Improving moisture durability of flax fibre composites by using non-dry fibres

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ABSTRACT

Non-dry flax fibre and polyester resin that has low sensitivity to moisture were used in the production of composites and the effects on flexural and moisture sorption properties of composites under wet-dry cycling were determined. Results showed that composites made of non-dry fibre have lower moisture sorption and degree of swelling and shrinking compared to composites made of dried fibre. Mean strength and modulus of composites made of non-dry fibre are 4–12% and 13–14% respectively, higher than composites made of dried fibre in longitudinal direction. Mean strength and modulus of composites made of non-dry fibre are 18–22% and 11–21% respectively, higher than composites made of dried fibre in transverse direction after the wet-dry cycling. The results suggest that composites made of non-dry fibre could be used for enhancing moisture durability of composites and lessen the time, cost and embodied energy to produce the composites, by omitting the step of drying the fibres.

Keywords: Natural fibres, Polymer-matrix composites, Moisture, Mechanical properties

1. Introduction

Natural fibre composites (NFC) present some advantages compared with composites made of synthetic fibres such as potentially lower material cost, lower density, and good specific strength and stiffness [1,2]. Moreover, natural fibres are a renewable resource whose production requires little energy and involves CO_2 absorption [3], with processing benefits which include less abrasion to tooling and less irritation to the skin and respiratory system of people handling them [4]. Despite the environmental and technical benefits of NFC, their use and applications have been limited due to their lower durability when compared to synthetic fibre composites, especially in outdoor conditions with high moisture content.

Durability of NFC involves resistance to different weathering factors that may threaten their performance over time which includes biodeterioration, photodegradation, moisture, or a combination of these influences [5,6]. The feasibility of substituting synthetic fibre composites by NFC in various applications has been the subject of many

studies since NFC exhibits strong sensitivity to moisture which leads to its degradation. Many studies suggested that to improve the durability of NFC, there should be good interfacial adhesion between the fibre and matrix and/or enhanced moisture resistance [7].

Generally, water sorption by fibres in NFC causes degradation in their mechanical properties [8]. Water sorption in NFC is dependent on many factors such as temperature, fibre loading, orientation of fibres, permeability of fibres, surface protection, area of the exposed surfaces, percentage and size of voids, hydrophilicity of the individual components and the quality of adhesion in the fibre-matrix interface [6,9]. Water sorption can lead to the swelling of the fibre, forming voids and microcracks at the fibre-matrix interface region which may result in a reduction of the mechanical properties and dimensional stability of the composites [7]. Water sorption can occur by diffusion, capillary transport and transport by microcracks, wherein, the latter two mechanisms are enhanced by ageing [6].

In the literature, several researchers have studied the effects of moisture on the durability of composites through various approaches. Some of the approaches used are Freeze-thaw cycles [10] and cold-warm cycles [11] for glass fibre composites. For NFC, some researchers used cyclic exposure to alternate wetting and drying cycles for jute fibre-reinforced phenolic composites [12] and artificial ageing through exposure to high humidity environments for flax/epoxy composites [13], while most researchers used a water immersion test on screwpine fibre/polyester composites [7], flax/epoxy composites [14–16], flax/polypropylene composite [17], hemp/polyester composites [18] and sisal/polyester composites [19]. Tensile and flexural strengths decreased by about 23–52% and 11–57% respectively, for jute fibre-reinforced phenolic composites after exposing the composites to increasing humidity levels [12]. Water sorption increased by 6–8% after 888h of water immersion while tensile strength decreased by 3–15% for 3 and 4 layers of hemp fibre/polyester composites, respectively from the same water exposure [18].

Poor adhesion between fibres and polymer matrix generates void spaces around the fibres in NFC which leads to a higher water uptake of the composites [20]. Several researchers have attempted to improve the adhesion between fibres and polymer matrix and also aimed to improve the water-resistant properties of NFC through physical and chemical surface treatment methods to change the surface structure of the fibres [2,4,8]. Mercerized sisal fibre/polyester composites showed 36% increase in tensile strength and 53% in Young's modulus while the permanganate-treated sisal fibre/polyester composites showed 25% increase in flexural strength. Moreover, treated fibre-reinforced composites sorbed less water at different temperatures compared to untreated fibre-reinforced composites [19]. Acetic anhydride-treated flax fibre/bio-based epoxy resin showed reduced moisture sorption by 65% and improved adhesion at the fibre/matrix interface [16]. Fibre treatments offer ways to improve the fibre-matrix interaction and moisture resistance property of NFC. But in some cases, however, the use of fibre surface treatments adds up to the processing cost of the fibres, making them unattractive materials for composites [2,8].

This study will use non-dry flax fibre and polyester resin that has low sensitivity to moisture in the production of composites [21] to take advantage of the moisture present in the fibre to enhance the water-resistant property of composites. Using non-dry fibre as reinforcement in composites eliminates the pre-drying step in composite preparation which would lessen the time, cost and embodied energy to produce the composites. The hypothesis to be tested is that, since the fibre already contains some moisture and is thus pre-swollen, the shrinking and swelling of the composite due to moisture sorption could

be limited, slowing its degradation and thereby improving its moisture durability. The effects of moisture sorption on NFC are commonly studied using a water immersion test and therefore the effects of swelling and shrinking of NFC due to changing environmental conditions are neglected. Although several researchers have studied the effects of moisture on NFC, few have reported the effects of high humidity – low humidity ("wet-dry") cycles, particularly on flax fibre-reinforced composites. In addition, most artificial ageing studies determined the mechanical properties of NFC right after the ageing study or after re-conditioning the composites to standard environmental conditions, but not in both conditions. Testing the mechanical properties of the composites immediately after the ageing study is useful to describe the condition of the composites at that instant while testing the composites after re-conditioning is also beneficial to determine whether the mechanical properties can be recovered from the damaging weathering exposure. Therefore, this study was set up to determine the effects of using non-dry fibre on the moisture sorption behaviour, thickness swelling and flexural properties (right after each (half) cycle and after re-conditioning) of the composites under wet-dry cycling.

2. Materials and methods

2.1. Composite material

Long unidirectional (UD) flax fibre (Flax Tape 200, Lineo) having an areal weight of 200 g/m^2 and 400 mm width was used in this study. No treatment was performed on the fibre surface. Unsaturated polyester resin (Synolite 1967-N-1, DSM) with peroxide curing agent (MEKP 50-VB, Vosschemie), accelerator (Nouryact CF-32, Akzo Nobel) and an inhibitor (NLC-10, Akzo Nobel) was used for the matrix of the composite. The concentrations of the materials used were 1% for both the curing agent and accelerator and 0.6% for the inhibitor. The Nouryact accelerator for polyester cure from Akzo Nobel is cobalt-free and it has low sensitivity to moisture.

2.2. Composite plate preparation

In order to study the effects of using non-dry fibre on composites, two kinds of composites were prepared for comparison; composite made from non-dry fibre and composite made from dried fibre. The non-dry fibre was produced by storing flax fibre in a regulated chamber at 80% RH (relative humidity) until equilibrium was reached. The 80% RH for the conditioning of non-dry fibre was chosen in this study to represent fibres stored in an outdoor environmental condition e.g. in Western Europe where the average outdoor RH in a year is about 80%. The moisture content (MC) of non-dry flax fibre was about 10%. Whereas, the dried fibre was obtained by drying in an oven at 80 °C to constant mass. The drying temperature of 80 °C, which is less than the standard for oven-drying plant fibres (105 °C) was chosen to prevent a significant drop in fibre strength and in a similar way in composite strength [22].

The composite laminates were made using vacuum-assisted resin infusion technique (VARI). Composites (dried fibre/polyester and non-dry fibre/polyester) with dimensions 300 mm x 300 mm and fibre volume fraction of ~35% were prepared in the study. A mould consisting of a top plate, bottom plate and two spacers was used to produce a composite with a thickness of 2 mm.

A vacuum of 0.6 bar was applied to the infusion process that was performed at ambient temperature. After the fibre was completely impregnated with the resin, the laminate was post-cured at room temperature for 24 h followed by oven drying at 70 °C for 12 h. At the end of this procedure, differential scanning calorimetry analysis of matrix

samples was used to determine if the curing process was completed. Using a band saw, the laminate was cut into smaller pieces for sample collection for the physical and mechanical tests. Then, they were ground using a double disc polishing/grinding machine with 300 and 800 grit size abrasive papers. After cutting, all samples both in longitudinal and transverse fibre orientations were equilibrated in a conditioned room maintained at 22 °C and 50% RH given sufficient time to equilibrate in mass. This was monitored by weighing 10 representative samples of each laminate at different time intervals until the MC reached saturation. Table 1 shows some of the properties of the composites after production and conditioning.

(Insert Table 1)

2.3. Accelerated ageing procedure

The ageing study was performed under "wet-dry" cycles, using an oven and salt solutions of potassium nitrate and magnesium chloride for 10 consecutive weeks. It consisted of five cycles divided into wet/humid and dry conditions. In the wet condition, the temperature and RH were maintained at 40 °C and 89%, respectively while for the dry condition, the temperature and RH were maintained at 40 °C and 32%, respectively. Before the ageing study, the samples were pre-conditioned at 22 °C and 50% RH, which is the average Benelux standard indoor condition as well as the international standard laboratory environment, for approximately 35 days.

A single cycle had a total period of two weeks and the wet and dry conditions were alternated every week. Six samples from each type of composite at each time step were used to monitor the change in weight, thickness and flexural properties of composites under the wet-dry cycles. Samples were not taken from one laminate, but e.g. 2 samples from laminate 1, 2 samples from laminate 2, and 2 samples from laminate 3, etc., to rule out manufacturing effects. The accelerated ageing procedure adopted in the study intends to simulate both hot/wet and hot/dry environmental conditions that might be experienced by the composites in either outdoor or indoor applications.

2.4. Physical and mechanical characterization

The physical and mechanical properties of samples from each composite type were evaluated. All tests were performed on six samples and the average values were calculated. Data were statistically analyzed by one-way ANOVA using Microsoft Excel with a confidence interval of 95%.

2.4.1. Flexural test

Flexural tests of the composites were performed using a universal testing machine (Instron 5567) with a 1 kN load cell and cross-head speed of 0.85 mm/min. All tests on aged samples were performed after each (half) cycle and after re-conditioning for 30 days in the conditioned room. Unaged samples were also tested to compare the obtained flexural test results with those of aged materials. Both longitudinal and transverse fibre-oriented composites were tested according to ASTM D790 standard on a 60 mm x 12.7 mm sample, with a test span of 32 mm.

2.4.2. Weight loss or gain

Weight change of samples subjected to the artificial ageing experiment (Δw) was determined using Eq. (1), where w_2 is the weight of the samples at the end of each (half) cycle and w_1 is the weight of the samples at initial time (conditioned at 22 °C and 50% RH). The weight of samples was determined using an analytical balance (sensitivity ± 0.0001 g) at pre-determined time intervals.

$$\Delta w = \left(\frac{w_2 - w_1}{w_1}\right) x100 \qquad (1)$$

2.4.3. Thickness swelling

Swelling in thickness (Δt) after the artificial accelerated ageing study was measured on 60 mm x 12.7 mm samples using Eq. (2), where t_2 is the average thickness of samples at the end of each (half) cycle and t_1 is the average thickness of the samples at initial time (conditioned at 22 °C and 50% RH). The thickness was measured using a Mitutoyo digital micrometre with a precision of ± 0.001 mm. Three measurements were performed along the length of each specimen and the average values were recorded.

$$\Delta t = \left(\frac{t_2 - t_1}{t_1}\right) x100 \qquad (2)$$

2.4.4. Composite microstructure

The microstructure of composites was determined before and after the artificial ageing study by computed tomography at micrometre scale (micro-CT) using a Phoenix NanoTom S and scanning electron microscopy (SEM) using a Philips XL-30 FEG SEM. For micro-CT analysis, the samples were examined with a voxel size of 2 µm and with applied voltage and current of 50 kV and 260 µA, respectively. A 2D image analysis tool, ImageJ was used to enhance the reconstructed image from micro-CT to examine the presence of different components and damage (if any) of the composites before and after the ageing study.

For SEM, the samples were coated with platinum of thickness 5 nm before the imaging and then examined using 10 kV acceleration voltage. Polishing was not performed during sample preparation to prevent its damaging effect on unaged and aged composites by mechanical damage or chemical damage from using liquids such as soapy water and alcohol. Therefore, some artefacts e.g. scratches and lines due to preparation of the fresh samples could still be seen in the SEM images.

3. Results and discussion

3.1. Weight loss or gain

The results are presented with a composite coding system. Composites made of dried and non-dry fibre in longitudinal fibre orientation are represented with the codes DL and UL, respectively. While composites made of dried and non-dry fibre in transverse fibre orientation are represented with the codes DT and UT, respectively.

The moisture sorption of composites and resin throughout the moisture cycling test is presented in Fig.1. The data points of all the figures in this study are actual experimental values and fitting curves were not used; data points were merely connected by straight lines to clearly visualize the trends. In Fig. 1a, the MC of samples after conditioning at 22 °C and 50% RH are shown as zero moisture sorption while in Fig. 1b, the graphs are constructed so that the oven-dried MC of samples are shown as zero moisture sorption. The samples sorb moisture when RH increases and desorb moisture when RH decreases. A similar trend for moisture sorption is observed for all samples. Reasonably, it can be observed that throughout the moisture cycling test the composites made of non-dry fibre have lower moisture sorption compared to composites made of dried fibre at wet condition and desorb moisture to less than the original moisture content

at dry condition (Fig. 1a). Polyester resin is relatively hydrophobic in nature and therefore, pure resin samples had negligible water sorption.

As shown in Fig. 1a, DL and DT sorbed 2.4% and 2.6% of moisture, respectively at cycle 0.5 while UL and UT sorbed 1.4% and 1.6% of moisture, respectively. The lower moisture sorption of UL and UT at wet condition may be expected since composites made of non-dry fibre still contains more moisture at the start of the ageing study (see table 1), hence, its capacity to sorb moisture is less compared to composites made of dried fibre. Water sorbed in fibres is attached to the hydrophilic groups of the fibre material and some other water molecules are attracted either to other hydrophilic groups or they may form further layers on top of the water molecules already sorbed [23]. Therefore, UL and UT have lower moisture sorption than DL and DT at wet condition, possibly because a greater part of the hydrophilic groups of the fibres are already occupied by water and therefore the composites made of non-dry fibre can only accommodate lesser amount of moisture compared to composites made of dried fibre. However, it is surprising (see Fig. 1b) that the dried samples eventually show a slightly higher absolute MC in wet condition than the non-dry fibres (when comparing longitudinal and transverse samples separately), although perhaps a larger constraining effect on fibre swelling would be expected in the dried case, because the composite was consolidated with fibres in shrunken state. This indicates that the dried samples may already show some damage (cracking due to fibre swelling) after the first half cycle.

(Insert Figure 1)

The amounts of sorbed and desorbed moisture of composites over time between 89% and 32% RH at 40 °C are shown in Fig. 2. Based on the plots it can be observed that no sample had reached the equilibrium moisture content after seven days in either wet or dry conditions. Similar trends were observed for the moisture sorption of samples in other cycles of this study which are not displayed in Fig. 2. This, by the way, does not reduce the usefulness of the current test set-up in artificially ageing the samples. The current test set-up was chosen so as to reach about 80% of saturation after 1 week, whilst still using realistic temperature conditions; this to prevent an extremely long testing protocol. In general, it can be observed that composites made of non-dry fibre have considerably lower moisture sorption and desorption behaviour than composites made of dried fibre. This again suggests that the fully dried samples already sustained damage in the first half cycle.

(Insert Figure 2)

3.2. Thickness swelling

Fig. 3 shows the swelling and shrinking in thickness of composites throughout the wet-dry cycles. In the graph, the thickness of composites after equilibration at 22 °C and 50% RH or prior to the wet-dry cycles is shown as zero thickness swelling. It can be observed that relative to their initial thickness, DL and DT swell more compared to UL and UT at wet condition while for UL and UT, at dry condition, the thickness shrinks to less than the starting thickness. This way the behaviour corresponds to the moisture sorption and desorption behaviour in Fig. 1.

Therefore, the swelling and shrinking of composites were logically observed as a result of loss or gain of moisture from the composites. Swelling and shrinking were approximately similar for DL and DT. A maximum of 2.3% and 2.4% swelling, and 0.2% and 0% shrinking for DL and DT respectively were recorded. Swelling and shrinking were slightly higher for UT compared to UL. A maximum of 0.9% and 0.6% swelling,

and -0.5% and -0.3% shrinking for UT and UL respectively were recorded. It was observed that thickness swelling values of UT and UL became negative at dry conditions which indicated the composites shrink to less than the original thickness. This is logical because the original thickness was after equilibration at 50% RH.

The graph generally shows that the degree of swelling and shrinking of UL and UT was lower compared to DL and DT. This result is reasonable since composite made of non-dry fibre has also considerably lower moisture sorption and desorption behaviour than composite made of dried fibre, as was shown in Fig. 1. Moreover, the conditioning of composites before the start of the wet-dry cycles could possibly have caused damage to composites made of dried fibre. During the conditioning, moisture sorption and swelling were observed for the pre-dried composites, as was shown in Table 1, which could already have caused some micro-damage.

(Insert Figure 3)

3.3. Flexural test

Fig. 4 shows the flexural properties of composites in longitudinal fibre orientation subjected to wet-dry cycles. Flexural properties of composites were determined right after each (half) cycle and after re-conditioning for 30 days in standard conditions. Composites were tested at 0, 0.5, 1, 2.5, 3, 4.5 and 5 cycles to measure the development of damage resulting from increasing number of cycles.

The results showed that composites made of non-dry fibre have higher flexural properties than composites made of dried fibre at different cycles. It was also evident that flexural properties of both composites were lower at wet conditions compared to dry conditions. At cycle 0.5, higher reductions in the flexural properties were observed in composites made of dried fibre compared to the flexural properties of composites made of non-dry fibre, both when tested directly or after re-conditioning. Reductions of 17% and 34% in the mean strength and mean modulus were recorded for DL respectively, while for UL, reductions of 8% and 27% in the mean strength and mean modulus were observed respectively when the composites were tested right after each (half) cycle. The mean strength and mean modulus of DL were reduced by 6% and 16% respectively, while for UL, the mean modulus was reduced by 3% while its mean strength remained approximately the same after the composites were re-conditioned before being tested.

The flexural properties of both composites decrease after subjecting the composites to wet conditions. Possible reasons for the reduction in flexural properties of composites are the swelling of the flax fibres when moisture penetrates into them, forming voids and microcracks at the fibre-matrix interface region which may result in deterioration of the mechanical properties and dimensional stability of the composites [7]. In addition, the sorbed water acts like a plasticizer influencing fibre, resin and interface simultaneously and therefore affecting mechanical properties of composites [24]. In contrast, the flexural properties of both composites increase or approximately remain the same after subjecting the composites at dry condition throughout the wet-dry cycles. These were shown in the flexural properties of composites at cycles 1, 3 and 5. The better retention of the flexural properties of the composites in dry conditions is likely due to the reversal of plasticization effects of the resin and fibre [25].

In general, it can be observed that at cycle 0.5, the flexural properties of composites made of dried fibre are lower (and sometimes significantly lower) compared to composites made of non-dry fibre, both immediately tested or after re-conditioning. The results of the flexural test also showed that the properties of the composites made of non-dry fibre seem to recover after subjecting the composites at dry condition until the end of

the wet-dry cycles compared to composites made of dried fibre. The mean strength of DL was reduced significantly (17%) when the composites were tested right after cycle 0.5, whereas a relatively low decrease (8%) was observed for UL. The observed behaviour is close to what is reported in the literature for natural fibre-based composites subjected to wet condition. In the case of jute fibre-reinforced composites, approximately 25% decrease in flexural strength has been found after subjecting the composites at 85% RH [12] while for pultruded jute/phenolic composites, a decrease of approximately 15% was observed after subjecting the composites at 95% RH and 50°C.

At the end of the wet-dry cycles, the mean strength and mean modulus of UL were 12% and 13% respectively, higher than the mean strength and mean modulus of DL when the composites were tested right after the (half) cycle. The mean strength and mean modulus of UL were 4% and 14% respectively, higher than mean strength and mean modulus of DL after the composites were re-conditioned before being tested.

It can be observed that most flexural properties of composites were lower when tested right after each (half) cycle than when re-conditioned before being tested. Overall, the composites made of non-dry fibre did not result in significant decreases in flexural properties throughout the wet-dry cycles when tested right after each (half) cycle compared to composites made of dried fibre.

(Insert Figure 4)

Fig. 5 shows the flexural properties of composites in transverse fibre orientation subjected to wet-dry cycles. It is immediately apparent that the differences between dried and non-dry samples are enhanced when compared to the longitudinal test results. This is logical because the longitudinal properties are dominated by longitudinal fibre properties, whereas the effect of damage due to swelling and shrinkage should be more visible in transverse direction.

At cycle 0.5, higher reductions in the flexural properties were observed in composites made of dried fibre compared to the flexural properties of composites made of non-dry fibre, both when immediately tested and after re-conditioning. Reductions of 32% and 37% in the mean strength and mean modulus were recorded for DT respectively, while for UT, reductions of 20% and 36% in the mean strength and mean modulus were observed respectively when the composites were tested right after cycle 0.5. The mean strength and mean modulus of DT were reduced by 22% and 31% respectively, while for UT, the mean strength and mean modulus were reduced by 5% and 19% respectively after the composites were re-conditioned before being tested.

(Insert Figure 5)

Similar to the flexural properties of composites in longitudinal fibre orientations at cycle 0.5, the composites made of dried fibre exhibited a high drop in the flexural properties in transverse fibre orientations. This is not the case for the composites made of non-dry fibre. Possibly, the composites made of dried fibre already have substantial damage in the very first half cycle, therefore, the flexural properties of composites did not recover to values at the start of the test after subjecting the composites at dry condition. Cross-sections of dried and non-dry materials after cycle 0.5 are compared in Fig. 6. DT appeared more degraded with several cracks at the fibre-matrix interface and inside technical fibre bundles when compared to UT, which showed little apparent damage.

(Insert Figure 6)

The wet-dry cycles affected the mean moduli of both composites in transverse fibre orientation more compared to the moduli of composites in longitudinal fibre orientation, both immediately after each cycle and after re-conditioning. At cycle 5, the mean moduli of DT and UT decreased by 37% and 22% respectively, compared to the initial moduli of both composites, when the composites were tested right after the cycle. The mean moduli of DT and UT decreased by 31% and 25% respectively, compared to the initial moduli, when both composites were re-conditioned before being tested. Overall, the results showed that the mean moduli of composites both in longitudinal fibre orientation and transverse fibre orientation were significantly more sensitive to wet-dry cycling than the mean strengths of composites. This was also observed in flax fibre/epoxy composites aged at 90% RH for 3 days, wherein the ageing study induced a significant reduction in the modulus (33%) but only a slight decrease in strength (12%) of composites [26].

At the end of the wet-dry cycles, the mean strength and mean modulus of UT were 22% and 21% respectively, higher than the mean strength and mean modulus of DT when the composites were tested right after the fifth cycle. While the mean strength and mean modulus of UT were 18% and 11% respectively, higher than the mean strength and mean modulus of DT after the samples were re-conditioned before being tested.

3.4. Composite microstructure

Fig. 7 shows the micro-CT and SEM images of unaged and aged microstructure of composites. All images are from DT samples immediately tested after cycle 5. Images of UT samples are not shown in the figure since both composites showed approximately the same surface microstructure at the end of the ageing study. This was perhaps expected because as was presented in Fig. 5, both composites eventually showed a decrease in mechanical properties after the wet-dry cycling. The DT samples showed substantially more mechanical decay though, but this is not easy to deduce from the images.

SEM provides valuable information regarding the morphology of the composites but is restricted only to the sample surface. Thus, composite microstructures before and after the ageing study have also been assessed using the micro-CT scan. The 2D images obtained using micro-CT with the help of a 2D image analysis tool, can be used to examine the presence of voids, size of different components of the composites and several damages in composites including debonding at the fibre-matrix interface and cracks in the matrix, once these features surpass the resolution of the equipment, which is in the order of 1 to 2 micron.

The micro-CT image (Fig. 7a) shows the porosity and fibres in light grey colour and resin in dark grey colour in composites prior to wet-dry cycles. After the accelerated ageing, some differences in the microstructure of composites can be observed. As shown in Fig. 7d and 7e, after the wet-dry cycles, the composite structure appeared degraded with several cracks caused by swelling and shrinking. This explains that the flexural properties of composites were affected by the ageing study.

(Insert Figure 7)

After the flexural examination, the broken samples of composites in transverse fibre orientations are shown in Fig. 8. Micro-CT images of the fractured part of unaged composites showed that fractures occurred in the matrix and at the fibre-matrix interface (Fig. 8a) while for aged composite, fractures occurred mostly in the fibre-matrix interface (Fig. 8d), but also within some technical fibres. The same result was also observed using SEM.

The appearance of the fibres before and after the ageing study is also relatively different. As shown in the SEM images (Fig. 8c, 8f), large amounts of matrix are still attached to the fibres of unaged composites, however, in the case of aged composites, the fibres appear to have a smooth surface and the amount of resin attached on the surface of the fibre has been reduced. This phenomenon indicates a poor fibre-matrix interfacial bonding after ageing and therefore, fractures occur at the fibre-matrix interface leading to the reduction of the flexural properties of composites [16,27].

(Insert Figure 8)

4. Conclusions

Composites made of non-dry fibre have considerably lower moisture sorption and desorption behaviour and degree of swelling and shrinking than composites made of dried fibre. Flexural properties of composites made of non-dry fibre were higher than composites made of dried fibre in longitudinal and transverse direction after the wet-dry cycles. The starting hypothesis that pre-swelling the fibres will lead to lower stresses and subsequent damage due to fibre swelling is endorsed. Too much pre-swelling however, would lead to stresses due to shrinkage in drier environments.

Although the composites did not reach the equilibrium moisture content in neither wet nor dry conditions, we could restrict the study duration to 10 weeks because of the ageing procedure used. The results suggest that composites made of non-dry fibre and resin that have low sensitivity to moisture could be used for enhancing moisture durability of composites and at the same time lessen the cost and time to produce the composites, by eliminating the drying step and drying precautions. Natural fibre composites should be manufactured with fibres conditioned in average humidity conditions they will face during their lifetime, so they are in equilibrium with their environment. This would mean e.g. conditioning for outdoor conditions in Western Europe at around 80% RH and at 50% RH for average indoor conditions.

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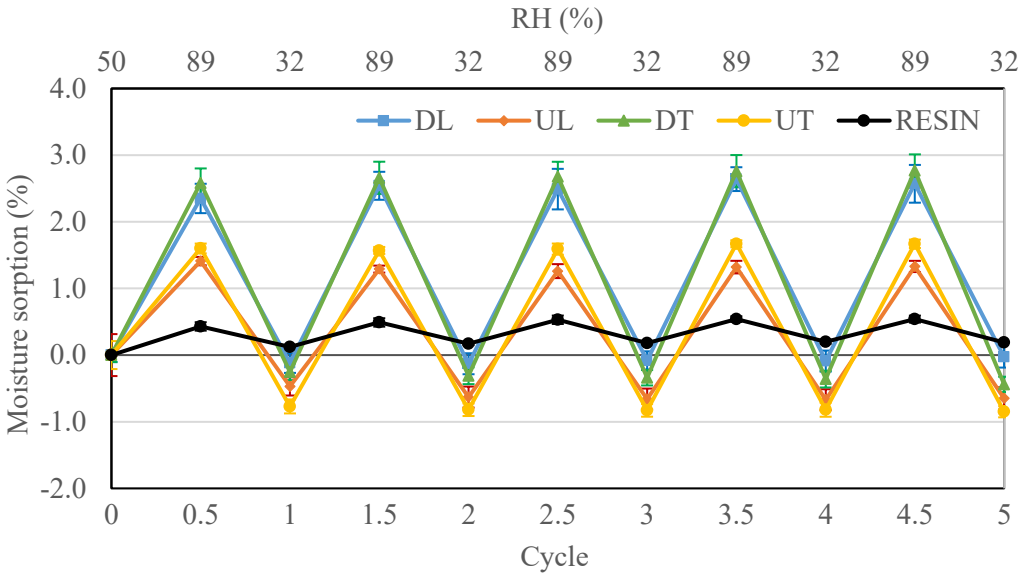
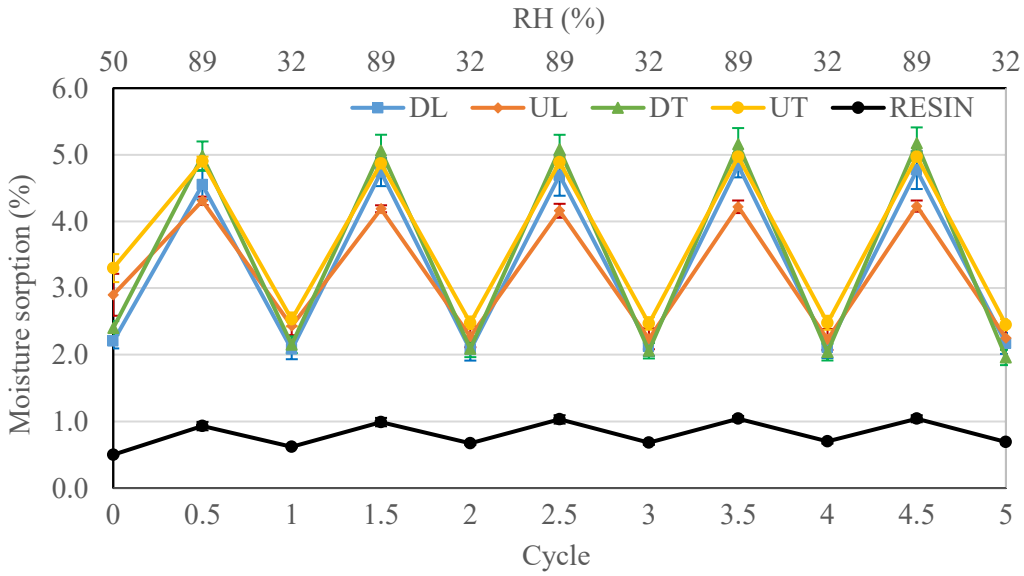


(a)



(b)

Fig. 1. Moisture sorption of composites and resin at different cycles: (a) 0% moisture sorption at start of test is for samples equilibrated at standard conditions (see Table 1) and (b) 0% moisture sorption at start of test is the oven-dried MC of samples; right after production the dried samples contain 0,9% of moisture and the non-dry samples 3,65% (see Table 1); DL = dried longitudinal, UL = non-dry longitudinal, DT = dried transversal, UT = non-dry transversal.

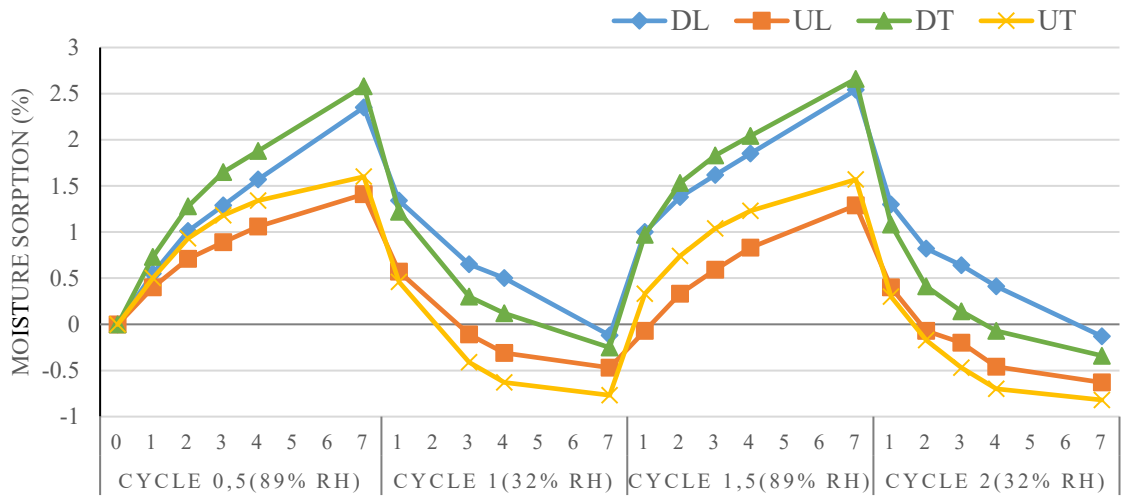


Fig. 2. Moisture sorption of composites over time between the wet and dry conditions.

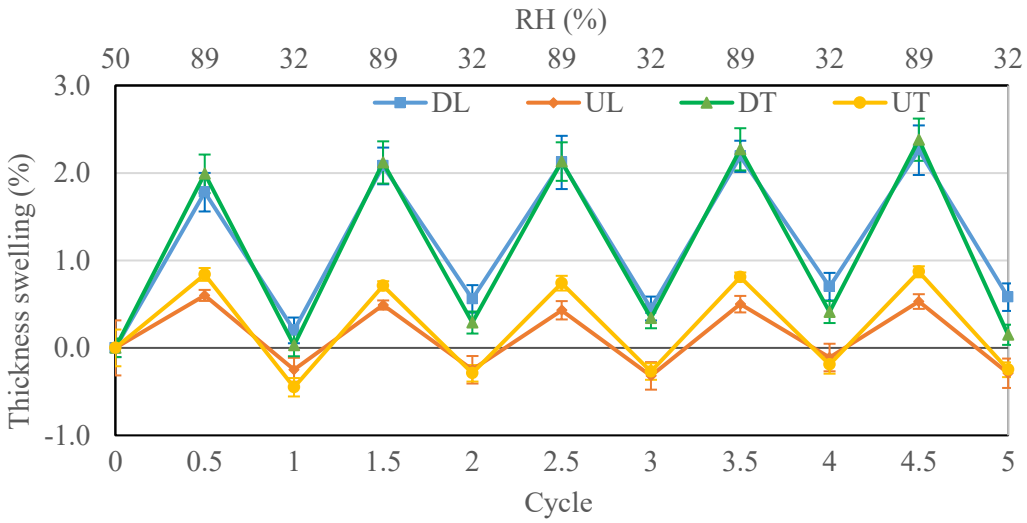
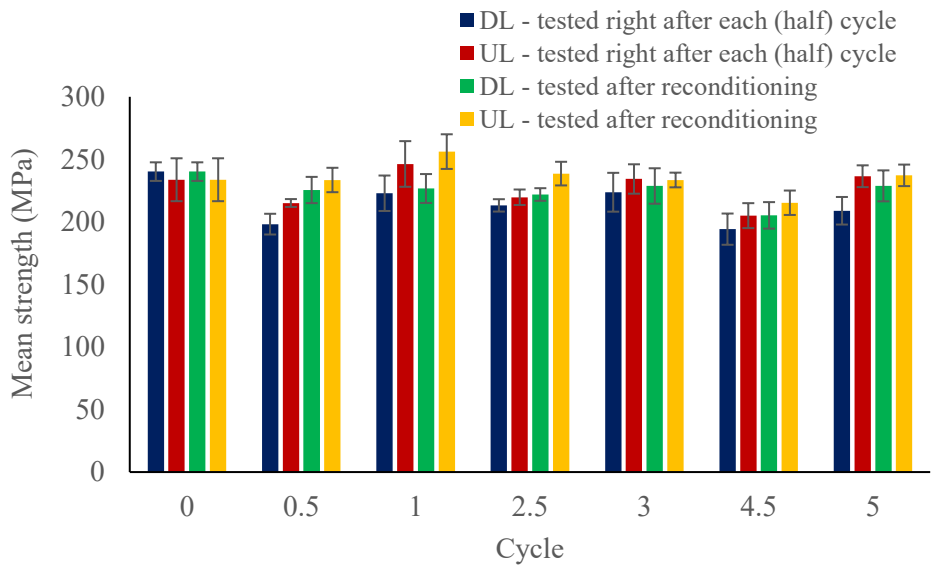
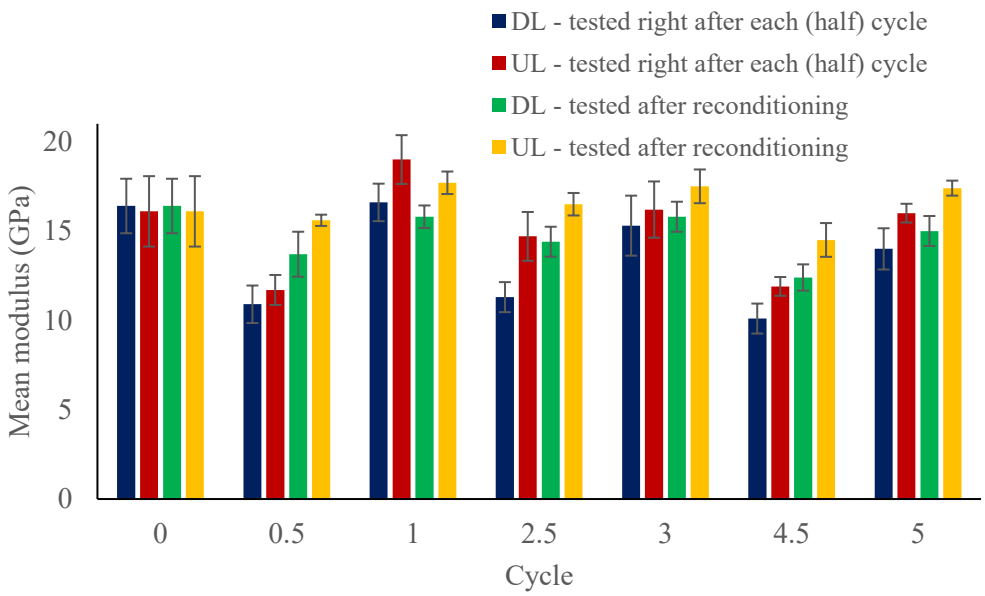


Fig. 3. Thickness swelling of the composites at different cycles; samples at start equilibrated at standard conditions (see Table 1).

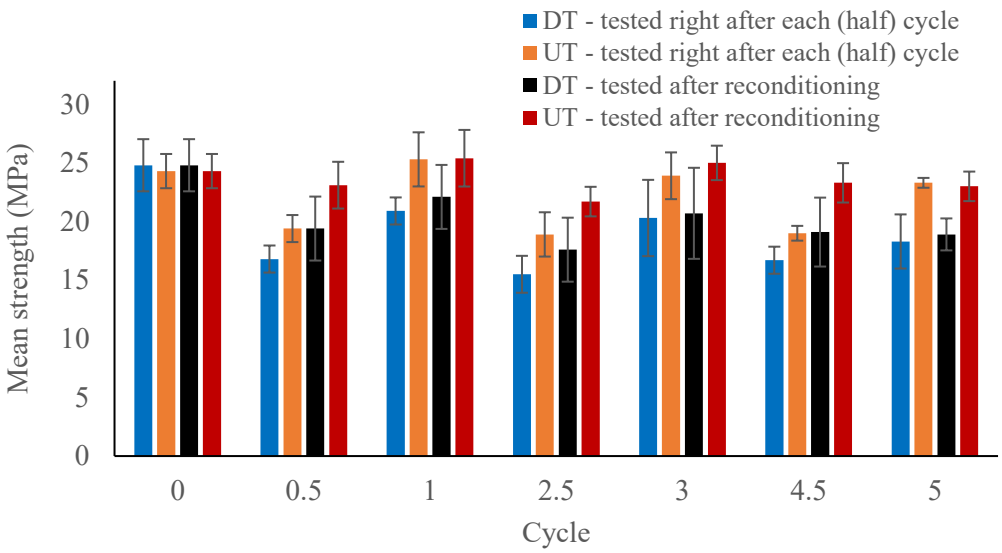


(a)

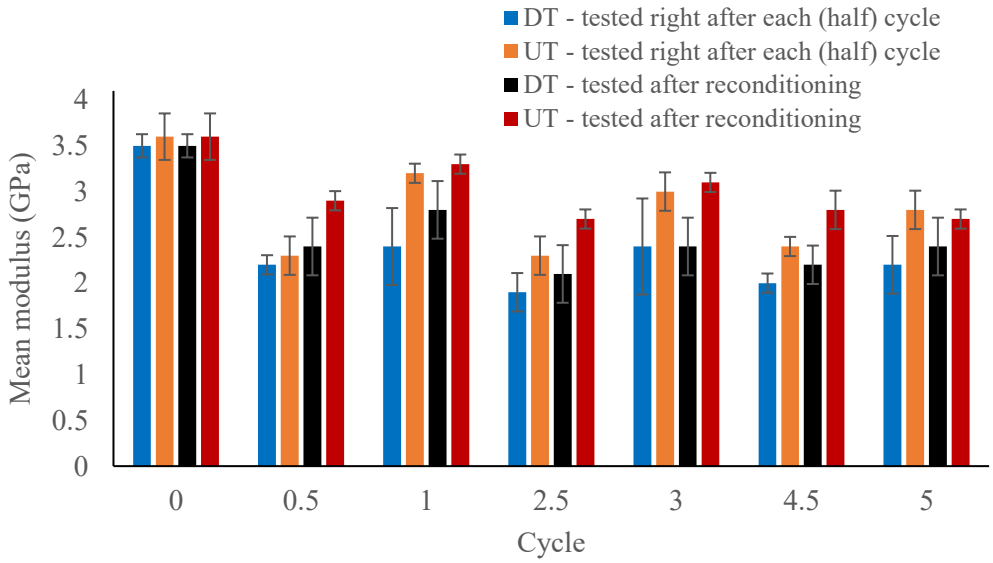


(b)

Fig. 4. Flexural properties of composites in longitudinal fibre orientation at different cycles: (a) mean strength and (b) mean modulus.



(a)



(b)

Fig. 5. Flexural properties of composites in transverse fibre orientation at different cycles: (a) mean strength and (b) mean modulus.

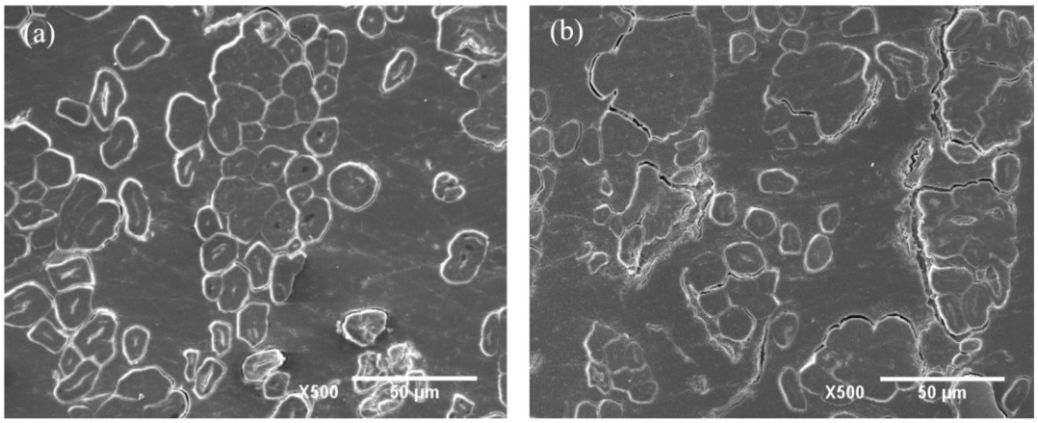


Fig. 6. SEM images of composites in transverse fibre orientation after half cycle: (a) UT and (b) DT.

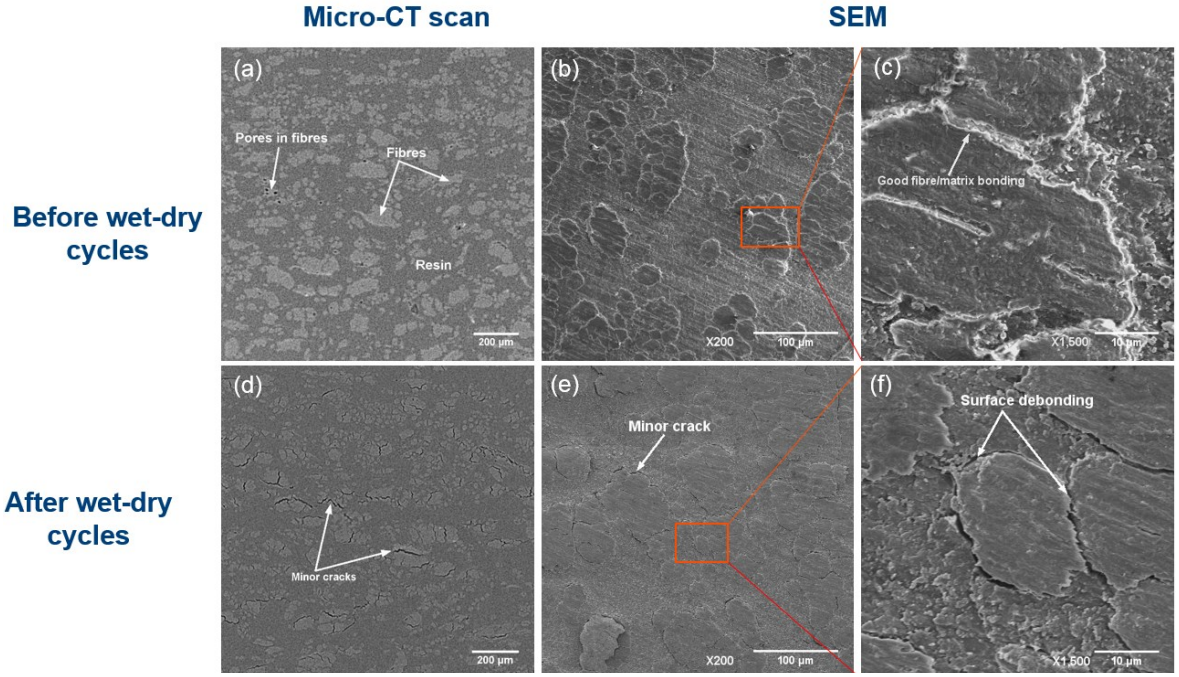


Fig. 7. Micro-CT (a,d) and SEM images of composites (DT samples) before and after the ageing study.

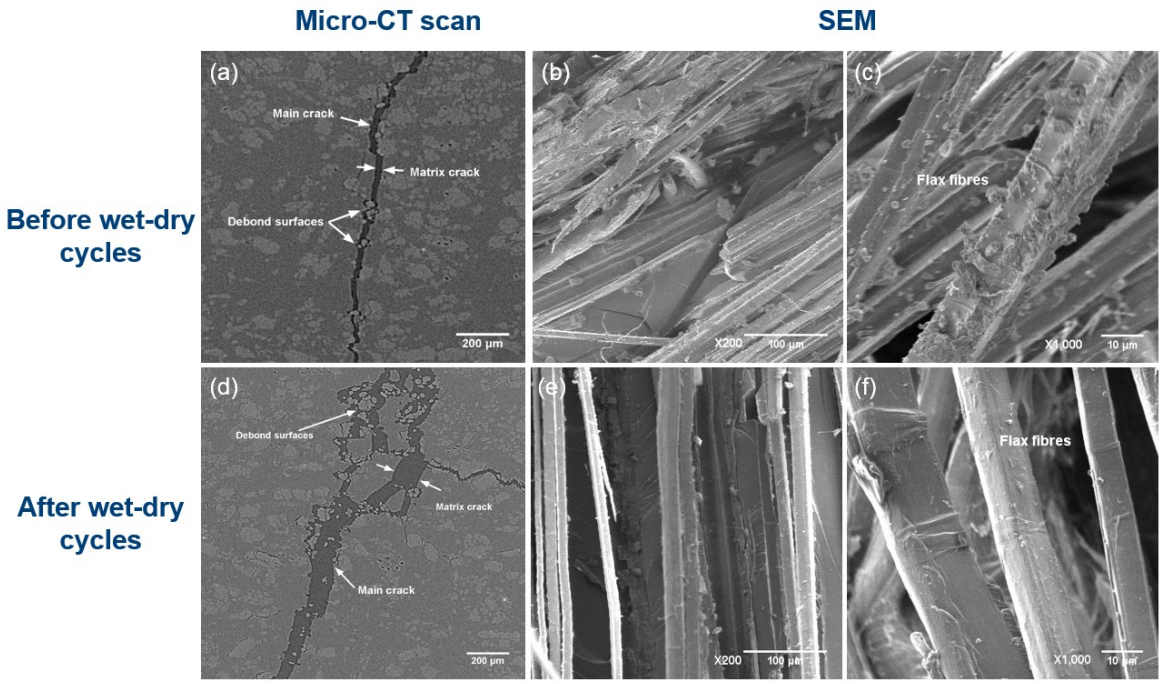


Fig. 8. Micro-CT (a,d) and SEM images of fractured part (transverse 3-point bending) of composites (DT samples) before and after the ageing study.

List of Tables

Table 1. Properties of composites made of dried and non-dry fibres in longitudinal and transverse directions after production and conditioning.

	Longitudinal		Transverse	
	Dried fibre	**Non-dry fibre**	**Dried fibre**	**Non-dry**
Production				
MC (%)	0.9 ± 0.3	3.6 ± 0.1	0.9 ± 0.3	3.7 ± 0.1
Thickness (mm)	2.034	2.028	2.061	2.091
Mean strength (MPa)	246.6 ± 15.2	221.8 ± 8.3	24.6 ± 2.4	23.1 ± 1.3
Mean modulus (GPa)	20.5 ± 0.8	16.7 ± 0.9	3.6 ± 0.1	3.2 ± 0.1
Conditioning				
MC (%)	2.2 ± 0.1	2.9 ± 0.3	2.4 ± 0.1	3.3 ± 0.2
Thickness (mm)	2.050	2.018	2.080	2.083
Mean strength (MPa)	240.2 ± 11.7	233.7 ± 27.0	24.8 ± 3.5	24.3 ± 2.3
Mean modulus (GPa)	16.4 ± 2.4	16.1 ± 3.1	3.5 ± 0.2	3.6 ± 0.4