



Fracture characteristics of acetylated young Scots pine

Karin Forsman¹ · Erik Serrano¹ · Henrik Danielsson¹ · Jonas Engqvist²

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Abstract

A study on the fracture characteristics of unmodified and chemically modified Scots pine (*Pinus sylvestris*) is presented. The investigated material consisted of small-dimension sawn timber originating from young logs (thinnings), aged 30–40 years. The modified samples were acetylated with acetic anhydride in an industrial scale process without the use of any catalyst, reaching an acetyl content of approximately 20%. Clear wood specimens, consisting of either heartwood or sapwood, were extracted and conditioned to equilibrium at a relative humidity of 60% and a temperature of 20 °C. The fracture energy for mode I loading in tension perpendicular to the grain was determined using single edge notched beam (SENB) specimens, subjected to three-point bending. Additionally, the modulus of elasticity along the grain and the tensile strength perpendicular to the grain were determined for sapwood specimens. The findings demonstrated a significant decrease (between 36 and 50%) in the fracture energy for the acetylated specimens, compared to the unmodified specimens. No significant effect of the acetylation process on the modulus of elasticity, nor on the tensile strength could be concluded. This study indicates that the acetylation process used results in an increased brittleness for Scots pine. Further studies are needed to analyse why the fracture energy is impaired, and to examine whether and how current timber engineering design provisions can or should be revised to account for the increased brittleness of acetylated Scots pine.

1 Introduction

Softwoods demonstrate low durability and poor dimensional stability when exposed to changes in moisture content, resulting in, for example, crack initiation caused by differential swelling, or loss of strength due to biological degradation. To overcome these drawbacks, without the use of toxic preservatives, different modification methods have been developed. The foundation of chemical modification methods lies in the possibility to change the properties of wood by changing its chemistry and these methods have

proven to be successful in limiting the hygroscopic characteristics of wood (Rowell 2006). As there is a change in chemistry of the cell wall polymers, there also is an impact on the physical properties of the wood (Rowell 1996).

Acetylation is one of the most studied chemical modification methods (Rowell 2006) and was introduced in Germany in 1928 by Fuchs (1928). The chemical process of acetylation involves a reaction of acetic anhydride with wood polymers, resulting in the esterification of accessible hydroxyl groups in the cell wall, as well as formation of the by-product acetic acid. The acetylation process is a single-addition reaction, meaning that one acetyl group is bound to one hydroxyl group, without any polymerisation (Rowell 1983). The number of free hydroxyl groups that normally bind water is reduced and substituted by hydrophobic acetyl groups. The combination of a lesser number of accessible hydroxyl groups and more hydrophobic fibres decreases the water sorption. This change in hygroscopicity results in a reduced equilibrium moisture content (EMC) and fibre saturation point (FSP) (Rowell 2006). Furthermore, acetylation impacts the wood volume. For acetylated wood with a weight percentage gain of approximately 20%, the oven-dry wood volume equals the original green volume. Hence, as the wood is in a permanently swollen state, acetylated wood

✉ Karin Forsman
karin.forsman@construction.lth.se

Erik Serrano
erik.serrano@construction.lth.se

Henrik Danielsson
henrik.danielsson@construction.lth.se

Jonas Engqvist
jonas.engqvist@solid.lth.se

¹ Division of Structural Mechanics, Faculty of Engineering LTH, Lund University, P.O. Box 118, 221 00 Lund, Sweden

² Division of Solid Mechanics, Faculty of Engineering LTH, Lund University, P.O. Box 118, 221 00 Lund, Sweden

exhibits fewer fibres per cross-section area, compared to its unmodified state (Rowell 1996).

Comprehensive studies have shown that acetylated wood exhibits enhanced dimensional stability and improved resistance to biological degradation; for compilations of studies see for example Rowell (1983), Rowell (2006) and Brelid (2013). Changing the chemical constitution of the cell wall polymers may also impact mechanical properties. For modified wood, well-studied mechanical properties are the modulus of elasticity (MOE) and modulus of rupture (MOR), determined through bending tests (see e.g. Dreher et al. 1964; Larsson and Simonson 1994; Bongers and Beckers 2003; Epmeier and Kliger 2005). Findings depend on wood species, climate conditions and utilised acetylation techniques. Previous studies have also reported that acetylated wood demonstrates improved hardness (Dreher et al. 1964; Bongers and Beckers 2003), improved compressive strength parallel and perpendicular to the grain (Dreher et al. 1964; Bongers and Beckers 2003), improved wet compressive strength (Goldstein et al. 1961) and a reduced relative creep, measured under cyclic relative humidity conditions (Epmeier and Kliger 2005). Insignificant effects from acetylation have been concluded regarding the impact strength (Goldstein et al. 1961; Bongers and Beckers 2003), while significant reductions in the shear strength have been demonstrated for various wood species (Dreher et al. 1964).

Whilst the effects of acetylation on dimensional stability, durability and basic mechanical properties have been investigated extensively over the last decades, less research concerning fracture properties has been performed. The occurrence of knots, holes, notches, moisture gradients etc., can induce large tensile stresses perpendicular to grain in structural elements which may lead to crack initiation and propagation (Gustafsson 2003). It is thus important to consider fracture properties when wood is used for structural applications. In the design of mechanical joints, fracture properties related to mode I and II are decisive, for example, when a load is applied at an angle to the grain, or in order to avoid brittle failure modes, such as splitting (Ehlbeck and Görlacher 2017). In particular, dowel-type joints have been researched in terms of brittle failure modes, for which both mode I and mode II fracture energy are of importance for the load bearing capacity (Sjödin and Serrano 2008; Jensen and Quenneville 2011; Cabrero et al. 2019). A study conducted by Reiterer and Sinn (2002) indicates a reduction in the mode I fracture energy with approximately 20% for acetylated spruce. The study also includes fracture characteristics of heat-treated spruce specimens, indicating even larger reductions in the fracture energy, in the order of 50%–80%.

The aim of this study is to further investigate the effects of acetylation on the fracture characteristics of wood, namely Scots pine. In the current paper, the term “fracture characteristics” refers to those material parameters that influence the

brittleness of the material, i.e. strength, stiffness and fracture energy. The work includes studies on the fracture energy for mode I loading in tension perpendicular to the grain, modulus of elasticity along the grain and tensile strength perpendicular to the grain for modified and unmodified specimens. The fracture energy is determined for specimens consisting of either sapwood or heartwood, whereas the stiffness and strength are only determined for specimens consisting of sapwood. The investigated material consists of wood from young logs from thinnings. Such wood is rarely used for structural purposes due to its poor durability and poor dimensional stability. By acetylation, the aim is to enhance both durability and dimensional stability. If it is possible to do so, without impairing the mechanical properties, it will enable the use of young Scots pine in outdoor load-bearing applications.

2 Materials and methods

2.1 Materials

This study investigates fracture characteristics of unmodified and modified samples of young Scots pine (*Pinus sylvestris*). The wood was provided by the sawmill Isojoen Saha, located in Finland. The logs used by the sawmill origins within 60 km from the mill, in an area well known for its fast-growing pine. For this study, the term young logs refers to wood from small-dimension sawn timber from thinnings, with an age of approximately 30–40 years, without visible unsound knots. The modification was conducted in a proprietary industrial scale process at Accsys Group’s acetylation plant in Arnhem, the Netherlands. The size of the boards was approximately 100 mm × 1000 mm × 30 mm, in the width, length and thickness directions, respectively. The acetylation process involves a reaction of the wood with acetic anhydride at an elevated temperature (approximately 120–130 °C) without the use of any catalyst, and after the reaction, the by-product acetic acid was removed (Rowell and Dickerson 2014). The wood material was acetylated according to the commercial production process for Accoya® radiata pine at Accsys Technologies, according to the standard process (European Patent No. 2818287A1, Giotra 2014). This process is developed for radiata pine, and it should be noted that it was not optimised for Scots pine. All boards used in this study were analysed for acetyl content by near infrared spectroscopy (NIRS) according to the method described by Schwanninger et al (2011). The analysis showed that all boards reached an acetyl content of approximately 20%.

The sawing pattern to extract specimens from each board is illustrated in Fig. 1. From each board three sticks were extracted. The two outermost, furthest from the pith,

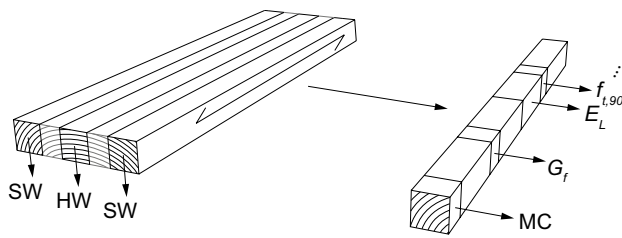


Fig. 1 Sawing pattern for each board, where 3 sticks consisting of either mainly sapwood (SW) or heartwood (HW) were extracted, from which specimens were extracted in the lengthwise direction

contained pure sapwood (SW), based on an ocular distinction between sapwood and heartwood. The inner one contained growth rings closer to the pith, i.e. contained higher fractions of juvenile wood and heartwood. Due to the variability in performance of heartwood as well as juvenile wood, results from these specimens are presented separately from pure sapwood specimens, henceforth referred to as heartwood specimens (HW). Aiming at identifying mechanical properties for clear wood specimens, knots and other imperfections were excluded when extracting specimens from each stick. Specimens originating from the same stick are referred to as nominally equal, due to similarities regarding growth ring orientations and expected limited variation in properties in general.

The fracture energy was determined for both sapwood and heartwood, based on four test groups: unmodified sapwood (USW); modified sapwood (MSW); unmodified heartwood (UHW); modified heartwood (MHW). The modulus of elasticity and the tensile strength were only determined for sapwood, based on two test groups: USW; MSW. The density was determined for each specimen, and the moisture content (MC) for one specimen in each series of nominally equal samples, i.e. one specimen per stick, see Fig. 1. The moisture content was determined by the oven dry method. Prior to testing, all samples were conditioned until equilibrium at a relative humidity (RH) of 60% and a temperature of 20 °C.

2.2 Fracture energy

The fracture energy, G_f , is defined as the energy needed to produce a unit area of traction-free crack and is measured in $[J/m^2]$. It is the energy dissipated in the fracture process zone, from fracture initiation to creation of traction-free crack surfaces, i.e. when stresses can no longer be transferred between the two fracture surfaces (Gustafsson 2003). There are a number of methods to experimentally determine fracture properties of wood. For mode I (the opening mode), some commonly used methods include the use of compact tension specimens (CT), double cantilever beams (DCB) or single edge notched beams (SENB). In this study, SENB specimens subjected to three-point-bending were used to

determine the fracture energy in mode I in tension perpendicular to the grain, according to the standard NT BUILD 422 (1993). The main reason for choosing this test method and the related evaluation methods was its simplicity. Using the approach stated in the standard, no assumptions about, for example, loading/unloading behaviour, bending/shear deformation of the specimen, the shape of the softening curve of the material, the tensile strength perpendicular to the grain, the size of the fracture process zone nor its development are necessary. Only the energy put into the system by the loading applied during the course of the test is evaluated. The most noticeable possible error involved herein is the influence of plastic dissipation at the loading point and at the supports, and plastic dissipation in the compression zone of the specimen. The influence of these sources of error is, however, expected to be negligible in relation to, for example, the variability of G_f between nominally equal specimens, and especially for small specimens like the ones used in the present study (Gustafsson 2003). The drawback when using the Nordtest method is that the information obtained is limited, i.e. only the fracture energy is evaluated. If additional parameters are of interest, for example, the shape of the softening curve of the material, including the strength of the material, more sophisticated (and complex) test and evaluation methods can be used. Such methods include, for example, the use of the so-called R-curve concept, and require additional assumptions regarding the fracture behaviour of the material (see e.g. de Moura et al. 2010; Dourado et al. 2011, 2015; Morel et al. 2005).

The fracture energy was determined for 16 unmodified and 16 modified sapwood specimens, as well as 12 unmodified and 11 modified heartwood specimens. Each specimen consisted of three wood pieces, glued together with polyvinyl acetate (PVAc), with geometry and material orientations defined according to Fig. 2a. The annual rings of the specimens were oriented aiming at a TL crack system, i.e. with the crack propagating in the longitudinal direction (fibre direction), L, and the normal to the fracture surface in the tangential direction, T. Due to variations in growth ring orientations, a pure TL crack system was difficult to achieve and the deviation was measured to be approximately 20–30° in the TR-plane, where R denotes the radial direction. The 10 mm deep notch parallel to the grain, illustrated in Fig. 2a, was cut right prior to the tests being conducted.

The specimens were simply supported and loaded in three-point-bending, according to Fig. 2b. At one end, the specimens were placed on a steel prism resting on a steel ball and at the other on a steel prism resting on a steel cylinder, which in turn rested on ball bearings to reduce influence of friction. The span between the supports was 120 mm and the load, P , was applied at the midpoint, through a rounded cross head and a steel prism with a mass of $m_{prism} = 2.69$ g. The load was applied with a material testing system, MTS

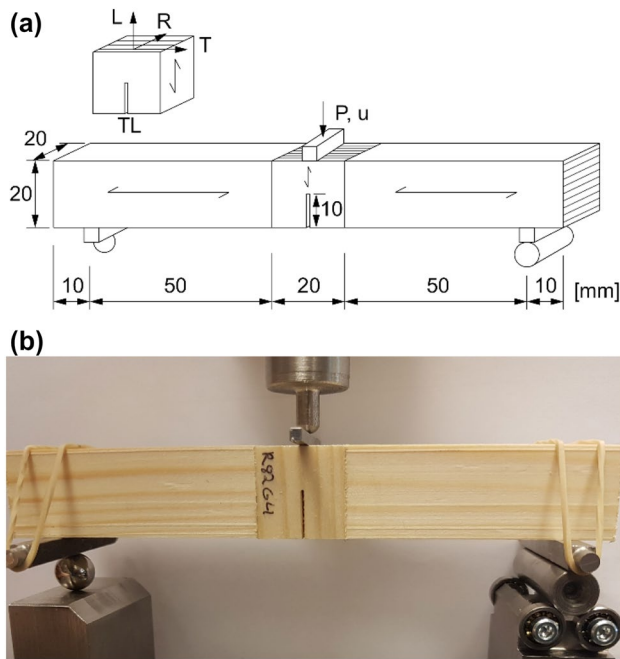


Fig. 2 SENB specimen tested in three-point-bending in the fracture energy tests, where the specimen is composed of three wood pieces glued together. **a** Specimen geometry and material orientations, where L denotes the longitudinal direction, R the radial direction and T the tangential direction of the growth ring orientation. **b** Test set-up with supports visualised

810, with displacement controlled movement of the cross head at a rate of 3 mm/min. All specimens were loaded until complete failure, recording load and mid-point displacement, the latter through the cross-head movement of the testing machine. The fracture energy was evaluated by the work done by the midpoint force and the dead weight of the specimen, divided by the fractured area, as indicated in the standard (NT BUILD 422 1993). Thus, the fracture energy perpendicular to the grain in mode I was evaluated as:

$$G_f = \frac{W + (5/6 m_{tot} + 2 m_{prism})gu_0}{A_c}$$

where m_{tot} the weight of the specimen, m_{prism} the weight of the steel prism placed under the applied midpoint force, u_0 the displacement of the cross head at failure, A_c the fractured area and g the gravity acceleration. The work done by the midpoint force, W , was determined by numerical integration of the load–displacement response, using the trapezoidal method implemented in the software MATLAB.

2.3 Modulus of elasticity

The modulus of elasticity in compression parallel to the grain, E_L , was determined for 16 modified and 16 unmodified

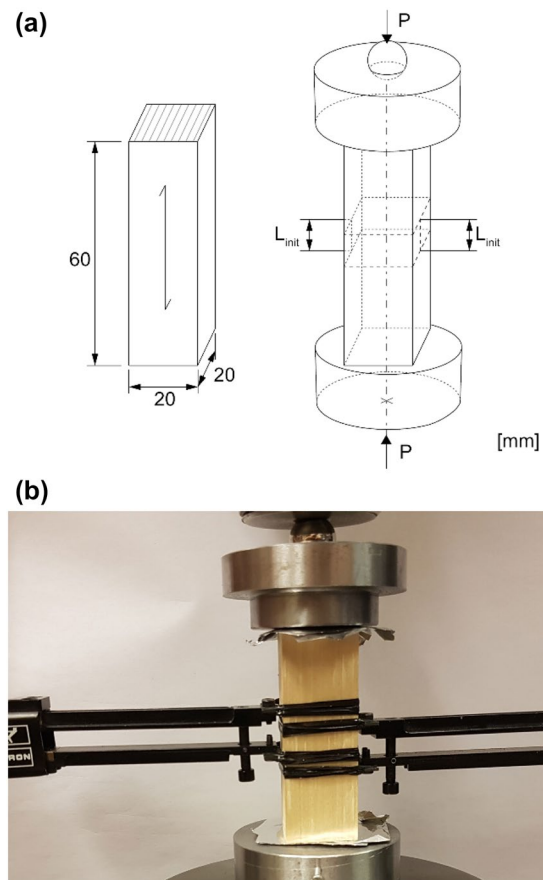


Fig. 3 Test set-up for determining the modulus of elasticity in compression along the grain. **a** Specimen geometry and location of extensometer pins, mounted on the specimen. **b** Test set-up with supports and extensometers visualised

samples consisting of sapwood. The geometry of each specimen was approximately 20 mm × 20 mm × 60 mm, in the radial, tangential and longitudinal directions, respectively, as illustrated in Fig. 3a.

The tests were conducted with a material testing system, MTS 810, and the specimens were loaded in the grain direction. On top of the specimen a steel cylinder was placed, with a steel ball placed in a centred cavity according to Fig. 3b. To reduce constraining shear forces, 3 layers of greased aluminium foil were placed in the interface between the specimen and the supports on both sides. The load, P , was applied with a displacement-controlled movement of the crosshead with a rate of 0.5 mm/min. The force was recorded by the testing machine, and the relative displacement by using two external extensometers, placed on opposite sides of the specimen as shown in Fig. 3. The initial distance between the extensometer pins mounted on the specimen was $L_{init} = 12.5$ mm.

The strain was determined based on an averaged value of the recorded relative displacements from the two

extensometers and the initial length L_{init} . The stress in the longitudinal direction was evaluated by the applied force, P , divided by the cross-section area. In order to find a proper fit to recorded data, a regression line was fitted to test data corresponding to load values in the range $2 \text{ kN} < P < 8 \text{ kN}$ (corresponding to stress values of 5–20 MPa), according to the method of least squares. The slope of the regression line was used to estimate the modulus of elasticity parallel to the grain.

2.4 Tensile strength

The tensile strength perpendicular to the grain, $f_{t,90}$, was determined for 5 unmodified and 6 modified samples consisting of sapwood. The geometry of each specimen was approximately $20 \text{ mm} \times 20 \text{ mm} \times 20 \text{ mm}$, in the radial, tangential and longitudinal directions respectively. A cylindrical notch with a radius of 5 mm was made, as illustrated in Fig. 4a. Prior to testing the specimens, they were glued with a cyanoacrylate adhesive to steel cylinders and stored at a RH of 60% and a temperature of $20 \text{ }^\circ\text{C}$ until the glue had cured. The cyanoacrylate adhesive was chosen based on pre-tests to find a suitable adhesive that would provide a combination of sufficient strength yet curing fast enough to simplify specimen handling.

The steel cylinders were connected to a material testing system, MTS 810, with hinged ends to allow mounting of the specimens without introducing any constraining forces prior to testing, see Fig. 4b. The specimens were loaded at a rate of 1 mm/min until complete failure and the applied tensile load was recorded by the testing machine. The tensile

strength, $f_{t,90}$, was evaluated by the maximum recorded load, P_{max} , divided by the fractured area A_c .

3 Results and discussion

3.1 Moisture content

Mean values and standard deviation of the moisture content (MC) for all examined test groups (USW; MSW; UHW; MHW) are presented in Table 1. The results clearly show a decreased moisture content for acetylated samples, which is an expected result (Rowell and Dickerson 2014). However, the measured moisture contents are slightly lower than values reported for acetylated Scots pine in previous studies (Epmeier and Kliger 2005). This is most probably attributed to a higher acetyl content of the samples investigated in this study, but it could also be a consequence of the examined samples being extremely dry prior to conditioning, or due to differences regarding acetylation techniques, climatic conditions and differences related to the origins of the material.

3.2 Fracture energy

All tests performed displayed a well-defined descending part of the load–displacement response, indicating a stable crack propagation. The work done by the midpoint force could thus be determined by numerical integration of the load–displacement response. In Fig. 5, typical load–displacement responses for unmodified and modified sapwood and heartwood are shown. Clear differences can be observed, for example, regarding the peak load, indicating a lowered fracture energy for the modified samples. Figure 6a illustrates a specimen under loading, as the crack propagates. In Fig. 6b, a typical fracture surface demonstrating a wavy pattern is shown, an attribute observed among all specimens.

Mean values and standard deviations for the fracture energy, G_f , and density, ρ , of corresponding specimens, are presented in Table 2. The difference in mean fracture energy between unmodified and modified sapwood, as well as heartwood, are presented in $[\text{J/m}^2]$ and $[\%]$. Compared

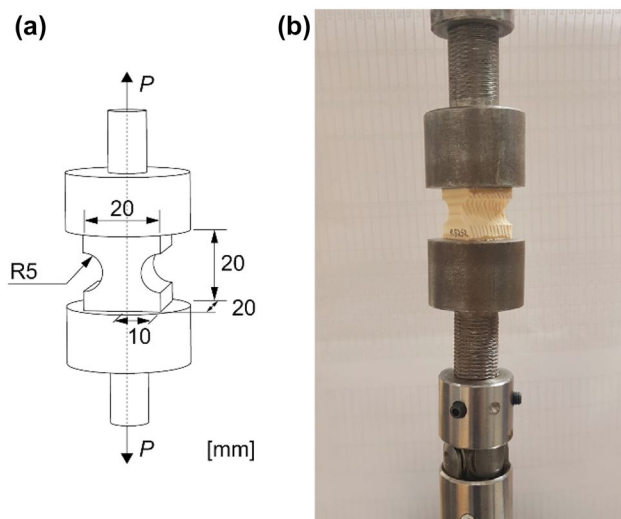


Fig. 4 Test set-up for determining the tensile strength perpendicular to the grain. **a** Specimen geometry and material orientations. **b** Test set-up with supports visualised

Table 1 Mean values of the moisture content for unmodified sapwood (USW), modified sapwood (MSW), unmodified heartwood (UHW) and modified heartwood (MHW), after conditioning at a RH of 60% and a temperature of $20 \text{ }^\circ\text{C}$ (numbers within brackets specify the standard deviations)

Test group	No. samples	MC [%]
USW	4	10.3 (0.3)
MSW	4	2.5 (0.2)
UHW	3	10.4 (0.6)
MHW	3	3.5 (0.5)

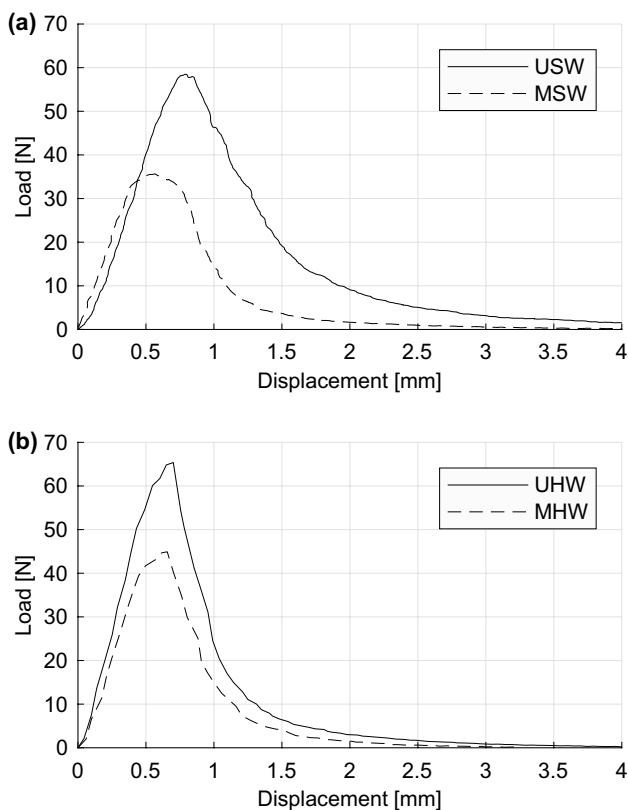


Fig. 5 Typical load–displacement responses from fracture energy tests. **a** Unmodified sapwood (USW) and modified sapwood (MSW). **b** Unmodified heartwood (UHW) and modified heartwood (MHW)

to unmodified samples, a decreased fracture energy was observed for the acetylated specimens. The mean values were 36% and 50% lower for acetylated heartwood and sapwood, respectively. The statistical significance of the difference is presented in the table through p-values and confidence intervals, according to a two-sample t-test, assuming unequal variances. There was an impact on the fracture energy at a level of significance greater than 99.9%. This observation clearly demonstrates an increased brittleness of

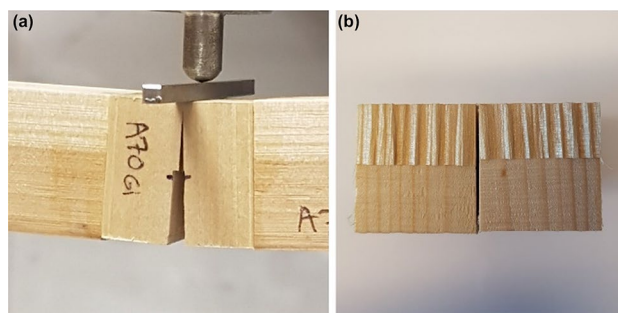


Fig. 6 Illustrations of a specimen during and after the fracture energy test. **a** Propagating crack. **b** Fracture surface

acetylated Scots pine, which is in agreement with previous studies for another species (Reiterer and Sinn 2002). However, the observed impact appears larger in the current study. This could be due to differences in acetylation techniques, acetyl content, climatic conditions or species-specific effects (Scots pine versus spruce). It should also be emphasized that the acetylation process used in this study was optimised for radiata pine and not Scots pine.

In Fig. 7, the correlation between measured fracture energy and density are illustrated for each test group: USW; MSW; UHW; MHW. Data from all tested specimens are presented and within each plot equal markers indicate data from nominally equal specimens. A regression line is fitted to the data according to the method of least squares. A previous study conducted by Larsen and Gustafsson (1990) showed a positive correlation between the fracture energy and density for unmodified European softwoods. In the current study, the observed values of fracture energies for the unmodified specimens correspond well with the values reported by Larsen and Gustafsson (1990). In the present study, however, the correlation between the fracture energy and density was very low for some cases ($R^2=0.12$, see Fig. 7a) and the corresponding trend lines indicated in Fig. 7 cannot be considered as being statistically significant. Hence, additional testing with a wider range of densities is recommended.

Table 2 Mean values of density and fracture energy for unmodified sapwood (USW), modified sapwood (MSW), unmodified heartwood (UHW) and modified heartwood (MHW), where numbers within brackets specify the standard deviation

Test group	No. samples	ρ [kg/m ³]	G_f [J/m ²]	ΔG_f [J/m ²]	ΔG_f [%]	p-value
USW	16	475 (16)	339 (27)			
MSW	16	511 (33)	169 (17)	$-170 \pm 30^*$	-50	1.5e-17
UHW	12	443 (46)	249 (23)			
MHW	11	482 (26)	158 (15)	$-91 \pm 31^*$	-36	4.9e-10

Differences in mean values of the fracture energy between unmodified and modified specimens, are presented in [J/m²] and [%]. Statistical significance according to a two-sample t-test, defined by a p-value and a confidence interval

*99.9% confidence interval

3.3 Modulus of elasticity

Results for the modulus of elasticity parallel to the grain, E_L , and the density, ρ , of the corresponding samples are presented in Table 3. Mean values and standard deviations are stated, as well as the difference in mean modulus of elasticity between unmodified and modified sapwood. The statistical significance is presented by a p-value, according to a two-sample t-test with unequal variances, and a confidence interval with a significance level of 95%. Based on

the statistical data, no significant impact on the modulus of elasticity could be concluded for specimens conditioned at the specified relative humidity and temperature.

The correlation between the modulus of elasticity and the density is presented in Fig. 8. The results are presented separately for unmodified and modified sapwood, and within each plot equal markers represent nominally equal specimens. The results suggest a positive correlation, i.e. an increasing modulus of elasticity for an increased density, which is expected (Kollmann 1968). However, as previously stated, statistically significant observations regarding

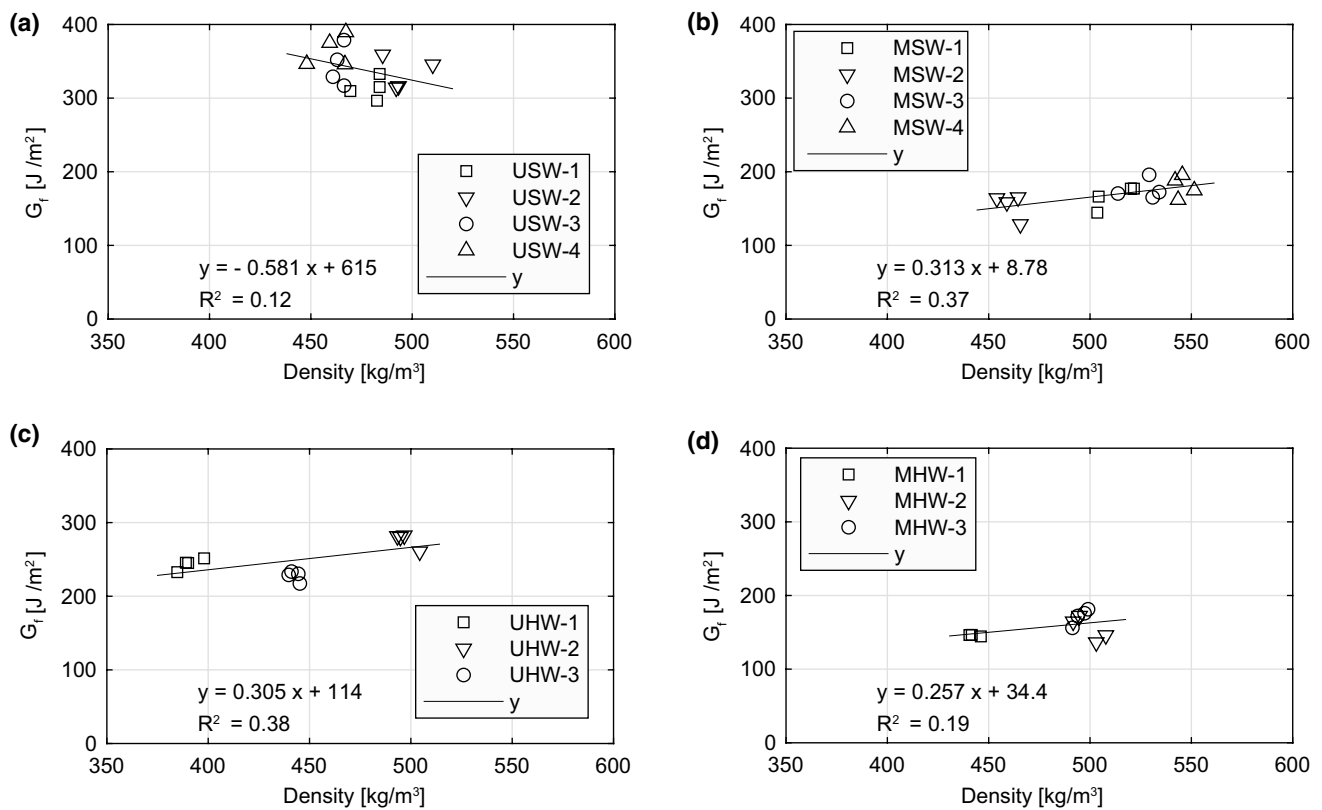


Fig. 7 Correlation between the fracture energy and the density, where nominally equal specimens are indicated with equal markers. **a** Unmodified sapwood (USW). **b** Modified sapwood (MSW). **c** Unmodified heartwood (UHW). **d** Modified heartwood (MHW)

Table 3 Mean values of density and modulus of elasticity parallel to the grain for unmodified sapwood (USW) and modified sapwood (MSW), where numbers within brackets specify the standard deviation

Test group	N.o. samples	ρ [kg/m ³]	E_L [GPa]	ΔE_L [GPa]	ΔE_L [%]	p-value
USW	16	472 (19)	12.6 (1.8)			
MSW	16	511 (35)	11.9 (0.9)	$-0.7 \pm 1^*$	- 6	0.18

Differences in mean modulus of elasticity between unmodified and modified specimens, presented in [GPa] and [%]. Statistical significance according to a two-sample t-test, defined by a p-value and a confidence interval

*95% confidence interval

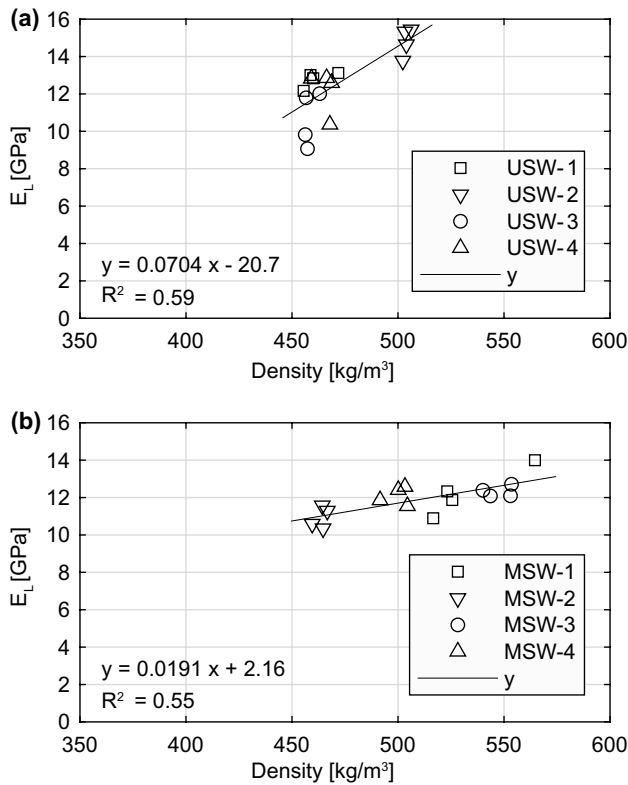


Fig. 8 Correlation between the modulus of elasticity and the density, where nominally equal specimens are indicated with equal markers. **a** Unmodified sapwood (USW). **b** Modified sapwood (MSW)

the correlation would require results from a wider range of densities and further testing is recommended.

3.4 Tensile strength

Mean values and standard deviation for the tensile strength, $f_{t,90}$, and the density, ρ , of the corresponding samples are presented in Table 4 and an example of a typical failure surface is shown in Fig. 9. The difference in mean tensile strength between unmodified and modified sapwood is presented in [MPa] and [%]. The statistical significance is defined by a p-value, according to a two-sample t-test with unequal variances, and a confidence interval with a significance level of

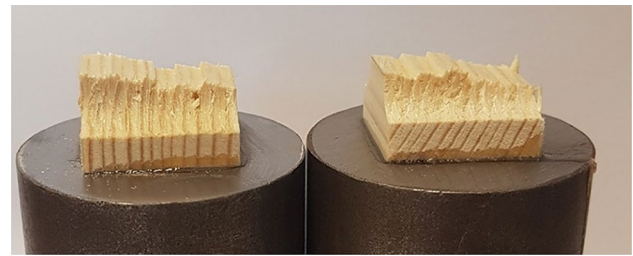


Fig. 9 A typical failure observed among the specimens in the tensile strength test

95%. Based on the result, no significant differences could be concluded regarding the tensile strength between unmodified and modified samples. However, for the tensile strength, it should be emphasised that these results only provide an indication of the effect of acetylation due to the limited number of samples.

In Fig. 10, the correlation between the tensile strength and the density is shown for unmodified and modified samples consisting of sapwood. Data from all tested specimens are presented, and within each plot, equal markers indicate data from nominally equal specimens. As for the fracture energy and the modulus of elasticity, conclusions regarding the correlation of the tensile strength to the density would require examination of a wider range of densities. Thus, further testing is recommended.

3.5 Discussion

As previously mentioned, the impact of chemical modification on mechanical properties can be considered to be a consequence of changed physical properties, caused by changing the chemistry of the cell wall polymers. As stated by Bongers and Beckers (2003) as well as Larsson and Simonson (1994), changes in the mechanical properties of modified wood can be regarded as a compilation of positive effects, gained by the lower moisture content, and negative effects, impaired by having less fibres per cross-section area. For unmodified wood, a lower moisture content correlates to increased strength and stiffness (Kollmann 1968). Based on

Table 4 Mean values of density and tensile strength perpendicular to the grain for unmodified sapwood (USW) and modified sapwood (MSW), where numbers within brackets specify the standard deviation

Test group	N.o. samples	ρ [kg/m ³]	$f_{t,90}$ [MPa]	$\Delta f_{t,90}$ [MPa]	$\Delta f_{t,90}$ [%]	p-value
USW	5	484 (8)	2.7 (0.3)			
MSW	6	501 (31)	2.5 (0.2)	$-0.2 \pm 0.4^*$	-9	0.20

Differences in mean values of the tensile strength between unmodified and modified specimens, are presented in [MPa] and [%]. Statistical significance according to a two-sample t-test, defined by a p-value and a confidence interval. *95% confidence interval

the results of the current study, no influence of acetylation on the strength nor the stiffness can be concluded. However, it should be emphasised that the current study considers samples conditioned at equal climates. Due to differences in hygroscopicity between unmodified and acetylated samples, a specific climate will result in different EMC for modified and unmodified wood. The mean moisture content for unmodified specimens was found to be 10.3–10.4%, while it was 2.5–3.5% for modified samples. Due to the strong correlation between the moisture content and the strength and stiffness for unmodified wood, the results and conclusions in this study might have been different if samples had been compared after conditioning at different climates yielding the same EMC.

In contrast to increased stiffness and strength for a decreased moisture content, studies have demonstrated that a low moisture content correlates to a decreased fracture energy (Phan et al. 2017; Reiterer and Tschegg 2002; Vasic & Stanzl-Tschegg 2007). In the current study, a significant decrease in the fracture energy was observed for the acetylated samples. To investigate why acetylation impairs the fracture energy, additional research is required. This could for instance include studies of the correlation between the fracture energy and the moisture content for modified and

unmodified samples. Findings from such a study could provide an indication of whether the decreased fracture energy is merely a consequence of a drier, hence more brittle, material. Yet, considering the use of acetylated wood in structural elements, it is still of a practical importance to compare samples subjected to equal climatic conditions, i.e. temperature and relative humidity. Similarly, this study examined samples of equal dimensions. The lower fracture energy could also be an adverse effect, caused by having less fibres per cross section area. Nevertheless, for engineering practice and in design applications, the current comparison is valid since structural design is based on nominal (gross) dimensions.

Another possible explanation for the decreased fracture energy, could be degradation of the cell wall polymers. During the acetylation process the material is subjected to elevated temperatures (approximately 120–130 °C) at drying prior to acetylation, in the reaction with acetic anhydride as well as during the removal of the by-product acetic acid. Previous studies on thermally modified wood have indicated a significant decrease in the fracture energy (Majano-Majano et al. 2012; Reiterer and Sinn 2002). Although temperatures applied in chemical modification methods are not as high as in thermal modification methods, the cell wall polymers may still be affected. Moreover, as stated by Bongers and van Zetten (2017) and Bongers and Uphill (2019), time, temperature and pressure are key parameters to attain a consistent and uniform treatment in the process of acetylation. It is important to base these parameters on a deep knowledge of the specific wood species. Otherwise, the acetylation process might lack in uniformity, resulting in an uneven distribution of acetyl groups, and might cause internal stresses, possibly resulting in the formation of cracks. As the examined material was treated with an acetylation process optimised for radiata pine and not Scots pine, it would be of interest to examine the microstructure of the wood prior to testing, to analyse the occurrence of already existing micro-cracks that might have affected the outcome of the study.

To enable the use of acetylated young Scots pine in outdoor load-bearing applications, the increased brittleness has to be regarded in the design of mechanical joints and in structures where tensile stresses perpendicular to grain appear. The fracture energy is, for example, important for the load-bearing capacity of joints, subjected to a load at an angle to the fibre direction, and when determining edge distances to avoid brittle failure modes. Some design formulae in Eurocode 5 are based on fracture mechanics, but include assumptions regarding fracture characteristics, determined through empirical testing. Further studies are needed to determine if current design codes have to be revised, to account for the increased brittleness of modified wood.

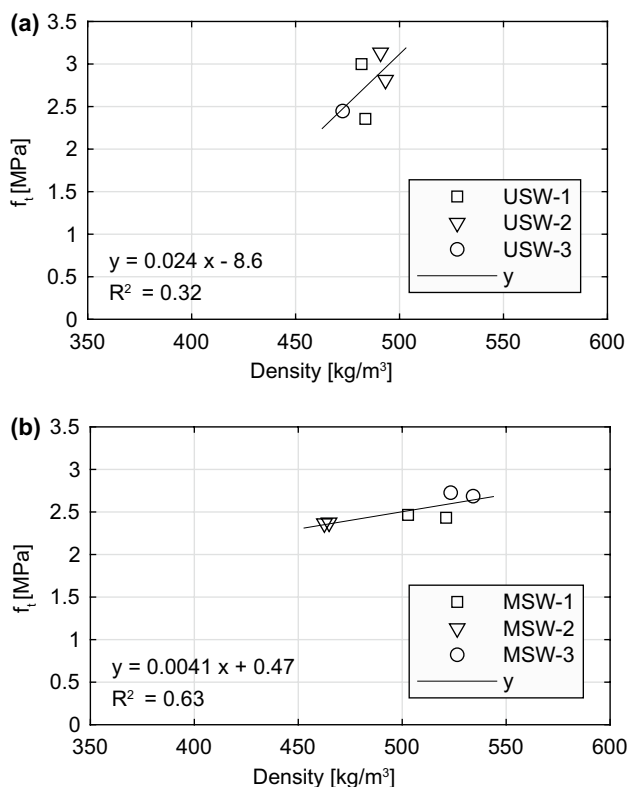


Fig. 10 Correlation between the tensile strength and the density, where nominally equal specimens are indicated with equal markers. **a** Unmodified sapwood (USW). **b** Modified sapwood (MSW)

4 Conclusion

In this study, unmodified and modified samples of Scots pine were examined. Modified samples had an acetyl content of approximately 20%, and all specimens were conditioned until equilibrium at a RH of 60% and a temperature of 20 °C. Acetylated samples demonstrated a significantly lower moisture content than unmodified samples. Significant differences were also observed regarding the fracture energy, where the mean value decreased with 36% and 50% for acetylated heartwood and sapwood, respectively. No significant effects of the acetylation regarding tensile strength perpendicular to the grain, nor modulus of elasticity parallel to the grain, could be concluded. The observations demonstrate an increased brittleness for acetylated Scots pine. This fact is important to regard in the design of mechanical joints as well as in structural elements where tensile stresses perpendicular to grain appear. Based on the knowledge gained, further studies will be conducted regarding structural applications to determine whether current design codes have to be revised, in order to account for the increased brittleness of acetylated Scots pine.

To further investigate the cause of the decreased fracture energy, both modified and unmodified specimens conditioned at a range of moisture contents should be examined. By doing so, the effect of MC on the fracture energy can be separated from other effects, such as the changed chemistry and the amount of wood fibres. Furthermore, it would be of interest to investigate the microstructure of the modified wood, to study the presence of micro-cracks and determine whether there is a degradation of the cell wall polymers.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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