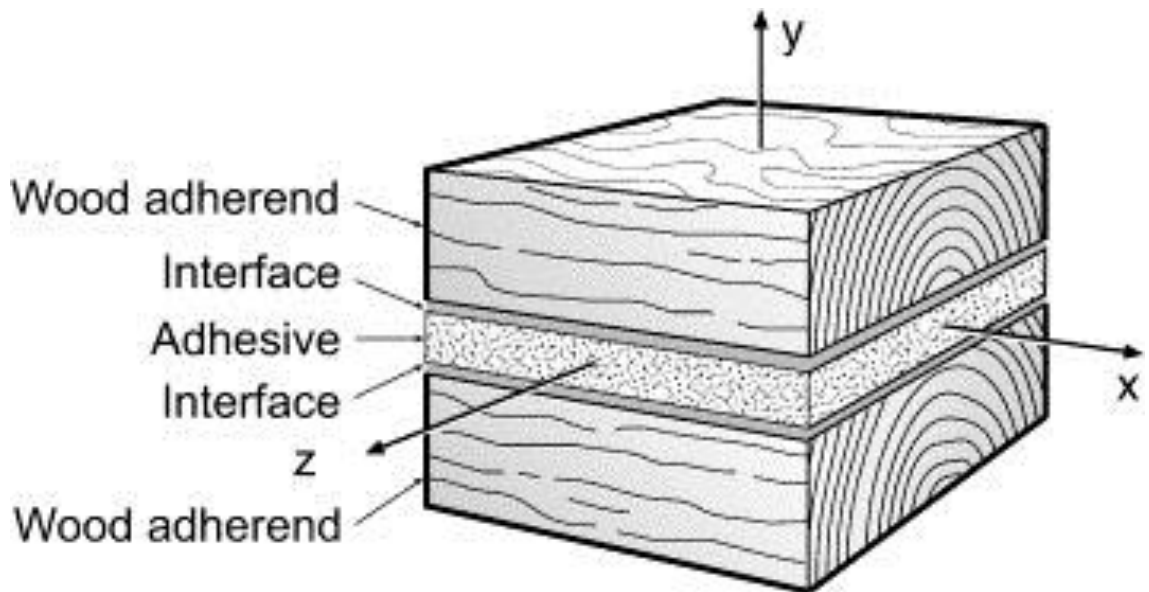


FIGURE 4. Model of a wood adhesive bond line (Serrano 2004, 26)

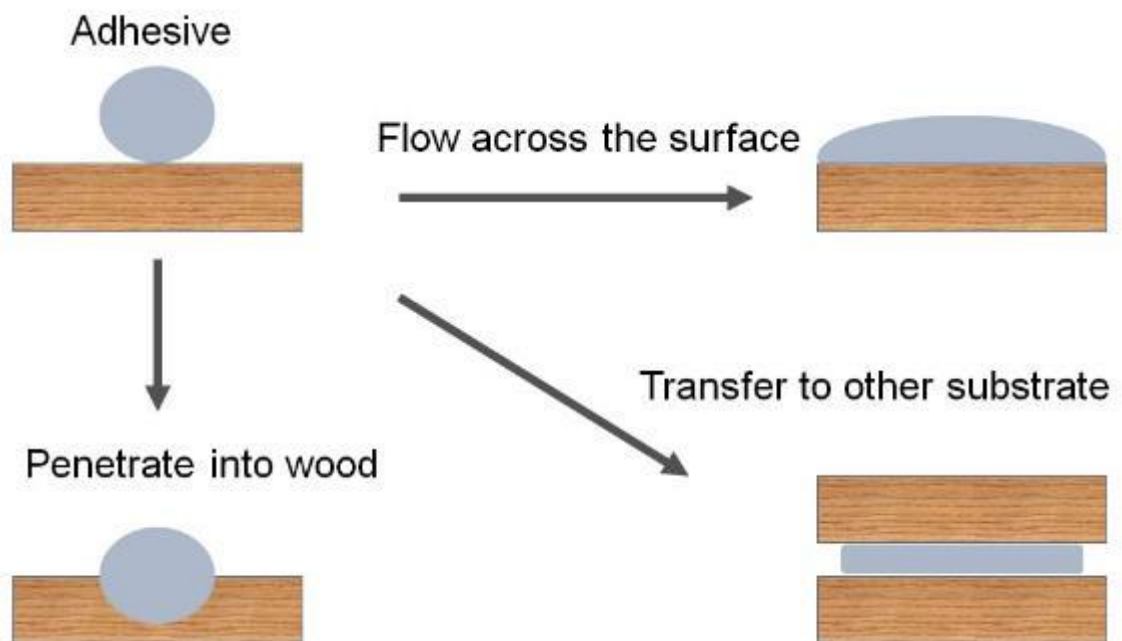


FIGURE 5. Spreading and penetration in wood bonding (Rowell 2005, 270 edited)

Frihart, (2009) in a paper on adhesive groups and their relationship to the durability of bonded wood, proposes a model of classifying weather resistant durable wood adhesives into two types, in-situ polymerized and pre-polymerized. In-situ polymerized adhesives are made up of small molecular weight (<2000) molecules that effectively cross-link during the bonding of wood. They penetrate into the middle lamella between cells in wood and into the cell wall layers. In-situ polymerized adhesives resist delamination by spreading the moisture induced swelling and shrinking stresses away from the bond line into the wood (figure 6). In the figure, the stress of moisture induced swelling is distributed into the wood in gradually decreasing steps that illustrate the penetration and wood strengthening ability of in-situ polymerized adhesives. An example of a pre-polymerized adhesive is phenol-resorcinol formaldehyde, known as PRF, which is the control adhesive used to evaluate new adhesives against in long term creep tests included in SFS EN 15425:2008. (Frihart 2009, 601-614; SFS EN 15425:2008.)

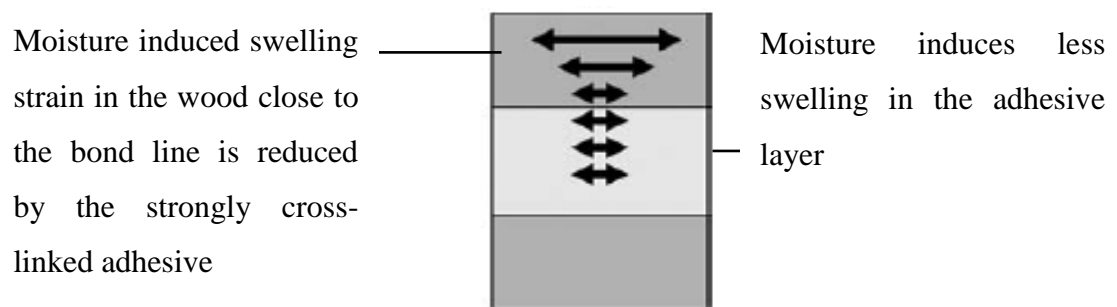


FIGURE 6. Mechanism of delamination resistance by in-situ polymerized adhesives (Frihart 2009, 611 edited)

The second group, pre-polymerized adhesives, includes the two adhesive types used in this thesis, 1K PUR adhesives and EPI adhesives. Pre-polymerized adhesives have long, large molecular weight backbones, which allow movement even when cross-linked. In 1K PUR the long flexible sections are the soft segments, from the polyol component, and in EPI adhesives they are made up of the polymer dispersion used. Because they have a large molecule size, these adhesives do not penetrate into the cell wall, however, they can fill the lumen, which is the space inside the inner layer, as well as the middle lamella to form lock and key polymerized structures. In figure 7, the stress release mechanism of pre-polymerized adhesives is shown. Because the adhesives retain some flexibility, stress maxima points do not form as easily and the wood swelling induced dimension changes are handled by the adhesive stretching cohesively.

Especially for 1K PUR adhesives, the carbon dioxide released during curing creates a foamy structure effectively dissipating strain without delamination. (Frihart 2009, 611-612.)

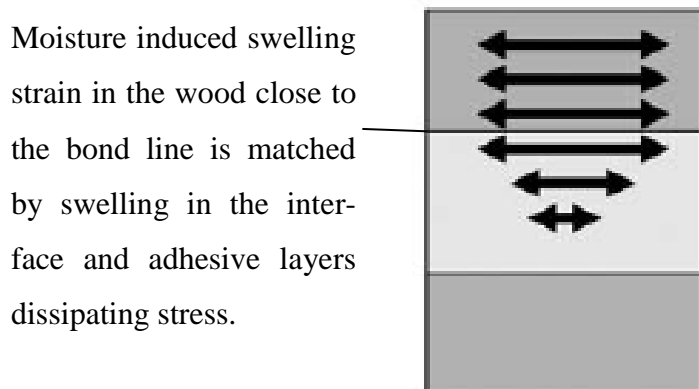


FIGURE 7. Behavior of pre-polymerized adhesives during moisture induced swelling (Frihart 2009, 611 edited)

2.5 Glued solid and laminated timber

Glued laminated timber or glulam, is made out of layers or laminations of finger jointed timber glued together along the grain direction. Glulam was used for the first time in 1893, in Basel, Switzerland to construct an auditorium (Forest products laboratory 1999, 11-3). Glulam was first produced in Finland in the 1940's and gained popularity from the 1960's onwards (Carling 2003, 9). In 2011 a total of 330,000 m³ of glulam, duo and triolam was produced in Finland. Duo and triolam are glulam beams made out of two and three laminates and are also called glued solid timber. A large part of the Finnish glulam production is exported to Japan. Glulam production has been growing over the past decade, and Finland is one of the largest producers of glulam in Europe. (Suomen liimapuuyhdistys, 2011.)

Glulam is used because of its many advantages over solid timber. According to the Forest products laboratory Wood handbook (2010), the five main advantages of glulam are as follows: size capabilities, architectural effects, seasoning advantages, varying cross sections and varying grades of timber. Size capabilities means that large trees are not required to produce large glulam beams but cost effective small trees can be used instead. Architectural effects can be created by bending the glulam beams into curved

shapes. Glulam is manufactured from seasoned i.e. dried timber, which is less likely to warp and crack in use. Very different sizes of beams can be produced, to match the required strength needed in different applications. Grading timber means assessing its quality. Because glulam is comprised of many pieces of graded timber together, it means the faults in the timber are spread out and affect the strength of the finished product less. (Forest products laboratory, 2010 11-17 -- 11-18.)

In addition to these benefits, glulam is also considered to have other useful properties. It is aesthetically pleasing, fire resistant, has a good strength/weight ratio, is resistant to chemical attack, is a good insulator and can be produced with very little waste. The life cycle of a glulam beam can be extended by reusing, if the original classification is known. (Carling 2003, 9.)

A comprehensive product certification for glulam is in place in Finland. Certified glulam produced in Finland has to be manufactured from Norway spruce (*Picea abies*) or Scots Pine (*Pinus sylvestris*) (Carling 2003, 209). The glued solid timber used for the thesis as well as the raw material used for sample preparation was Scots Pine.

2.5.1 The manufacturing process of glulam

Glulam goes through many stages during production. In figure 8, the typical phases of glulam production are summarized (Carling 2003, 11). The focus of this thesis is on the factory lamellae pressing quality control, as well as developing adhesives for the glue application and lamellae pressing phase of production. However, each production stage is important for a good final product.

The production process starts with harvesting timber from forests. After the timber is sawn, it is dried in large kilns to remove excess moisture. The timber is dried to a maximum moisture content of 18 % in glued solid timber production (RAK 21 2004, 5). The factory humidity and temperature environment must be equivalent to the equilibrium of the dried timber. The stress grading can be an automated or visual process, in which the number of faults are checked and the wood is graded into a strength grade according to SFS 5878 INSTA 142. After grading, the lamellae are finger-jointed with an adhesive that is suitable for finger jointing. This process and quality control of fin-

ger joints are also covered by standards. Finger-jointing enables different length lamellae to become the same length for manufacturing into a glulam beam. The lamellae are then knife-planed, which means the surfaces are cut so that the lamellae is the same thickness throughout. A tolerance of 0,1 mm is allowed between thickness measurements from both sides of the lamellae. Knife-planing also produces a good surface for the adhesive to be applied on, because it removes a layer of wood that can be dirty, have broken cells or excess amounts of extractives. Knife-planing is preferred over abrasive planing. Abrasive planing crushes the cells close to the bond line, resulting in worse delamination resistance under accelerated and actual weathering (Murmanis, River & Stewart 1983, 115). This is due to the formation of a weak boundary layer. The lamellae to be made into glulam have to be bonded within 24 hours of planing. (RAK 19 2004, 1-13.)

Adhesive application onto lamellae in a factory is done according to the adhesive manufacturer's instructions. An even spread of a prescribed adhesive amount is important. The open and closed assembly time of the adhesive must also not be exceeded. The open assembly time of an adhesive is the time it takes from application to the lamellae being stacked. The closed assembly time is the time from being stacked to the bond line pressure being applied. If the open and closed assembly time is exceeded, poor bond quality will result. This can be due to premature curing or over penetration of the adhesive. Thus the certified adhesive chosen for production should meet the time requirements of production. (Frihart 2005, 221.)

The lamellae are pressed for at least the minimum time specified by the adhesive manufacturer. The pressure under which they are pressed depends on the thickness of the lamellae. Lamellae of 35 mm thickness or under are pressed at 0,6 N/mm² and lamellae of 35 mm - 45 mm thickness are pressed at 1,0 N/mm². After pressing, the glulam has to be allowed enough time for the bond line to harden. Then the beam is planed into its final shape and wrapped for transportation to a building site. Wrapping protects the beams from dirt and weather conditions before installation. (RAK 19 2004, 9.)

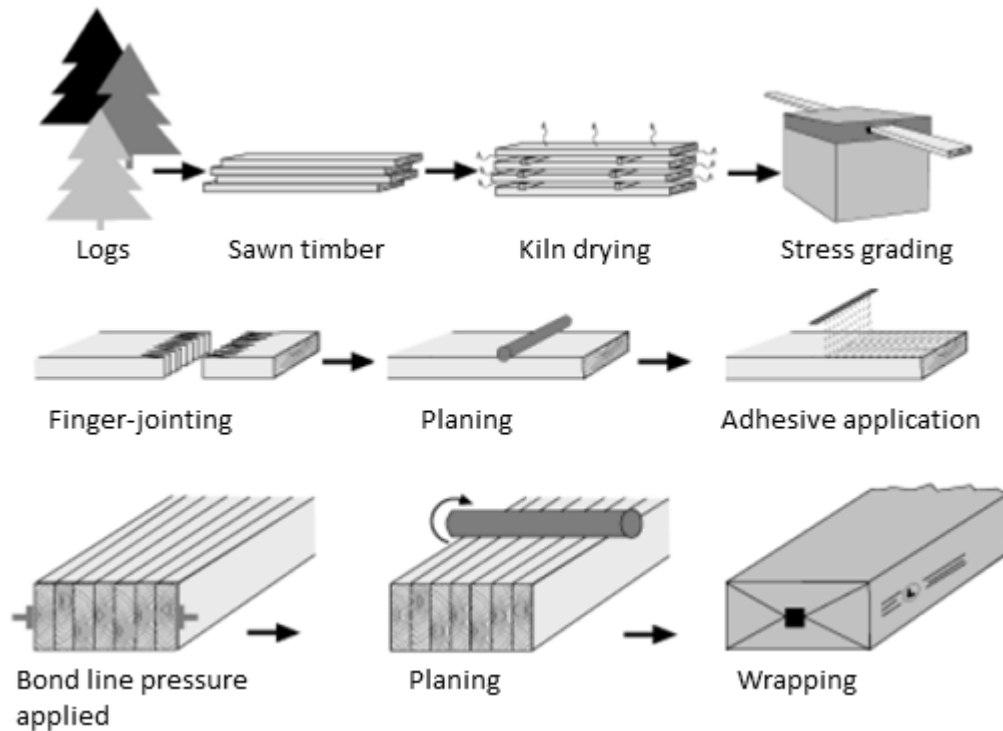


FIGURE 8. Schematic of the phases of glulam production (Carling, O. 2003, 11 edited)

2.6 Delamination tests of glulam and glued solid timber

The development of accelerated weather exposure tests started during the Second World War. There was a need to build ships quickly and from the materials available, which were laminated beams. (Truax & Selbo 1956, 2.) Laminated beams that are exposed to weather undergo moisture changes, which causes the dimensions of the wood to change. Wood swells with increasing moisture content up to its fiber saturation point. When the moisture content is decreased, wood shrinks. These changes create stresses at the bond lines, which can lead to delamination of bond lines. The development of new adhesives would not be practical, if every new adhesive would need to be tested for the duration of its typical service life, which is at least 50 years. Accelerated exposure tests aim to produce similar stresses in the bond lines in a much shorter time than outdoor exposure.

A March 1956 report from the Forest products laboratory in Madison, Wisconsin, USA describes the development of the first accelerated weather tests. The first accelerated tests took 180 days, and were a cycle of 30 days water soaking and 30 days drying at

normal temperatures. A low amount of delamination in this test was found to correlate with low amounts of delamination in beams exposed to weathering over longer periods of 1-4 years. It was also found that delamination increased only slightly after 1 year exposed to weather. This is due to irreversible relaxation of internal stresses. Bond strength measured by block shear tests was found not to correlate well with the amount of delamination due to weather exposure over a long period. (Truax & Selbo 1956, 3-6.)

The discovery that application of vacuum and pressure alternately under water increases the wood moisture content quickly to the fiber saturation point and beyond, led to an even faster test method being developed. The new method involved a cycle of vacuum and pressure and soaking for one day under water, then drying for 6 days in a controlled environment of 27 °C and 30 % relative humidity. This test cycle was repeated a total of three times, for a test period of 21 days. The test samples were cut from beams that were placed outside and subjected to weather for 1 year and the results were compared. Individual results were not consistent, but a clear general trend was visible, which was that small and large delamination amounts in the samples subjected to the 21 days test correlated with small and large delamination amounts in the beams which were exposed to weather for 1 year. This relationship was plotted below (figure 9), which is taken from the report. In figure 9, it can be seen that the correlation between the 21 days three cycle test and outdoor weather exposure results with pine and fir is very good. The correlation for oak and maple is not as good, but a similar trend is present. (Truax & Selbo 1956, 3-23.)

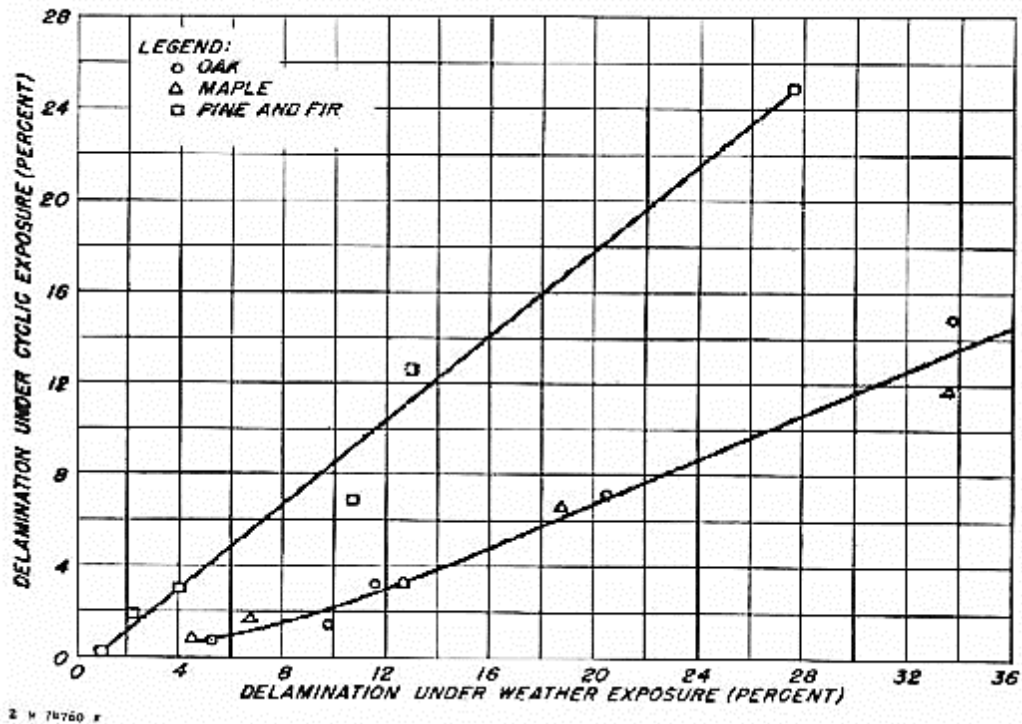


FIGURE 9. Correlation of delamination under cyclic exposure and weather exposure (Truax & Selbo 1956, 21)

With increased experience, the delamination cycles were shortened further. Increased drying air temperatures and a higher velocity air stream contributed towards shortening the test to 12 days in 1950, 8 days in 1959, and further to the three days and one day cycles in use today. The process of shortening test cycles may have affected the correlation with the original weathering exposure tests. (Winandy & River 1986, 28.) A three cycle vacuum-pressure delamination test, SFS EN 302-2 that takes four days to complete, is the current form in Europe for evaluating the suitability of adhesives for structural timber use. The one or two cycle, two, three or four day test methods that are described in SFS EN 391 are in use currently as factory production quality control tests in Finland.

2.7 Standards and certifications for glulam and glued solid timber

The CE-marking of glulam in Finland is covered by the SFS EN 14080:2005 (Timber structures – Glued laminated timber and glued solid timber – Requirements) standard. The requirements for the adhesives used for bonding the CE-marked glulam are also covered by the same SFS EN 14080:2005 standard and the specific adhesive standards

it refers to. The CE-marking of products is possible when the product has a harmonized product standard, and the product meets the requirements at each stage of production. CE-marking is also allowed with a European Technical Approval (ETA), which can be issued on a case by case basis. EN standards are created and approved by the European Committee for Standardization, known as CEN, which is mandated by the European Union. The aim of the CEN is to provide harmonized product standards that facilitate free trade in safe products among the CEN members. The CEN has 33 members, 27 EU members as well as Croatia, The Former Yugoslav Republic of Macedonia, Turkey, Switzerland, Iceland and Norway. EN standards are voluntary, openly available standards created by Technical Committees (TC) at CEN. The standards are then adopted into each member nation's national legislation. The Finnish standardization body, Suomen standardisoimisliitty ry (SFS) is a member of CEN. The CE-marking of building products in Finland is mandatory from 1.7.2013, including glulam and glued solid timber. The national bodies in Finland that have a certificate to audit CE-marked glulam production are the VTT Technical research center of Finland, Inspecta Oy and Finotrol Oy. (CEN 2010; Ympäristöministeriö 2012.)

The standard specified in SFS EN 14080:2005 for factory production quality control of bond lines is SFS EN 391:2001 (Glued laminated timber. Delamination test of glue lines). The standard SFS EN 391:2001 specifies three methods for continuous quality control of glulam and glued solid timber. Methods A and B are for type I adhesives (service classes 1,2 & 3) according to SFS EN 301:2006 classes, while method C is for type II adhesives. SFS EN 14080:2005 is in the process of being replaced with a new version of EN 14080. There is a final draft version of a new replacement, FprEN 14080:2013, that could update the current performance requirements, but the final standard is not in use yet as of January 2013.

1K-PUR adhesives for glulam and glued solid timber manufacturing in Finland have to meet the requirements in SFS EN 15425:2008 (Adhesives. One component polyurethane for load bearing timber structures. Classification and performance requirements). To produce CE-marked glulam and glued solid timber, the adhesive also has to fulfill extra requirements specified in annex C of SFS EN 14080:2005 (Annex B. of FprEN 14080). Currently, there is no standard that specifies producing CE-marked glulam or glued solid timber with EPI adhesives in use in the CEN countries. The new FprEN 14080 final draft, however, includes EPI adhesives. In the FprEN 14080 final draft, EPI

adhesives can be used for service classes 1 & 2 if they meet the same requirements as 1K-PUR adhesives. In addition there is a provision for small dimension glulam or glued solid timber produced with EPI adhesives, which can be certified by completing tests with a 0,3 mm bond line thickness instead of the 0,5 mm specified in SFS EN 15425:2008.

2.8 Development of new adhesives

Developing new adhesives is based on understanding the chemical nature of the surface of the substrate, as well as the interface between the adhesive and the substrate. In addition to the ability to bond substrates, standard based requirements and production requirements each shape the properties needed for an adhesive to become a commercially successful product. In demand are new adhesives that are faster curing, faster curing at lower temperatures, have better resistance to delamination due to chemical stress, are better at bonding difficult substrates, as well as have specific optical and electrical properties. The means of achieving these functionalities can be synthesis of new raw materials, adding functional components to the formulations or introducing new combinations of known raw materials. Tests simulating the lifetime of bonded components are based on standards and evaluate the suitability and performance of adhesives. Information about the causes of adhesive failure are essential to develop new adhesives. (IFAM 2013.)

3 MATERIALS AND METHODS

For experience with the easyQ DLA appliance, comparing EN 391 method B test results with Kontio production quality control results was chosen. Kontio provided samples that were bonded using a commercial, EN 14080:2005 certified 1K PUR adhesive. The test method specifies taking 2 samples from each batch to be tested, an A and B sample. Kontio quality control tested the A sample, and the surplus B samples were sent to the research and development laboratory at Kiilto for testing using the new appliance. A total of 36 glued solid timber samples of various shapes and sizes were received to perform this section of the thesis.

For comparing adhesives in development, the SFS EN 302-2:2004 method was chosen as a reference. Passing the requirements is one of the prerequisites for an adhesive to become a certified product for CE-marked production. This method is the one used for the delamination test in the adhesive certification. Because of equipment limitations, some modifications to the standard were made with regards to the samples. A test body certified adhesive was used as a control sample.

3.1 EN 391 method B

The SFS EN 391 method B is described fully in the standard “EN 391:2001 Glued laminated timber. Delamination test of glue lines”. The method described in the standard involves cutting two 75 ± 5 mm wide cross-sections from a laminated beam, and testing one of them under a single cycle of vacuum-pressure immersed in water, then drying the sample. The results are analyzed after the sample has dried to within 110 % of its original weight. The total delamination percentage is then calculated according to formula (3) and the maximum single bond line percentage calculated according to formula (4) (SFS EN 391:2001, 8).

$$100 \frac{l_{tot,delam}}{l_{tot,glueline}} = \text{Total delamination percentage} \quad (3)$$

$$100 \frac{l_{max,delam}}{2 l_{glueline}} = \text{Maximum delamination percentage} \quad (4)$$

Only the cut sample surfaces are evaluated. In formula (3), $l_{tot,delam}$ is the sum of the delaminated sections in mm on the bond lines of the two faces of the samples, and $l_{tot,glueline}$ is the sum of the bond lines on the same faces. In formula (4), $l_{max,delam}$ is the sum of the delaminated sections on the two faces of the same bond line, while $2l_{glueline}$ is the total length of the two exposed sections of that bond line.

The sample quality control requirements for the glued solid timber samples used in this thesis were; less than 8 % for total delamination percentage (3) after one cycle, and less than 40 % delamination (4) for a single bond line. If the sample does not meet the first requirement of less than 8 % delamination, it is subjected to a second cycle of vacuum-pressure soak, and the delamination percentage is calculated after the second cycle is complete. The limit of acceptance for the second cycle is less than 16 % total delamination. In the final draft version of FprEN 14080:2013 the limit for samples tested for two cycles is lowered to less than 12 % total delamination. The limits for glulam are 4 % total delamination for the first cycle, and 8 % total delamination after the second cycle. If the A sample does not meet the requirements, the B sample is tested in the same fashion. If both samples fail to meet the requirements, the production batch is rejected.

3.1.1 EN 391 method B sample preparation

A total of 36 samples were sent to the research and development laboratory at Kiilto. The samples were marked, then weighed, and then the bond line lengths were recorded. The samples were of various sizes, differing in the amount of lamellae and the lamellae thickness. The samples were chosen by Kontio. The samples were kept wrapped in a black plastic covering until being tested to keep their moisture content as similar to when leaving the factory as possible.

3.1.2 SFS EN 391 method B vacuum-pressure soak

The standard, EN 391, specifies using a pressure vessel capable of 700 kPa absolute pressure and a vacuum of 85 kPa. The easyQ DLA appliance is a pressure vessel equipped with pumps and a control interface designed specifically for standardized de-

lamination tests on timber. The appliance can be seen in picture 2. The main parts are the pressure vessel, a vacuum pump, a water pump, and the control screen.



PICTURE 2. easyQ DLA appliance

The samples should be placed inside the pressure vessel in such a way that they do not touch each other, or the walls of the pressure vessel as can be seen in picture 3. The samples are fixed in position using a wire mesh and prevented from floating using a lockable bar. The size of the pressure vessel is 100 liters, which sets a limit on the amount of samples that can be tested simultaneously. The samples were placed in the pressure vessel, and the lid was fastened by hand. The easyQ DLA appliance has a control interface, from which it is possible to choose the EN391 method B program. The computer program controls automatic valves and the pumps, and the program goes from start to finish automatically.

The EN 391 method B vacuum-pressure soak starts by filling the pressure vessel three quarters full of water. This places the samples below the water line, but leaves an air space in the top quarter of the pressure vessel. Then a -75 kPa below ambient air pressure vacuum is drawn by the vacuum pump for 30 minutes. Following the release of the

vacuum, a pressure of 550 kPa above ambient air pressure is created by the water pump in the pressure vessel for two hours.



PICTURE 3. SFS EN 391 method B samples inside the pressure vessel

The test cycle saturates the samples above their fiber saturation point with water, increasing their weight 1,5 – 2,5 times depending on the sample. Each sample of wood increased in weight by a different amount due to each sample of wood being unique. The saturated samples were then dried in a dry heat chamber or in a weather chamber at Kiilto. The SFS EN 391 method B states that samples should be dried at 70 ± 5 °C, 8-10 % relative humidity and with an air flow of 2-3 m/s. The equipment used at Kontio for performing SFS EN 391 method B included a drying chamber, which was custom built for drying samples, but did not include a wind speed meter or a relative humidity measuring device.

Two deliveries of samples were sent to Kiilto from Kontio, and the experiences from the first batch were used to partly decide the drying conditions for the second batch. In addition to different drying conditions, the samples were of various shapes and sizes, providing a representative cross-section of production. The smallest samples, comprised a single bond line with a total bond length of 344 mm and the largest, 8 lamellae, bonded together in two vertical sections of four lamellae, with a total bond length of 1540 mm.

The samples were dried according to table 2 to find out the effect of different drying conditions on the delamination results, compared with the equipment used at Kontio for quality control purposes. Samples dried with the different methods were designated A, B, C and D using running numbering. Neither the weather cabinet or dry-heat chamber had controllable fans, so it was not possible to adjust the air flow speed. The weather chamber allowed humidity to be set at a specific value but the temperature chamber did not have a humidity setting. The dry-heat chamber has small dimensions, which makes it unsuitable for continuous testing.

TABLE 2. The different drying conditions and number of samples per test batch.

Drying chamber	Temperature (°C)	Relative humidity (%)	Number of samples	Test batch designation
Dry-heat chamber	65	n.a.	4	A
Weather chamber	65	9	16	B
Weather chamber	70	9	8	C
Weather chamber	75	9	8	D

3.1.3 Evaluation of EN 391 method B samples

The dried samples were evaluated individually after drying to 100-110 % of their original weight. The evaluation takes place in two stages. The first stage is weighing the samples, then recording the delaminated bond line sections by drawing vertical lines to mark the edges using a ball-point pen as in the example in picture 4. In the SFS EN 391-B method, using a magnifying glass and 0,1 mm thick feeler gauge are recommended to assist in marking the delaminated bond line sections.

The first evaluation stage should take place within an hour of removing the samples from the drying process, and in practice it was completed in under 15 minutes. It is important to complete this stage quickly, because wood immediately starts returning to its

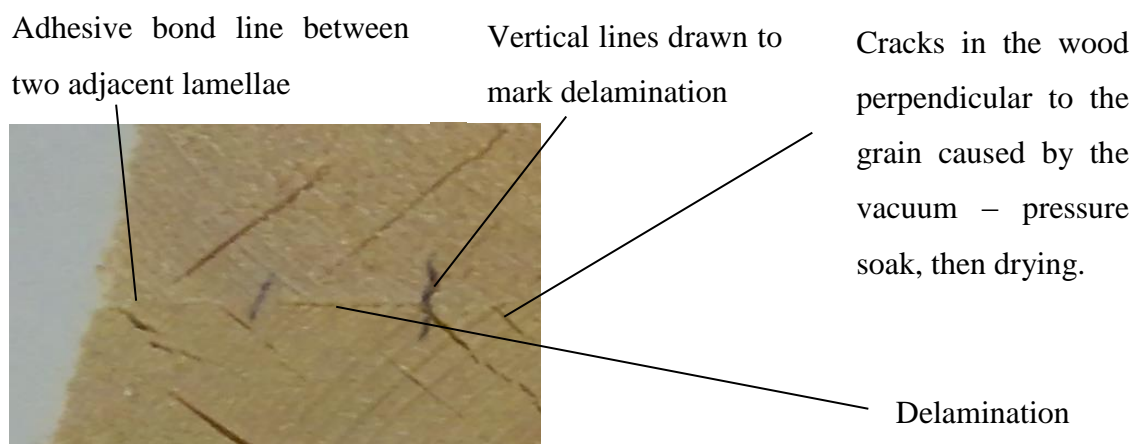
equilibrium size in normal room temperature and humidity, shrinking the delaminated sections and making them harder to notice.

The second stage is studying the delaminated sections and recording the results. The SFS EN 391 (2001) standard section 6.4.2.2. gives the criteria for valid delaminations, that are:

1. Cohesive failure in the adhesive layer.
2. Bond line failure precisely between the adhesive and the wood layer.
3. Wood failure within the first two wood cell layers, which is not a logical extension of cracks due to the growth ring or grain angle structure of the wood.

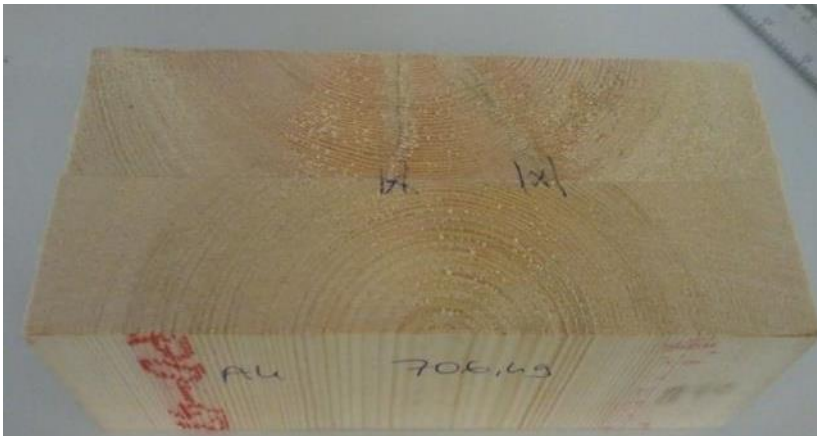
Section 6.4.2.3 of the SFS EN 391 (2001) standard gives examples of delamination that are not considered to be due to adhesive failure, and therefore are to be excluded:

1. Wood failures more than two cell layers away from the bond line due to growth rings or the angle of the grain.
2. Under 2,5 mm wide openings that are more than 5mm away from other openings.
3. Delaminated sections that are due to knots, visible or hidden, as well as resin pockets.



PICTURE 4. Marking delaminated sections in the first stage of evaluation

Hidden knots can be discovered by splitting the samples along the bond line with a chisel or wedge and a hammer. Excluded sections are marked with a cross, as seen in picture 5. The knots are indicated by the dark sections of wood leading to the two excluded sections. Valid delaminated sections according to 6.4.2.2. from the SFS EN 391 standard were then measured with a ruler ($\pm 0,5$ mm) and a magnifying glass, and the sum for each bond line was entered into an excel chart with the other sample details such as the production batch, sample original weight, sample weight after drying, sample amount and length of bond lines, total bond line length and sample drying time. Using formulas (3) and (4) each bond line is given a maximum delamination percentage, as well as a total delamination percentage. An example of an evaluation table is given in table 3. Excel charts such as table 3 with pre-entered formulas are useful for quick result analysis.



PICTURE 5. Sample with excluded bond line sections marked with a cross

TABLE 3. An example of an evaluation table

Sample	Original weight (g)	Final weight (g)	Percentage original weight (%)	Drying time (h)	Length of bond line (mm)	Length delaminated (mm)	Maximum delamination %	Total delamination %	Remarks
A.1	942	1021	108,6	20	346	46	13,3	13,3	Second cycle needed

3.2 EN 302-2:2004 laboratory method

SFS EN 302-2:2004 (Adhesives for load-bearing timber structures. Test methods. Part 2: Determination of resistance to delamination) is a laboratory method used in the adhesive certification process. Passing the SFS EN 302-2:2004 test is part of the requirements included in SFS EN 15425:2008 for one-component polyurethane adhesives for CE-marked glulam and glued solid timber. The method for type I adhesives was chosen. Type I adhesives can be used in all service classes, which correspond to the conditions likely to be encountered in use. SFS EN 15425:2008 specifies type I as being adhesives that can have prolonged exposure to high temperatures (such as ≥ 50 °C) as well as full exposure to weather. Minimum requirements for resistance to delamination for type I adhesives are specified in section 5.3 and 6.3 of the SFS EN 15425:2008 standard as being <5 % for all test samples tested according to SFS EN 302-2. Certified laboratories that perform the SFS EN 302-2:2004 test are the Nordisk Treteknisk Institutt (NTI) in Oslo, Norway and Materialprüfungsanstalt Universität Stuttgart (MPA Stuttgart) in Germany. A modified test method similar to the SFS EN 302-2 standard method was developed for testing adhesives according to table 5.

3.2.1 Moisture-cure one component polyurethane adhesive formulation

Moisture-cure polyurethane adhesive sample batches were formulated in the laboratory. Because the adhesives cure on contact with atmospheric water content, a quick method for adhesive compilation was chosen to prevent premature curing. The SpeedMixer DAC 400 FVZ is a dual asymmetric centrifuge manufactured by Hauschild Engineering based in Hamm, Germany. Dual asymmetric centrifuges are used to mix various substrates, such as liquids, powders and pastes. Some typical uses according to the manufacturer are blending adhesives and sealants. As can be seen (figure 10), dual asymmetric centrifuges (DAC) have dual rotations simultaneously in opposite directions. (SpeedMixer 2011.) The SpeedMixer cup is placed in a basket on the end of an arm. The arm is spun clockwise while the basket spins counterclockwise. This is different from a regular centrifuge, which only has one direction of rotation. The dual rotations induce mixing of the substrates quickly. Homogenous mixing of adhesive substrates is usually checked visually, and by measuring the Brookfield viscosity (mPas) of the adhesive.

Raw materials were weighed to 0,01 g precision into a SpeedMixer cup on an electronic scale. Mixing was performed using a SpeedMixer DAC 400 FVZ. The sample cup was spun at 800 rpm for two minutes, then 2000 rpm for one minute. The compounded adhesives were then sealed into glass jars with metal lids. The jars were protected from excess amounts of atmospheric moisture by wrapping them with Parafilm “M” laboratory film. From opening the raw material containers to sealing the jars took less than 5 minutes per sample. All the adhesive samples were visually inspected for homogeneity after mixing with the SpeedMixer DAC.

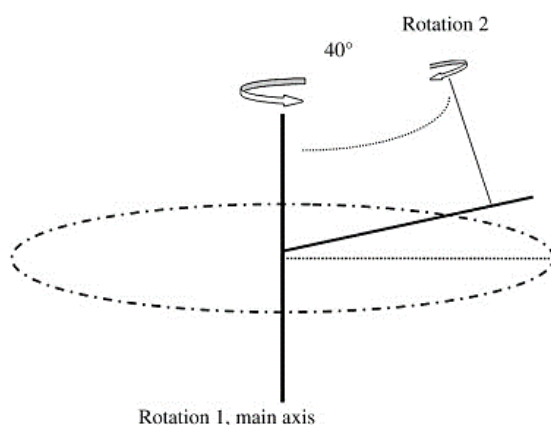


FIGURE 10. Principle of the SpeedMixer centrifuge (Tian et al, 2010)

3.2.2 Modified SFS EN 302-2:2004 method

The SFS EN 302-2:2004 method was modified to be used for testing experimental adhesives at Kiilto, due to equipment restrictions and raw material availability. SFS EN 302-2:2004 standard method for type I adhesives describes a three cycle vacuum-pressure soak and drying test for testing resistance to delamination. The test method developed for Kiilto modifying the SFS EN 302-2:2004 standard has the following steps with the corresponding original SFS EN 302-2:2004 steps in brackets:

1. Two (*four*) members for each adhesive sample are prepared. One (*two*) with short closed assembly time and one (*two*) with long closed assembly time. Closed assembly time is the time taken between the assembly being complete and the press applying the bonding pressure on the completed assembly. For each closed assembly time sample, 5 (*6*) lamellae of 145 (*150 ± 5*) mm wide, 28

(30 ± 1) mm thick and 350 (500) mm long Scots pine (*spruce*) are bonded with adhesive applied to both faces. The bonding pressure is ($0,6 \pm 0,1$) N/mm². The amount of adhesive used, pressing time and closed assembly time are reported by the adhesive manufacturer. In this thesis, they were estimated from prior testing of the adhesive properties. Adhesives were evenly spread on both sides of the bond line using a 200 μ m spiral bar applicator. Open assembly time is the total time taken to spread the adhesives on the lamellae. The open assembly time should be less than 5 minutes. Picture 6 shows a bonded assembly after completing the first step.



PICTURE 6. Bonded Scots pine assembly for Kiilto EN 302-2 method testing

2. For seven days, the bonded members are conditioned in (20 ± 2) °C and a relative humidity of (65 ± 5) %. Three (*two*) cross-section samples that are more than 50mm from the ends and that are (75 ± 5) mm wide are cut. These samples are weighed and the weight recorded into the excel sheet.
3. The samples are placed in the vacuum-pressure vessel and fixed in place as in SFS EN 391:2001 method B. The SFS EN 302-2:2004 test method for type I adhesives is selected, which has four phases that are laid out in table 4.

TABLE 4. EN 302-2:2004 Type I adhesive vacuum-pressure cycle

Step (values are absolute pressure)	Time (min)
Vacuum: (25 ± 5) kPa	15
Pressure: (600 ± 25) kPa	60
Vacuum: (25 ± 5) kPa	15
Pressure: (600 ± 25) kPa	60

4. After the test cycle is complete, the samples are dried for 20 hours in air that is 65 °C (65 ± 3), has a relative humidity of 12,5 % (10-15) and an air speed of (2,25 ± 0,25) m/s until the samples are within 110 % of their weight before the next vacuum-pressure cycle.
5. Steps 3 and 4 are repeated twice, for a total of three complete impregnation and drying cycles.
6. Evaluation of the samples is carried out within one hour of the end of the third drying cycle. Using a magnifying glass and strong lighting, the bond lines are checked for delamination according to the same principles as in SFS EN 391:2001 method B. After the evaluation and calculations, if all the six (*eight*) samples have less than 5 % delamination, the adhesive fills the requirements of SFS EN 302-2:2004.

The changes to the SFS EN 302-2:2004 method for the above steps 1, 2 and 4 for testing adhesives at Kiilto were mainly to the number of samples and sizes. Spruce of the standard dimensions was not available, therefore Scots pine was chosen as the substrate. Scots pine of the dimensions in step 1. was sourced from Lautta Oy. The four adhesives tested are listed in table 5. A total of 24 samples, six for each adhesive with three parallel samples per short and long closed assembly time were tested. The three parallel samples were obtained by cutting cross sections 75 mm wide cross sections from the assembly (figure 11). Modified EN 302-2 method instructions were written for further use at Kiilto.

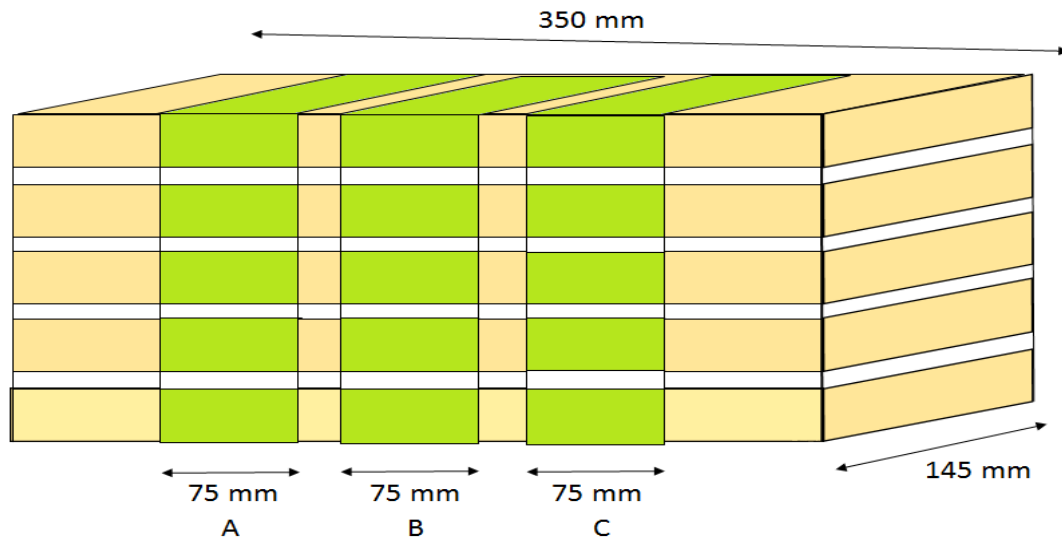


FIGURE 11. Cutting scheme for three parallel samples, A, B and C

TABLE 5. Tested adhesives, short and long closed assembly times.

Adhesive	Parallel Samples	Closed assembly time (min)
Commercial PUR	Control A, B and C	2
Commercial PUR	Control A, B and C	6
PUR A	A, B, and C	2
PUR A	A, B, and C	6
PUR B	A, B and C	2
PUR B	A, B and C	10
EPI	A, B and C	7
EPI	A, B and C	12

During pressing, some assemblies with short closed assembly time slipped horizontally while pressure was applied, causing the bond line lengths to differ from the nominal value in figure 11. Kiilto method samples were dried after the vacuum – pressure soak cycle in the same weather chamber used with the SFS EN 391 method B, with the settings in step 4. Picture 7 shows twelve samples in the drying configuration, with a minimum of 50 mm between samples to ensure even airflow. Final results after the two evaluation stages were entered into an excel chart, into which formulas (3) and (4) were coded.



PICTURE 7. Kiilto test method samples drying in the weather chamber

4 RESULTS

All samples underwent a vacuum – pressure soak cycle in the easyQ DLA appliance, and were then dried to between 100 % and 110 % of their original dry weight. Delamination results were evaluated according to the guidelines posted included in SFS EN 391 method B, as well as using the experience of a visit to the Kontio quality control workstation. The parallel testing of B samples from Kontio was completed first. The requirements for sample acceptance and rejection were taken from SFS EN 14080:2005. Parallel Kontio production quality control results were kindly provided. Evaluating delamination amounts included several judgment factors that are not covered in the guidelines in SFS EN 391 method B, or other published standards and were from Kontio.

4.1 Evaluation of delamination

The presence of knots very often leads to delamination of the bond line. In the SFS EN 391:2001 standard, the instructions are: to not regard openings due to knots as valid delaminations. In several cases, however, the delamination of the bond line occurs not only directly at the location of the knot, but in an area around the knot. For the evaluation of results, the immediate knot affected area were disregarded as delaminated, but areas perceived to not be caused by the knot in question were included as delaminations in the calculations of maximum bond line delamination and total delamination percentages.

In some cases, delaminated sections can have a jagged appearance, with multiple short (0,5 - 2 mm) delamination interspersed with non-delaminated bond line sections. In this case, a guideline of the length of the jagged section divided by two was used as a measurement of the delaminated area. For openings that were less than 2,5 mm wide but further than 5 mm away from the nearest delaminated section, a single vertical line was drawn through the opening while evaluating the samples.

4.2 SFS EN 391:2001 method B results

4.2.1 Results in different drying conditions

The maximum (max) delamination percentages and total (tot.) delamination results were calculated for all the batches according to formulas (3) and (4) along with the final accept / reject evaluation of the samples. Test batch A samples were dried in a temperature chamber in which the only controllable variable was temperature. Test batches B, C and D were dried in a weather chamber, at different temperatures of 65 °C, 70 °C and 75 °C. The weather chamber had larger dimensions than the temperature chamber, allowing for multiple samples to be dried simultaneously and the air to circulate freely. The results for test batch A are shown in table 6, results for test batch B in table 7, results for test batch C in table 8, and test batch D results are in table 9. The sample A.2 from test batch A was rejected after the first cycle, due to a total delamination percentage of 21,2 %, which was above the 8 % limit for the first cycle, and 16 % limit for the second cycle. None of the samples in test batch D required a second cycle.

TABLE 6. Delamination results according to SFS EN 391 method B. Test batch (A) dried in a temperature chamber at 65 °C

Sample	Max delamination after first cycle (%)	Max delamination after second cycle (%)	Tot. delamination after first cycle (%)	Tot. delamination after second cycle (%)	Result (accept / reject)
A.1	13,3	15,0	13,3	15,0	Accept
A.2	21,2	-	21,2	-	Reject
A.3	4,7	-	4,7	-	Accept
A.4	20,1	20,1	9,0	10,0	Accept
Mean \bar{x}	14,8		12,0		

TABLE 7. Test batch B delamination results according to SFS EN 391 method B. Dried in a weather cabinet at 65 °C with 9 % relative humidity

Sample	Max delamination after first cycle (%)	Max delamination after second cycle (%)	Tot. delamination after first cycle (%)	Tot. delamination after second cycle (%)	Result (Accept / Reject)
B.1	1,2	-	1,2	-	Accept
B.2	0,0	-	0,0	-	Accept
B.3	5,7	-	5,7	-	Accept
B.4	0,0	-	0,0	-	Accept
B.5	4,2	-	2,9	-	Accept
B.6	27,8	27,8	10,4	10,4	Accept
B.7	5,6	-	4,2	-	Accept
B.8	2,2	-	0,7	-	Accept
B.9	7,7	-	3,5	-	Accept
B.10	0,0	-	0,0	-	Accept
B.11	3,2	-	1,6	-	Accept
B.12	11,8	11,8	11,8	11,8	Accept
B.13	2,0	-	2,0	-	Accept
B.14	2,3	-	2,3	-	Accept
B.15	4,2	-	4,2	-	Accept
B.16	0,0	-	0,0	-	Accept
Mean \bar{x}	4,9		3,2		

TABLE 8. Test batch C delamination results according to SFS EN 391 method B. Dried in a weather cabinet at 70 °C with 9 % relative humidity

Sample	Max delamination after first cycle (%)	Max delamination after second cycle (%)	Tot. delamination after first cycle (%)	Tot. delamination after second cycle (%)	Result (Accept / Reject)
C.1	21,1	21,1	9,0	11,7	Accept
C.2	25,1	-	8,0	-	Accept
C.3	8,8	-	7,4	-	Accept
C.4	1,0	-	0,9	-	Accept
C.5	21,3	21,3	12,4	12,4	Accept
C.6	10,4	-	6,3	-	Accept
C.7	8,4	-	4,2	-	Accept
C.8	5,6	-	3,1	-	Accept
Mean \bar{x}	12,7		6,4		

TABLE 9. Test batch D delamination results according to SFS EN 391 method B. Dried in a weather cabinet at 75 °C with 9 % relative humidity

Sample	Max delamination after first cycle (%)	Tot. delamination after first cycle (%)	Result (Accept / Reject)
D.1	5,4	3,0	Accept
D.2	9,1	5,2	Accept
D.3	10,0	5,0	Accept
D.4	18,2	4,3	Accept
D.5	10,7	7,5	Accept
D.6	14,8	7,3	Accept
D.7	12,9	5,1	Accept
D.8	10,7	7,0	Accept
Mean \bar{x}	11,5	5,5	

4.2.2 Comparison with Kontio quality control results

For each of the 36 samples tested at Kiilto, data was provided on the parallel samples tested routinely as part of the quality control process. The parallel samples were from the same bonded lamellae assemblies as the samples sent to Kiilto, but were cut from an adjacent section. The parallel results were calculated by Kontio using the same formulas (3) and (4) as the samples at Kiilto. A summary of the maximum delamination percentage results from the parallel samples and the Kiilto samples is shown in figure 12. All the 72 samples tested, by Kontio as well as at Kiilto, passed the requirement of less than 40 % maximum delamination of both sides of the same bond line. The highest maximum delamination values were for sample B.6, 27,8 % for the Kiilto sample and 16,6 % for its corresponding parallel sample (figure 12). The total delamination results comparison is shown in figure 13. All 36 parallel samples tested by Kontio passed the 8 % first cycle requirement. Of the 36 samples tested at Kiilto, one sample failed to meet the 8 % and 16 % requirements of the first and second cycle and was rejected. In general the samples tested at Kiilto resulted in higher total delamination percentages than those tested by Kontio (figure 13). The highest Kontio percentage was 7,8 % for parallel sample B.6. For the samples tested at Kiilto, 21,2 % for sample A.2 was the highest total delamination percentage.

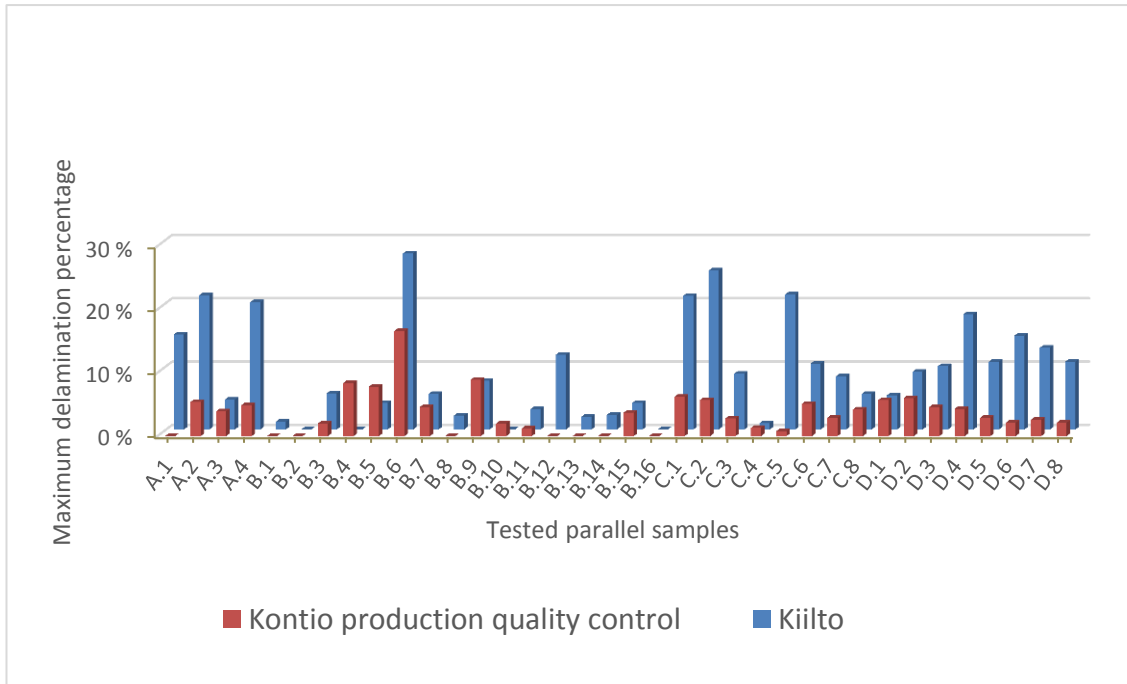


FIGURE 12. Kontio quality control SFS EN 391 method B maximum delamination results compared with Kiilto

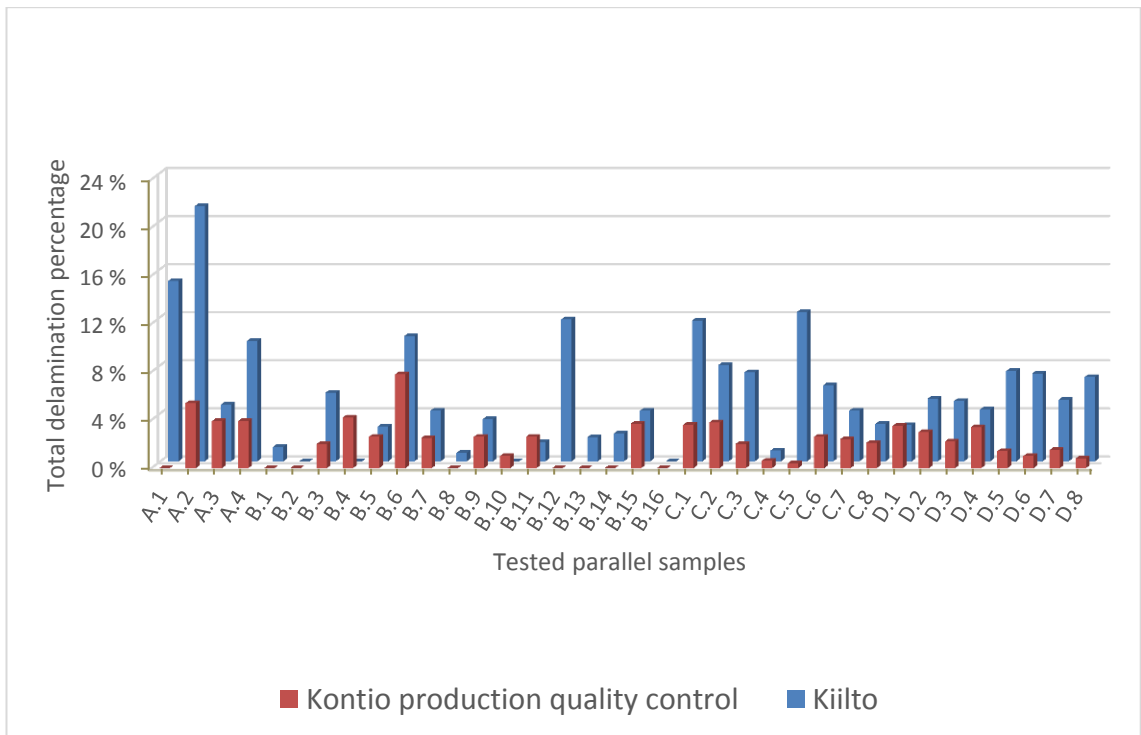


FIGURE 13. Kontio quality control SFS EN 391 method B total delamination results compared with Kiilto

A comparison of the different drying condition test batches compared with quality control results for maximum delamination percentage is shown in figure 14. Total delamination mean results for each test batch compared with quality control mean results for the parallel samples is shown in figure 15. Because the test batches had different sample amounts and bond line total lengths, weighted means by sample amounts and total lengths is shown in table 10.

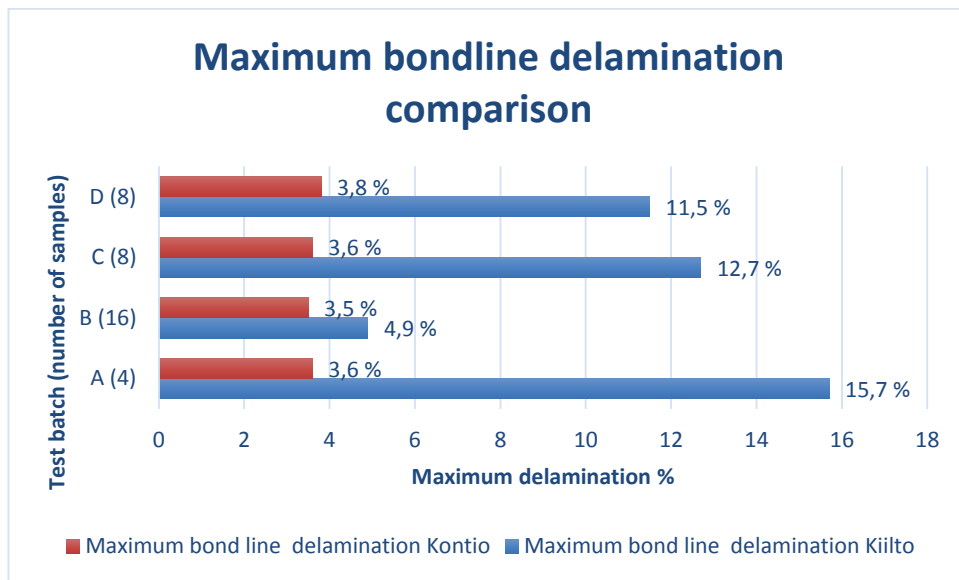


FIGURE 14. Maximum bond line delamination comparison by drying method test batch

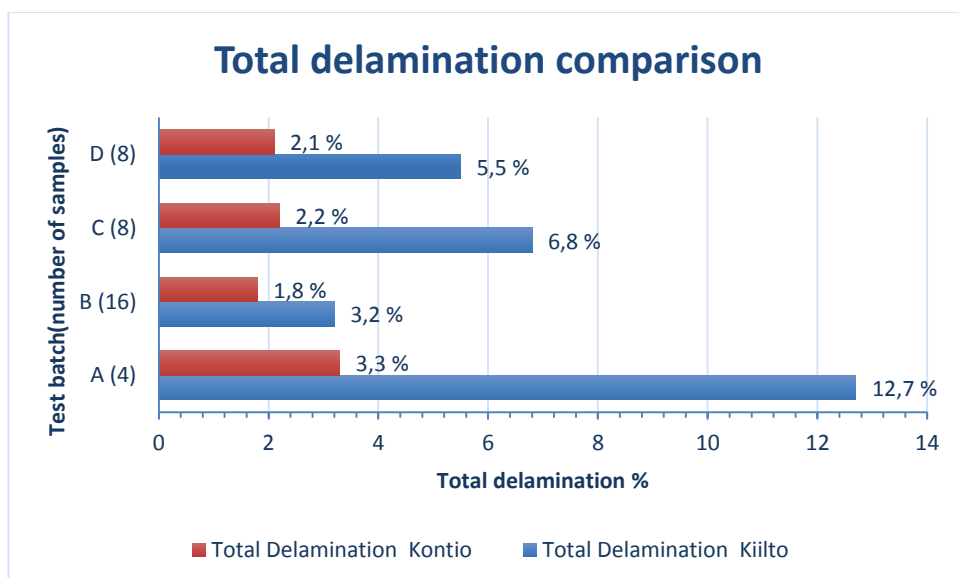


FIGURE 15. Mean results of Total delamination percentage by drying method test batch

TABLE 10. Mean results of Total bond line delamination percentage

Calculation	Total Delamination (%) Kiilto	Total Delamination (%) Kontio
Mean (weighted per sample)	5,6	2,1
Mean (weighted per total bondline)	5,1	2,4

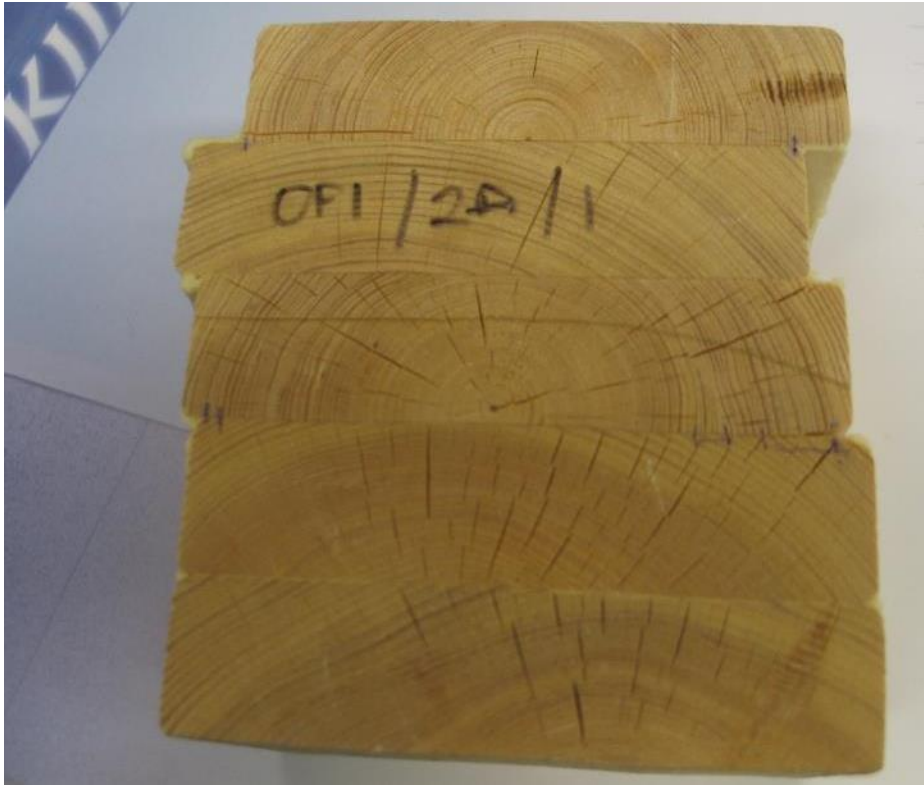
A total of five samples tested at Kiilto had total delamination percentages between 8 % and 16 % after the first cycle, and were retested for a second cycle. Three samples did not delaminate further, and two samples delaminated further by a small amount (table 11).

TABLE 11. Results of samples that were tested for two cycles

Sample	Total delamination after first cycle (%)	Total delamination in second cycle (%)	Final total delamination (%)	Result (Accept / Reject)
A.1	13,3	1,7	15,0	Accept
A.4	9,0	1,0	10,0	Accept
B.6	10,4	0,0	10,4	Accept
B.12	11,8	0,0	11,8	Accept
C.5	12,4	0,0	12,4	Accept
Mean \bar{x}	11,4	0,5	11,9	

4.3 Kiilto test method (modified SFS EN 302-2 test method) results

Kiilto test method pass and fail criteria were taken from SFS EN 15425:2008, being less than 5 % delamination for all test pieces after the three vacuum pressure impregnation cycles for type I adhesives. A picture of a test piece (Commercial 1K PUR A, short closed assembly time) with a low delamination percentage is shown in picture 8. Of note is the extensive cracking caused by the vacuum-pressure impregnation and drying cycles. The second lamellae from the top has slipped during pressing, reducing the length of the bond line. The criteria for valid or invalid delamination was the same as for EN 391, and the same principles were used for evaluation. Where needed, the bond lines were opened with a knife and hammer to check for hidden knots (Picture 9).



PICTURE 8. Commercial 1K PUR 2 minute closed assembly time sample A after the Kiihto test method



PICTURE 9. A hidden knot in an opened bond line

In the Kiilto method as well as SFS EN 302-2, the maximum delamination percentage is not calculated, as only the total delamination percentage is used for passing or failing adhesives. The evaluation results were recorded into an excel chart individually for each sample, and the mean of the three parallel samples calculated. Pass and fail are highlighted in green and red (table 12). Two assemblies achieved a pass status, the short closed assembly time samples for the commercial 1K PUR and 1K PUR B.

TABLE 12. Kiilto test method results matrix (modified EN 302-2)

Adhesive	Closed Assembly time (min)	Tot. delamination Sample A (%)	Tot. delamination Sample B (%)	Tot. delamination Sample C (%)	Mean \bar{x} (%)	Pass / Fail
Commercial 1K PUR	2	1,5	3,5	4,2	3,1	Pass
	6	17,0	21,6	12,3	17,0	Fail
1K PUR A	2	7,3	11,9	5,1	8,1	Fail
	6	33,5	37,2	27,7	32,8	Fail
1K PUR B	2	3,6	1,9	3,8	3,1	Pass
	10	9,2	20,1	15,8	15,0	Fail
EPI	7	20,9	13,3	18,2	17,4	Fail
	12	6,0	4,4	6,6	5,7	Fail

5 CONCLUSIONS AND DISCUSSION

This thesis came about from the decision to invest in a vacuum – pressure impregnation device, which could provide Kiilto with the opportunity to perform accelerated delamination tests. Both goals of the thesis were based upon making the new device as useful as possible: to better serve customers and to be prepared for the start of mandatory CE-marking of building products, starting from the first of July 2013, as well as for internal use developing and testing adhesives. For the purposes of the thesis, the help received from the Kontio glued solid timber and glulam factory was invaluable. Both goals were partially successful, with areas for improvement identified.

The samples tested according to SFS EN 391 method B results showed that samples tested at Kiilto resulted in a higher delamination percentage mean than Kontio, with a total difference of 3,5 %. Calculating the mean weighted by sample length, to account for the sample dimension variations, the results are closer together, with a difference of 2,6 % (table 10). The difference in the results means that, compared to Kontio, short total bond line length samples tested at Kiilto proportionally delaminated more than the samples that had long bond line lengths. This was caused by the testing of shorter bond line length samples in the dry heat chamber, and in the weather chamber at higher temperatures, while large samples were mostly tested in the weather chamber at a lower temperature.

Delving deeper into the results of different drying batches (A, B, C and D), in figure 15, the total delamination percentage mean in the dry heat chamber was 12,7 %, while drying the test samples in 65 °C and 9 % relative humidity (test batch B) resulted in a mean of only 3,2 %. Test batch B had the lowest total delamination percentage mean overall and was the closest to the results from Kontio. The dry heat chamber could be unsuitable for drying the samples, because of harsher drying conditions than in the weather chambers. The lack of humidity control can result in extremely dry air, speeding up the drying process and drying the samples too fast, meaning that stresses are not released gradually. In addition, the dry heat chamber has small dimensions, meaning the air cannot circulate freely. From the results it can be concluded that three factors: temperature, humidity and air circulation are important in the delamination of test samples. The recommendation is for a drying-, possibly a weather chamber with a control-

lable fan and humidity control, in which it would be possible to control the variables to match all the standard requirements.

Even though the samples were wrapped in plastic, moisture and temperature variations during the journey from Kontio to Kiilto could have developed stresses in the bond line that resulted in more delamination. The entire evaluation process of the delaminated samples could produce slightly different results from similar samples, since it contains value judgments to do with invalid delamination. Vacuum-pressure impregnation equipment differences could cause variations in delamination percentages between Kontio and Kiilto. In addition, as the method deals with wood as a substrate, each sample has differences that affect bonding performance. In particular the presence of a larger amount of extractives or compression wood can affect results. The goal, however, that Kiilto can test customer samples and provide results, was achieved to some extent with test batches B, C and D, but work for more accurate and representative results needs to continue on the basis of the results from test batch B.

The second goal of the thesis was to test adhesives formulated in the research and development laboratory with respect to the delamination according to the SFS EN 302-2 method for type I adhesives. The results from the modified Kiilto method showed that achieving good delamination results was difficult, with only two out of eight assemblies managing to fulfill the requirements. Looking at the results in table 12, for all the 1K PUR adhesives, the results from the long closed assembly time are significantly worse than for the short closed assembly time. The EPI adhesive achieved better results with a longer closed assembly time. This difference in the 1K PUR results clearly points to the working time of the adhesives having been exceeded, leading to poor bonding performance. When a 1K PUR adhesive is applied, it immediately begins to react with moisture in the air and in the wood lamellae. If the time taken to begin to press the assembly is longer than the working time, the adhesive can prematurely cure, resulting in a weak boundary layer. That the delamination results clearly show the effect of closed assembly time, means that the method can be used to evaluate maximum closed assembly times, which can lead to more precise product manuals and recommendations. In addition, the method showed a different level of delamination for short closed assembly time between the commercial 1K PUR and 1K PUR A, which could be due to significant differences in their formulation.

The EPI adhesive results were mixed, with the short closed assembly time of 7 minutes giving a delamination mean of 17,4 %, which is well above the <5 % limit. On the other hand, the longer closed assembly time of 12 minutes gave a result of 5,7 %, which was quite close to the minimum requirements and the short assembly time 1K PUR results. This result suggests that 12 minutes did not exceed the adhesives pot life, and was within the range of working life. The 17,4 % for the 7 minute assembly sample could have been due to uneven mixing of the dispersion and hardener and tests should continue to discover the reason.

From analyzing the results from the Kontio samples, as well as samples prepared in the laboratory, important information about adhesive performance as well the delamination test drying conditions was discovered. For future use of delamination tests, both for samples produced at Kiilto or elsewhere, the goal should be to try and minimize the variables involved in the testing process, to make accurate and repeatable results possible. The substrate should be as homogenous and of good quality as possible, which would mean less need for invalid delamination assessment from results, removing judgment issues and increasing the repeatability of the results. To provide information on adhesive properties, samples should be tested well within their working life, instead of the theoretical maximum values. Testing the adhesives should be in the same drying conditions each time. Delamination is a fascinating subject, and with so many factors affecting results there is definitely scope for a lot more study.

REFERENCES

- Carling, O. 2003. Limtre handbok. Translated into Finnish as Liimapuu Käsikirja, Pystynen, H. 2003. Helsinki: Wood Focus Oy/Suomen Liimapuuyhdistys ry.
- CEN. 2010. Compass. CEN The World of European Standards. European Committee for Standardization. PDF. Published September 2010. Read 12.12.2012.
<ftp://ftp.cen.eu/cen/AboutUs/Publications/Compass.pdf>
- Forest.fi. 2012. Indigenous tree species in Finland. Read 28.11.2012.
<http://www.forest.fi/smyforest/foresteng.nsf/allbyid2/DD4FE732E6A5EDFCC2256F3400440DCA?OpenDocument>
- Forest products laboratory. 1999. Wood handbook: Wood as an Engineering Material. General Technical Report FPL-GTR-113. Madison, Wisconsin: US Department of Agriculture, Forest service.
- Forest products laboratory. 2010. Wood handbook-Wood as an Engineering Material. General Technical Report FPL-GTR-190. Madison, Wisconsin: US Department of Agriculture, Forest service.
- Frihart, C. 2005. Wood adhesion and adhesives. Rowell, R (ed.) Handbook of wood chemistry and composites. Chapter 9. Boca Raton, Florida: CRC Press.
- Frihart, C. 2009. Adhesive groups and how they relate to the durability of bonded wood. Journal of Adhesion Science and Technology. Adhesion Science and Technology 23, 601-617.
- FprEN 14080. Timber structures. Glued laminated timber and glued solid timber. Requirements. 2013. Helsinki: Suomen Standardisoimisliitto ry.
- Greenberg, M. & Sekizawa, J. 2000. Diphenylmethane diisocyanate (MDI). Concise International Chemical Assessment Document 27. World Health Organization. Published 2000. Read 11.03.2013.
<http://www.inchem.org/documents/cicads/cicads/cicad27.htm>
- Grøstad, K. & Pedersen, A. 2010. Emulsion Polymer Isocyanates as Wood Adhesive: A Review. Journal of Adhesion Science and Technology. Adhesion Science and Technology 24 (8-10), 1357-1381.
- IFAM, 2013. Development of adhesives. Fraunhofer Institut Fertigungstechnik Materialforschung. PDF. Read 7.2.2013.
<http://www.ifam.fraunhofer.de/content/dam/ifam/de/documents/IFAM-Bremen/2804/fachinfo/broschueren/en/Broschuere-2804-EN-Klebtechnik-Klebstoffentwicklung.pdf>
- Kiilto Oy, 2013. About Us. R&D. Read 13.2.2013.
http://www.kiilto.com/en/about_us/kiilto_oy/r_d/
- Kontio, 2013. Kontio pähkinänkuoressa. Read 30.3.2013.
<http://www.kontio.fi/fin/Kontiotuote-Oy/Kontio-pahkinankuoressa.632.html>

Missouri S&T. 2012. Wood connections. Lecture notes. Missouri University of Science and Technology. Printed 20.11.2012.
http://classes.mst.edu/ide120/lessons/wood/cell_structure/index.html

Murmanis, L., River, B. & Stewart, H. 1983. Microscopy of abrasive-planed and knife-planed surfaces in wood-adhesive bonds. *Wood and Fiber Science* 15(2), 102-115.

Öertel, G. 1985. *Polyurethane Handbook*. Munich: Carl Hanser Verlag.

Peura, M. 2007. Studies on the cell wall structure and on the mechanical properties of Norway Spruce. University of Helsinki. Academic Dissertation.

Pocius, A. 1997. *Adhesion and Adhesives Technology. An Introduction*. Cincinnati: Hanser-Gardner.

Ragauskas, A. 2013. The Structure and Physical Properties of Pulpwood Fibers. From Doctoral thesis by Vander Wieler, L.C. Technical review. Georgia Institute of Paper Science Tech and Technology. Read 20.1.2013.
http://ipst.gatech.edu/faculty/ragauskas_art/technical_reviews/Proerties%20of%20Pulpwood%20Fibers.pdf

RAK 19. 2004. Liimapuorakenteiden valmistuksen sisäisen laadunvalvonnan ohje. SFS-INSPECTA SERTIFIOINTI OY – Rakennustuotteiden laaduntarkastus – Puutuotteet, Tekninen toimikunta. 20.04.2004.

RAK 21. 2004. Liimahirren valmistuksen sisäisen laadunvalvonnan ohje. SFS-INSPECTA SERTIFIOINTI OY – Rakennustuotteiden laaduntarkastus – Puutuotteet, Tekninen toimikunta. 20.04.2004.

Ren, D. 2010. *Moisture-Cure Polyurethane Wood Adhesives: Wood/Adhesive Interactions and Weather Durability*. Virginia Polytechnic and State University. Doctoral Dissertation.

Rowell, R. 2005. *Handbook of Wood Chemistry and Wood Composites*. Boca Raton, Florida: CRC Press.

Senese, F. 2010. What is cellulose? General chemistry online. Frostburg state university. Read 12.02.2013.
<http://antoine.frostburg.edu/chem/senese/101/consumer/faq/what-is-cellulose.shtml>

Serrano, E. 2004. A numerical study of the shear-strength-predicting capabilities of test specimens for wood-adhesive bonds. *International Journal of Adhesion and Adhesives* 24 (1), 23-35.

SFS-EN 391:en. Glued laminated timber. Delamination test of glue lines. 2001. Helsinki: Suomen Standardisoimisliitto ry.

SFS-EN 15425:en. Adhesives – One component polyurethane for load bearing timber structures – Classification and performance requirements. 2008. Helsinki: Suomen Standardisoimisliitto ry.

Sjöström, E. 1981. *Wood Chemistry. Fundamentals and Applications*. New York: Academic Press, Inc.

SpeedMixer 2011. Press Release. Time to make a stir. PDF. Updated 17.8.2011. Read 5.9.2012.

<http://www.speedmixer.com/PDF/Stirring%20Times.pdf>

Suomen liimapuuyhdistys. 2011. Suomalaisen liimapuun tuotanto kasvoi edelleen vuonna 2011. Toimialakatsaus. Read 20.09.2012.

http://www.liimapuu.fi/liimapuu/?__EVIA_WYSIWYG_FILE=28654&name=file

Szycher, M. 2012. *Szycher's Handbook of Polyurethanes*. Second Edition. Boca Raton, Florida: CRC Press.

Tian, W., Schulze, S., Brandl, M., & Winter, G. 2010. Vesicular phospholipid gel-based depot formulations for pharmaceutical proteins: Development and in vitro evaluation. *Journal of Controlled Release* 142 (3), 319-325.

Ulrich, H. 1996. *Chemistry and Technology of Isocyanates*. Chichester: John Wiley & Sons Ltd.

Wang, F. 2012. Henkel. EWP Bonding technology. PDF. Published 7.8.2012. Edited 17.8.2012. Read 13.2.2013.

<http://www.ctss.org.cn/Editor/UploadFile/201281711367377.pdf>

Winandy, J. & River, B. 1986. Evaluation of a method for testing adhesive-preservative compatibility. *Forest Products Journal* 56 (1), 27-32.

Ympäristöministeriö. 2012. CE-merkintä. Published 13.1.2012. Read 12.12.2012.

<http://www.ymparisto.fi/ce-merkinta>