Original article

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Moisture-dependency of the fracture energy of wood: a comparison of unmodified and acetylated Scots pine and birch

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Abstract: The moisture-dependency of the fracture energy for unmodified and acetylated Scots pine (*Pinus sylvestris* L.) and birch (*Betula pendula* Roth) has been investigated. Specimens were conditioned at relative humidity levels of 20, 75, and 97%, as well as dry and water-saturated. At moisture contents below 15%, the fracture energy increased with increasing moisture content for both unmodified and acetylated wood, while it decreased for untreated wood at higher moisture contents. A significant difference in moisture-dependency was found, indicating higher fracture energy for unmodified wood compared to acetylated wood at similar moisture contents. Additionally, to assess the impact of the increased brittleness for structural applications, the fracture energy was compared at equal relative humidity levels. The largest difference was seen at 75% relative humidity with approximately 50% lower fracture energy for acetylated wood. No significant differences were found for water-saturated samples. The moisture-dependency of the fracture energy, combined with the reduced hygroscopicity of acetylated wood, is suggested to be one, but not the only, contributing factor to the lower fracture energy of acetylated wood compared to unmodified wood at equal humidity levels. These observations have

importance for structural design since design codes often assess material parameters based on ambient humidity.

Keywords: acetylation; chemical modification; fracture energy; modified wood; moisture content; sorption isotherm.

1 Introduction

In order to reach milestone targets in mitigating the climate change, many operators within the building sector are exploring possibilities to increase the use of timber in load-bearing structures. An increased use of wood in *outdoor load-bearing* structures would open possibilities for new architectural expressions. This should, in turn, increase the awareness about those possibilities and about the environmental and climatic benefits associated with the use of wood in constructions. However, wood used in outdoor conditions must be protected from moisture to avoid excessive swelling and shrinking, as well as fungal degradation. To overcome these drawbacks, many different wood modification methods have been studied. These methods typically aim at modifying the physical properties of the wood to achieve a more hydrophobic material, without introducing harmful preservative substances (Rowell 2006). One promising method is acetylation, which is based on a chemical reaction between acetic anhydride and the wood polymers, resulting in the esterification of accessible hydroxyl groups in the cell wall (Rowell 1983). The resulting change of the chemical constitution of the cell wall affects most physical attributes of the material: acetylated wood exhibits a decreased equilibrium moisture content, a lower maximum cell wall moisture content (Rowell 2006), and due to bulking of the cell wall, it exhibits less fibres per cross section area compared to its unmodified state (Rowell 1996).

As reported by e.g. Brelid (2013) and Rowell (1983, 2006), acetylation indeed results in increased durability and improved dimensional stability. Today, acetylated wood commercially available is made from wood imported to Europe (*Pinus radiata* D. Don) and it is largely limited to non-structural applications, such as decking, furniture and

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facades. To reduce the climatic impact of such products, raw materials readily available in Europe could be used instead, if technically possible. Demonstrating the feasibility of using European raw materials, would also increase the economic incentive for the European forest-based industry to promote the use of acetylated wood. To make use of acetylated wood in load-bearing structures, the impact on the mechanical properties of the acetylation process must be well understood and quantified. Previous studies have investigated bending stiffness and strength of acetylated wood (Bongers and Beckers 2003; Dreher et al. 1964; Epmeier and Kliger 2005; Larsson and Simonson 1994) and results have also been reported on the impact of acetylation on compressive strength (Bongers and Beckers 2003; Dreher et al. 1964; Goldstein et al. 1961), hardness (Bongers and Beckers 2003; Dreher et al. 1964), shear strength (Dreher et al. 1964), and impact strength (Bongers and Beckers 2003; Goldstein et al. 1961).

One identified challenge is that acetylation seems to increase the brittleness of the material (Forsman et al. 2020; Lai and Plönning 2019; Reiterer and Sinn 2002). This finding is important to verify and quantify prior to large-scale use of acetylated wood in load-bearing structures, since the occurrence of knots, holes, notches, moisture gradients etc., can induce large stress concentrations, which may lead to crack initiation and propagation (Gustafsson 2003). Based on the limited number of studies on fracture characteristics of acetylated wood, the loss in mode I fracture energy has been estimated to be approximately 20–50% (Forsman et al. 2020; Lai and Plönning 2019; Reiterer and Sinn 2002). However, previous studies on the fracture characteristics have only considered unmodified and acetylated samples conditioned at equal climatic conditions, i.e. at equal temperature and in equilibrium with the same relative humidity (RH). Due to the decreased hygroscopicity of acetylated wood, this means that fracture characteristics of acetylated and unmodified samples have so far only been compared at unequal moisture contents. Thus, it is not yet known whether the increased brittleness of acetylated wood is simply an effect of the lower moisture content.

In this study, the fracture energy of unmodified and acetylated Scots pine and birch conditioned to equilibrium at various RH levels has been investigated. The results were used to examine the correlation between the moisture content and the fracture energy for unmodified and acetylated wood. The wider range of moisture contents/RH levels examined herein as compared to previous studies, makes it possible to estimate the significance of the increased brittleness of acetylated wood for moisture conditions relevant in the design of load-bearing structures. The wood materials

examined in this study have today only limited or no use for structural purposes outdoors, due to poor durability and dimensional stability. By acetylation, it is possible to increase both durability and dimensional stability. However, research on the impact of the acetylation process on fracture characteristics of the material is essential before its use in outdoor load-bearing applications.

2 Materials and methods

2.1 Wood materials

The wood materials used were Scots pine (*P. sylvestris* L.) and birch (*B. pendula* Roth). The pine was provided by a sawmill in Finland, Isojoen Saha. It consisted of small-dimension sawn timber from thinning's, aged 30–40 years in a close-by area, well-known for its fast-growing pine. The birch originated from Sweden and was provided by Vanhälls Säg AB. For birch, matched boards were investigated, i.e. one long board was split in two parts, where one part later was acetylated and the other kept unmodified. Hence, the unmodified and the modified samples of the birch had similarities in origins, density, width of growth rings, growth ring orientations etc. For the Scots pine, the unmodified and the acetylated samples originated from the same batch but not exclusively the same board.

2.2 Acetylation procedure

The modification of the acetylated boards was performed in a proprietary industrial scale process, at the Accsys Group's acetylation plant in Arnhem, the Netherlands. The standard process used in the commercial production process of Accoya radiata pine at Accsys Technologies was applied (Giotra 2014). This process involves a chemical reaction between acetic anhydride and wood polymers at elevated temperatures of approximately 120–130 °C, without the use of catalysts. It should be noted that the process parameters were not optimized neither for Scots pine nor birch, i.e. no adjustments were made regarding time, temperature, pressure or concentration levels. The modified boards were analysed for acetyl content using near infrared spectroscopy, a method described by Schwanninger et al. (2011). All the examined acetylated boards demonstrated a weight percentage gain (WPG) above 20%. The oven-dry density of the unmodified and the acetylated specimens, determined after drying at 105 °C, is presented in Table 1.

2.3 Sorption isotherms

Absorption isotherms at 20 °C were determined in a sorption balance (DVS Advantage, Surface Measurement Systems, Ltd., London, UK). A sorption balance monitors the mass of a specimen (resolution 0.1 µg) while the RH is incrementally changed in pre-programmed steps, see e.g. Williams (1995). Scots pine, acetylated Scots pine, birch and acetylated birch were cut to thin small pieces using a razor blade. The total sample mass for each wood type was about 10 mg. The sample was placed in the sample pan and dried at 0% RH/20 °C for 24 h. The sample was then equilibrated at the following RH levels: 20, 40, 60, 80

Table 1: The mean value of the oven-dry density (ρ) determined after drying at a temperature of 105 °C. The numbers within brackets specify the standard deviation.

Species	Treatment	Density, ρ (kg m ⁻³)
Pine	Unmodified	406 (5)
	Acetylated	532 (12)
Birch	Unmodified	647 (17)
	Acetylated	746 (10)

and 95%. The equilibration criterion used at a certain relative humidity step can be expressed either as a specific time or as a criterion based on the rate of change of mass with respect to time, i.e. a dm/dt -criterion. Due to uncertainties related to the use of dm/dt -criteria (Glass et al. 2017, 2018) a time criterion was chosen in the present study. The time at each RH level was 12 h, except at 95% RH where the time was set to 24 h. The sample was then dried at 60 °C for 8 h using the pre-heater in the instrument, followed by a 2 h thermal stabilisation period at 20 °C before the dry mass was determined. The equilibrium moisture content at each RH level was then evaluated as mass of water, i.e. the total mass at equilibrium minus the dry mass, divided by the dry mass of the wood. For the acetylated specimens, the moisture contents were corrected for the increase in dry mass obtained by the acetylation process, i.e. the moisture content was determined as (Thybring 2013):

$$u_0 = u_{\text{mod}} (1 + R_{\text{mod}}) \dots \% \quad (1)$$

where u_0 is the moisture content based on the dry mass before acetylation, u_{mod} is the moisture content based on the dry mass after acetylation, and R_{mod} is the relative mass increase due to the acetylation procedure. For the untreated wood, $R_{\text{mod}} = 0$.

2.4 Sample preparation

From each board consisting of either Scots pine, acetylated Scots pine, birch or acetylated birch, specimens sized 20 mm × 20 mm × 20 mm were extracted according to the pattern shown in Figure 1. To avoid influence of heartwood and juvenile wood, two sticks were extracted from the outer part of the board, one from each side of the pith (Figure 1a). In the lengthwise direction of each stick, nominally equal samples, replicates (Figure 1b), were extracted for conditioning to different RH levels (Figure 1c). For each specimen intended for fracture energy testing (denoted G), one adjacent specimen was extracted for which density and moisture content were determined (denoted U). The extracted specimens (G) were glued with polyvinyl acetate (Dana Lim, Wood Glue D3 Outdoor 430) to two wood pieces according to Figure 1d.

2.5 Sample conditioning

Prior to conditioning, all specimens were dried at 60 °C in order to ensure conditioning to the absorption isotherm and to reduce the influence of hysteresis. Specimens were conditioned at five RH levels, C1–C5 (Table 2). The specimens conditioned at C1 were oven-dried at 60 °C for seven days, and then placed in a desiccator containing molecular sieve (0.4 nm, Merck KGaA, Germany) to remain dry until tested. The specimens in conditions C2–C4 were conditioned using saturated salt-solutions in sealed boxes for 60 days, kept at a

Figure 1: The pattern used for extracting specimens from each board: Two sticks extracted from each side of the pith of the board (a). Specimens extracted for each replicate, i.e. nominally equal samples (b), and conditioning (c), where G denotes specimens for the fracture energy test and U specimens for the density and the moisture content. Specimens for the fracture energy test glued together with two more wood pieces (d).

temperature of 20 °C while measuring RH to ensure stable humidity levels over time. For condition C2, sodium hydroxide (NaOH) was used, with an expected RH level of 9% (Greenspan 1977). A higher RH was, however, noted (approximately 20%), which was most likely attributed to that the solution was not fully saturated. For conditions C3 and C4, sodium chloride (NaCl) and potassium sulphate (K_2SO_4) were used, with expected RH levels of 75 and 97% respectively (Greenspan 1977). These levels were confirmed by measurements and remained stable over time. For condition C5, the specimens were vacuum-saturated to achieve a well-defined, fully-saturated state. The water saturation was performed by placing the specimens in vacuum (<1 mbar) in a glass desiccator for 1 h, deionized water was then added while running the vacuum pump, and finally atmospheric pressure was re-established. The specimens were then kept in water for eight days before the fracture energy tests were performed.

The number of replicates for each condition and test group, i.e. series 1 and series 2 (Figure 1a) combined, is presented in Table 2, where specimens excluded due to sources of error prior or during test are noted. The number of specimens assigned to conditions C1 and C5 was lower due to an in-process decision to use some of the specimens originally assigned to condition C1, to also include water-saturated samples, C5. Moisture contents were determined by the oven-dry method and corrected for the increase in dry mass for acetylated wood (Equation (1)). For the water-saturated specimens (C5), an estimation of the cell wall moisture content was made since the fracture energy presumably is affected by the cell wall moisture content rather than the total moisture content. This estimation was performed as follows. For the unmodified wood, the volume of the voids outside of the cell walls was calculated from the measured dry bulk densities of the

Table 2: The number of specimens in the fracture energy test for conditions C1–C5 and each test group. The numbers within brackets specify the original number assigned for each test group/condition, noted where specimens were excluded due to sources of error prior to or during testing.

Species	Treatment	C1	C2	C3	C4	C5
		OD	RH 20%	RH 75%	RH 97%	WS
Pine	Unmodified	4	8	7 (8)	6 (8)	4
	Acetylated	4	8	8	8	4
Birch	Unmodified	4	8	8	8	4
	Acetylated	4	8	7 (8)	8	4

untreated wood (Table 1) and literature values of cell wall densities for the two wood species (Plötze and Niemz 2011). The amount of water in the cell walls in the saturated state was then determined by subtracting the amount of water in voids outside of cell walls from the total amount of water at saturation. In Thybring (2013), the relation between the WPG obtained by modification and the moisture exclusion efficiency is shown for cell wall bulking modifications. Based on this relation, and the estimated cell wall moisture content of the untreated specimens, the cell wall moisture content in the saturated state for the acetylated wood was determined. This methodology gave cell wall moisture contents for untreated Scots pine well in agreement with values for the same wood species measured using Low Field Nuclear Magnetic Resonance (Telkki et al. 2013). Also, the values estimated for acetylated Scots pine were in the same range as cell wall moisture contents for acetylated radiata pine in Beck et al. (2018).

2.6 Fracture energy tests

The fracture energy is the energy dissipated in the fracture process zone, from crack initiation to creation of traction-free crack surfaces. It is the energy needed to produce a unit area of traction-free crack, measured in (J m^{-2}) (Gustafsson 2003). In this study, the fracture energy was determined according to the standard NT BUILD 422 (Nordtest Method 1993), i.e. single-edge notched bend (SENB) specimens subjected to three-point bending were used to determine the mode I fracture energy in tension perpendicular to the grain. The main reason for choosing this test method and the related evaluation methods, was its simplicity: it only requires the evaluation of the energy put into the system by the load applied, and the only assumption made is the assumption of a negligible influence of plastic dissipation. The influence of plastic dissipation on the results can, as stated by Gustafsson (2003), be considered negligible for small specimens, like the ones used in this study. The current study thus included measurements of fracture energy but not measurements of the fracture process itself in terms of e.g. crack opening displacement (COD) or tracking of the crack formation and propagation. Such measurements could be an interesting further development for future studies using digital image correlation techniques (DIC). Such characterisation has been done for wood adhesive bonds (Serrano and Enquist 2005) and in more recent work for wood fracture by Dourado et al. (2015), Majano-Majano et al. (2019) and Ostapska and Malo (2020).

Prior to the test, a 10 mm notch was made in the fracture energy specimen (Figure 2b). For specimens made from pine, a rectangular notch was made, according to the standard NT BUILD 422 (Nordtest

Method 1993). For birch, an adjustment of the standard procedure was made regarding the notch geometry and a triangular ligament was realised instead of the rectangular one described in the standard. This adjustment was made in order to obtain a stable test performance, which would otherwise be difficult to obtain as found in (Lai and Plönnig 2019). The central piece was oriented aiming at a TL -crack propagation system, i.e. with the crack propagating in the longitudinal direction (L) and the normal to the fracture surface in the tangential direction (T) (Figure 2a). The deviation from a pure TL -crack propagation system was measured as 20° – 30° in the RT -plane, where R denotes the radial direction and T the tangential direction.

As shown in Figure 3, the SENB specimens were simply supported and the span between the supports was 120 mm. At one end, the specimen was placed on a steel prism resting on a steel ball, and at the other end on a steel prism resting on a steel cylinder, which in turn rested on ball bearings. The specimens were loaded in three-point bending by a load, P , applied at the midpoint with a rounded loading-nose, through a steel prism with a mass of 2.69 g. The tests were performed with a Material Testing System (MTS-810), and the load was applied with displacement-controlled movement of the cross head at a rate of 3 mm min^{-1} . All specimens were loaded until complete failure, and the load was recorded by a load cell (MTS 661.11B-02) and the midpoint displacement via the cross-head displacement of the testing system, through the build-in transducer of the machine. The fracture energy was evaluated by calculating the work done by the midpoint force and the dead-weight of the specimen, and divide this work by the fractured area, as described in the standard NT BUILD 422 (Nordtest Method 1993). The work done was determined by numerical integration of the load–displacement response, using the trapezoidal method, *trapz*, implemented in the software MATLAB (MATLAB R2017b, The MathWorks Inc., Natick, MA, US).

2.7 Evaluation of stable responses

According to the standard NT BUILD 422 (Nordtest Method 1993), only stable responses should be evaluated. Stable responses refer to load–

Figure 2: Geometry and orientations of the SENB-specimen (a) with notch geometries shown by an intersection (b) for Scots pine (rectangular ligament) and birch (triangular ligament). The coordinate systems are defined by the longitudinal (L), tangential (T) and radial (R) directions.

Figure 3: Test-setup and geometry for the fracture energy test (a), where the SENB-specimen was simply supported and loaded in three-point bending (b).

displacement responses for which it is possible to record the softening branch of the load versus deformation curve. However, no criterion for classification of the response as being stable or unstable is given in the standard. To evaluate the influence of possible unstable responses, the maximum relative load drop between two consecutive sampling points (relative to the maximum load recorded) was chosen to represent the degree of stability of the response. Thus, a restriction criterion, LC , was introduced:

$$LC = \frac{|P_i - P_{i+1}|}{P_{\max}} \times 100 \dots (\%) \quad (2)$$

with definitions according to Figure 4. In this study, the influence of varying the allowable level of LC was investigated, with the aim of verifying the sensibility of the fracture energy evaluation method to

Figure 4: Definition of the maximum load ($P_{\max}$), and the load in two consecutive sampling points (P_i and P_{i+1}), used to determine the criterion LC (Equation (2)), which defines instability of the load–displacement response.

minor instabilities. Four different LC -levels were investigated, representing allowing any, 15, 10 and 5% load drop, respectively.

3 Results and discussion

3.1 Moisture contents and sorption isotherms

As wood is a hygroscopic material, its moisture content will depend on ambient RH and temperature. To establish this relation for the materials investigated, sorption isotherms were evaluated for each test group. Figure 5 shows the sorption isotherms for the unmodified and the acetylated Scots pine and birch. As expected (Rowell and Dickerson 2014), a decreased hygroscopicity was observed for both wood species when acetylated. The moisture contents for acetylated specimens did not exceed 11% at RH levels up to 97%.

Figure 5: Sorption isotherms for the unmodified and the acetylated Scots pine (a) and birch (b), measured at a temperature of 20 °C in a sorption balance. Mean values of the measured moisture contents for the specimens (denoted U) conditioned at condition C1–C4, are marked with triangles (unmodified wood) and squares (acetylated wood). The moisture contents for acetylated wood is corrected according to Equation (1).

Moisture contents for the specimens used in the fracture energy test (evaluated for samples denoted *U* according to Figure 1) were determined after conditioning at C1–C5. Mean values of the moisture content for C1–C4 are shown in Figure 5, together with the absorption isotherms obtained by sorption balance measurements. As the expected RH levels may lack in accuracy due to various sources of error, and since a long equilibration time is needed for large specimens in climate boxes, the moisture contents for the specimens used in the fracture energy test were compared to the measured absorption isotherms. Figure 5 shows that the moisture contents for the larger specimens conditioned in climate boxes were in general well in line with the sorption isotherms determined for smaller samples. Thus, the 60-days equilibration time was indeed enough to reach equilibrium at each RH level. The results in Figure 5 also confirm that the RH level for condition C2 was higher than intended (20% instead of 9%). It can also be observed that the moisture content for the unmodified specimens tested at condition C1 deviated from zero. To remove all moisture, higher temperatures are required, but the temperature of 60 °C was chosen not to interfere with the binding of acetyl groups and wood polymers.

3.2 Typical load–displacement responses

According to the method presented, load and displacement were recorded during the fracture energy tests. To account for variations in notch geometries, and to make a comparison between the responses easier, the recorded load values, *P*, were converted to nominal stress values, σ_f , by:

$$\sigma_f = \frac{PL}{4W} \begin{cases} W = \frac{bh_c^2}{6} & \text{rectangular notch} \\ W = \frac{bh_c^2}{24} & \text{triangular notch} \end{cases}$$

where *W* is the elastic section modulus, *h_c* the height of the fractured area (Figure 2b), *b* the width of the specimen (Figure 2b), and *L* the span width between the supports (Figure 3). Typical stress–displacement responses, based on this conversion and for each condition, are shown in Figure 6. Two observations were made for both wood species: (1) No impact of the RH on the initial stiffness was seen for the acetylated samples, whereas a decreased stiffness was observed for the unmodified samples at RH levels exceeding 75%. (2) For an increased RH level above 75%, the maximum stress before softening decreased for the unmodified samples while no impact was noticed for the acetylated samples. These observations were expected as lower moisture contents correlate to higher stiffness and

Figure 6: Typical stress (σ_f) versus displacement (δ) responses for unmodified and acetylated Scots pine (a) and birch (b), after conditioning at C1–C5. The responses for each condition are presented with a 2 mm offset from each other along the x-axis.

strength (Kollmann 1968). That the initial stiffness and failure strength for the acetylated samples were not affected in the same manner can possibly be explained by the decreased hygroscopicity of acetylated wood.

3.3 Evaluation of stable responses

Depending on the restriction criteria applied to discard unstable test performance results (the relative load drop, *LC* [Equation (2)]), mean values and standard deviations of the fracture energy may vary. Figure 7 shows the fracture energy statistics for each RH level considering different *LC*:s (all, 15, 10 and 5%). As can be observed, the influence of excluding tests with unstable responses was found to be of minor importance. The overall tendencies observed (influence of RH and interrelation between unmodified and acetylated specimens) were not affected. In addition, no major impact was found regarding the mean values within the test groups. Thus, it can be concluded that including all load–displacement responses in the estimation of the fracture energy was reasonable for this study, and all the

Figure 7: The mean values and the standard deviations of the fracture energy (G_f) for unmodified and acetylated Scots pine (a) and birch (b), depending on applied restriction criterion, LC (Equation (2)). The number of included responses, n , for the different criteria are denoted in the corresponding bar.

following results will be based on the number of specimens given in Table 2, i.e. criterion “All”.

3.4 Fracture energy versus moisture content

Table 3 presents the fracture energy, G_f , and the moisture content, u_0 (Equation (1)), for RH levels C1–C5. Note that for the water-saturated samples (C5) an estimation of the cell wall moisture content, u_{CW} , is presented. As previously noted by several researchers (Bongers and Beckers 2003; Larsson and Simonson 1994) the impact of the

acetylation process on the mechanical properties of wood can be regarded as a consequence of the altered attributes (increased density, lower moisture content, less fibres per cross-section area). To visualize a possible effect of the lower moisture content, the fracture energy is plotted versus the moisture content in Figure 8. For both wood species, the following could be concluded: For unmodified wood, an increased moisture content resulted in an increased fracture energy for moisture contents up to approximately 15%, while a decreased fracture energy was observed for moisture contents exceeding this value. For acetylated wood, an increasing fracture energy was found for all the included moisture contents.

Thus, a peak (local maximum) in fracture energy was observed for the unmodified wood. Due to the limited number of data points (number of climates used for conditioning), it was not possible to draw any precise conclusions about the exact location of the peak. However, the results indicated this peak to be located at approximately 12–18% moisture content. This observation aligned with previous studies for a TL-oriented crack propagation system, where a similar behaviour was detected for untreated beech and ash at moisture contents around 13% (Majano-Majano et al. 2012) and for red pine around 18% moisture content (Smith and Chui 1994). Amorphous polymers undergo softening, i.e. they go from a glassy to a rubbery state when the temperature changes. The temperature at which this occurs is called the glass transition temperature. The transition from a glassy to a rubbery state is, however, not only dependent on the temperature, it is also moisture dependent. For hemicellulose, which is the most hygroscopic polymer in wood, glass transition occurs in the region 60–90% RH at room temperature, which corresponds to moisture contents between 10 and 20% (Engelund et al. 2013). The peak in fracture energy observed for moisture contents around 12–18% could thus be attributed to this transition. Worth noting is that in previous studies, the observed peak has only been noted for TL-oriented crack propagation systems, and the moisture-dependency of fracture energy for RL-oriented crack propagation systems has been suggested to be monotonic (Majano-Majano et al. 2012; Reiterer and Tschegg 2002). For the acetylated wood, no peak was identified in the data set. This could either be because it occurs at a moisture content where no data were obtained in the present study (between data points), or, because acetylation increases the glass transition temperature. It has, in previous studies, been speculated that acetylation may change at which moisture content/humidity level glass transition occurs at room temperature (Hunt et al. 2018; Zelinka et al. 2016).

To evaluate differences in the moisture-dependency of the fracture energy between unmodified and acetylated

Table 3: The mean values of the fracture energy (G_f) and the moisture content (u_0) for each test group/condition. The numbers within brackets specify the standard deviations. Note that for C5, an estimation of the cell wall moisture content (u_{cw}) is included, and the standard deviation given here is a result of the spread in wood density.

Species	Treatment	C1		C2		C3		C4		C5		
		O–D		RH 20%		RH 75%		RH 97%		W–S		
		G_f (J m ⁻²)	u_0 (%)	G_f (J m ⁻²)	u_0 (%)	G_f (J m ⁻²)	u_0 (%)	G_f (J m ⁻²)	u_0 (%)	G_f (J m ⁻²)	u_0 (%)	u_{cw} (%)
Pine	Unmodified	220 (26)	1.7 (0.1)	242 (19)	4.9 (0.0)	387 (18)	13.7 (0.1)	348 (16)	26.1 (0.2)	283 (18)	216.5 (3.5)	37.4 (3.1)
	Acetylated	165 (5)	0.4 (0.0)	175 (13)	1.3 (0.1)	197 (19)	4.6 (0.0)	256 (17)	8.3 (0.1)	299 (20)	144.5 (7.9)	20.3 (1.7)
Birch	Unmodified	198 (12)	1.2 (0.0)	243 (26)	4.1 (0.0)	460 (36)	13.6 (0.1)	336 (37)	29.8 (1.6)	272 (39)	118.9 (1.9)	30.9 (4.2)
	Acetylated	172 (8)	0.3 (0.0)	183 (25)	1.5 (0.1)	218 (28)	5.6 (0.1)	279 (21)	10.2 (0.2)	364 (71)	82.2 (2.6)	17.3 (2.3)

Figure 8: The fracture energy (G_f) versus moisture content (u) for unmodified and acetylated Scots pine (a) and birch (b). Markers denote the mean values and error bars the standards deviations of the fracture energy. Moisture contents for acetylated wood are corrected according to Equation (1), and for C5 the estimated intervals of the cell wall moisture contents, u_{cw} , are presented.

wood, data need to be compared within a relevant interval of moisture contents, i.e. an interval for which data is available for acetylated wood. Results corresponding to moisture contents below 15% are presented for each test group in Figure 9, along with regression lines and confidence intervals with a 95% level of significance. The results obtained were found to align with previous results for unmodified and acetylated Scots pine (Forsman et al. 2020) and birch (Lai and Plönning 2019). By means of a one-way analysis of covariance using the function *aocool* in MATLAB (MATLAB R2017b, The MathWorks Inc., Natick, MA, USA), the slopes of the regression lines for the unmodified and acetylated wood were compared, and were found to be different at a level of significance greater than 98 and 99% for Scots pine and birch, respectively. For dry wood, i.e. comparing the intercepts of the regression lines, no difference was found for birch while a significant difference at a level of 97% was suggested for Scots pine.

The findings demonstrated larger fracture energy for unmodified wood compared to acetylated wood for the moisture content levels investigated (0–15%). Thus, the increased brittleness observed for acetylated wood when compared to unmodified wood at equal *RH* levels cannot solely be explained by the reduced hygroscopicity of acetylated wood, i.e. that the moisture content was lower. Previous research (Phan et al. 2017) has suggested that the crack-bridging mechanism is predominant for the increased fracture energy at higher moisture contents, while the effect of the micro-cracking phenomenon is suggested to remain constant. This statement is also supported by

Figure 9: Regression lines along with confidence intervals for the fracture energy (G_f) versus the moisture content (u_0) for Scots pine (a) and birch (b), fitted to moisture contents below 15%. Markers denote the mean values and error bars the standards deviation of the fracture energy.

scanning electron microscope images of fracture surfaces presented by Reiterer and Tschegg (2002). In the present study, the discrepancy in fracture energy between the unmodified and the acetylated wood was found to increase along with the moisture content. Considering the findings in Phan et al. (2017) and Reiterer and Tschegg (2002), it could, thus, be hypothesized that this difference is due to differences in crack-bridging.

3.5 Fracture energy versus relative humidity

To provide an estimation of when the impact of an increased brittleness should be considered in structural design of acetylated wood, as well as the magnitude of that impact, the fracture energy for acetylated and unmodified wood were compared at equal RH levels. Accordingly, Figure 10 illustrates the difference in mean fracture energy for acetylated wood relative to unmodified wood at different RH levels, along with 95% confidence intervals. It is worth noting that the impact at different RH levels varied in a similar manner for Scots pine and birch. The following three observations were made for both wood species: (1) At RH levels up to 97%, a statistical significance of a reduced fracture energy for the acetylated wood was found. (2) The largest impact on the fracture energy was identified at 75% RH, where the loss in fracture energy for the acetylated wood compared to the unmodified wood was approximately 50%. (3) No significant impact on the fracture energy was found for the water-saturated samples.

As previously discussed, the lower fracture energy observed for acetylated wood compared to unmodified wood at equal RH levels can partly, but not solely, be explained by the reduced hygroscopicity of acetylated wood combined with the moisture-dependency of the fracture energy: Since the equilibrium moisture content of acetylated wood is lower compared to unmodified wood (according to the sorption isotherms), acetylated wood will demonstrate a lower fracture energy (according to the moisture-dependency of the fracture energy). This observation is important in structural design, where the influence of moisture on the material parameters of wood is typically considered by modification factors linked to *serviceability classes*. Such serviceability classes are typically defined in structural design codes and

Figure 10: The change in mean fracture energy (G_f) for acetylated wood relative to unmodified wood for examined conditions. The error bars represent a 95% confidence interval, based on a two-sample *t*-test assuming unequal variances.

standards based on the RH levels of the ambient climate. The observations presented in this study suggested an increased brittleness for all outdoor conditions, and due to the yearly fluctuation of the RH, the worst impact should be considered, i.e. a reduced fracture energy of 50%. As an example, the design provisions of Eurocode 5 for dowel type joints include the use of minimum edge distances in order to avoid brittle failure modes. The validity of such provision for acetylated wood can be questioned, based on the findings of this research. On the other hand, acetylated wood exhibits an increased dimensional stability and less moisture induced stress gradients. This indicates lower stress concentrations for applications where acetylated wood is used, which is when the brittleness would be decisive. Moreover, the reduced hygroscopicity of acetylated wood will have further implications on structural design. For example, reduced creep and mechano-sorptive creep has been found for acetylated wood compared to unmodified wood (Epmeier and Kliger 2005), and a study on Accoya wood (Marcroft et al. 2014) has shown a decreased impact on strength values at conditions corresponding to service class 3.

4 Conclusions

A clear moisture-dependency of the fracture energy was demonstrated for unmodified as well as acetylated wood, and a lower fracture energy for acetylated wood when compared to unmodified wood was found also at equal moisture contents. Thus, previous findings, demonstrating an increased brittleness of acetylated wood when compared to unmodified wood at equal RH levels, cannot solely be a consequence of the reduced hygroscopicity of acetylated wood. Nevertheless, the reduced hygroscopicity of acetylated wood along with the moisture-dependency of the fracture energy, contributes to the lower fracture energy found for acetylated wood when compared to unmodified wood at equal RH levels; acetylated wood exhibits a lower moisture content, thus, a lower fracture energy. This observation is of importance in practical applications, such as structural design. In this study, acetylated wood demonstrated significantly lower values of the fracture energy at relative humidity levels up to 97%. The largest impact was identified as approximately 50%, observed at 75% relative humidity. As the relative humidity outdoors has a yearly fluctuation, the worst impact should be regarded in the design of load-bearing structures.

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