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ABSTRACT

Moisture present in plant fibres is considered to be detrimental to the performance of composites. In general, a drying stage is performed on the plant fibre fabrics before manufacturing the composites since it is seemed to allow better output. This work provides an analysis of the effect of moisture in flax fibres on the overall quality of epoxy/flax biocomposites. Flax fibre fabrics were conditioned at different relative humidity (RH) environments and composites were manufactured by vacuum infusion technique. Composites were characterized by mechanical and microstructural analysis. Results showed that manufacturing composites with highly humid fabrics (95% RH) generates post processing deformation of finished parts and also leads to poor microstructural quality. The moisture in the fibres with different RH reduced the stiffness (from 23.74 to 17.67 GPa for Young's modulus and from 16.28 to 11.82 GPa for flexural modulus) but increased their fracture strain (from 1.87 to 2.64). Tensile strength displayed an optimum value (287.96 MPa) for fabrics conditioned at 50% RH, but flexural strength decreases continuously from 225.12 to 152.34 MPa as the moisture in the fabric increases.

摘要

植物纤维中存在的水分被认为对复合材料的性能有害。一般来说，在制造复合材料之前，在植物纤维织物上进行干燥阶段，因为它似乎允许更好的输出。这项工作提供了一个分析的亚麻纤维水分的影响，对整体质量的环氧/亚麻复合材料。在不同相对湿度环境下对亚麻纤维织物进行了整理，并采用真空浸渍技术制备了复合材料。通过力学和微观结构分析对复合材料进行了表征。结果表明，用高湿织物（95% RH）制造复合材料，加工后零件变形较大，显微组织质量差。不同RH纤维的湿度降低了刚度（从杨氏模量的23.74到17.67 GPa，从弯曲模量的16.28到11.82 GPa），但增加了断裂应变（从1.87到2.64）。拉伸强度显示在50% RH条件下织物的最佳值（287.96MPa），但弯曲强度不断下降，从225.12到152.34MPa，因为织物中的水分增加

Introduction

The growing concern of environmental issues has promoted the use of bio-materials replacing various synthetic materials over the past few decades (Pickering, Efendy, and Le 2016). In this context, biocomposites are being manufactured by using natural fibres, usually derived from plants, which are renewable, abundant in nature and cheaper than synthetic ones. Additionally, these fibres are less abrasive, reduce tool wear and the health risk to workers and operators. So, the initial low investment and friendly processing atmosphere of natural fibres reduce the dermal and respiratory

irritation are one of the main advantages (Cristaldi et al. 2010; Dhakal, Zhang, and Richardson 2007; Hajihah, Sain, and Mei 2014). However, the lignocellulosic plant fibres possess inherent polar and hydrophilic nature. The natural tendency of plant fibres to absorb moisture may cause swelling of the fibres and micro-cracks or voids can be created at the fibre-matrix interface. This may result in poor mechanical properties of the composites and reduced dimensional stability (Biagiotti, Puglia, and Kenny 2004; John and Anandjiwala 2008).

Vegetable fibres can be extracted from different parts of the plant such as: the bast fibres (jute, kenaf, hemp, ramie, flax), leaf fibres (sisal), fruit fibres (coconut), seed fibres (cotton), and among others. Flax, an important member of bast family is widely used in industries for composite fabrication. Flax fibre reinforced composites have the potential for a wide use in sports industries (bicycle frames, standup paddles, most kind of boards, etc.), automotive accessories (dash boards, door panels, etc.), maritime industry (small to medium sized boats). Nevertheless, flax prepregs with heavy areal weight (300 to 550 gm/m^2) are in development phase for future applications in the aeronautical industry (Suddell et al. 2005). George, Chae, and Bressler (2016) reported that flax based composites possess reduced weight and emit less carbon during their processing. They further indicated about the minimum dependence on foreign oil and less chance of volatility during manufacturing as their potential benefits. Each vegetable fibre (diameter between 50–100 μm) is composed of several elementary fibres (10–20 μm) glued together with lignin and hemicellulose (Nishino 2004). The cell wall of the elementary fibres has four different layers of cellulose micro fibrils embedded in a matrix of hemicellulose and lignin. These micro fibrils, which are mainly responsible for the mechanical properties of the fibre, follow a spiral angle along the elementary fibre that are composed of alternated crystalline and amorphous regions. The open channel in the centre of the elementary fibre is called the lumen. A small percentage of pectin, fat, wax and other water soluble substances are also found in the constituents (John and Anandjiwala 2008; Pandey et al. 2010). Plant fibres are highly hydrophilic, due to the presence of hydroxyl (OH) groups associated with the cell wall macromolecules: cellulose, hemicellulose and lignin. Cellulose units possess a high crystalline content (inaccessible to water molecules) but also a paracrystalline component to which water molecules can gain access. Lignin occurs in plant fibres in varying amounts, and is an amorphous crosslinked polymer composed of phenolic units and has a relatively low OH to carbon (C) ratio as compared with polysaccharides. The cell wall also contains a hemicellulose and pectic component, which is predominantly amorphous polysaccharide, is highly accessible to water molecules and has a high OH to C ratio (Hill, Norton, and Newman 2009). As the cell wall absorbs moisture, the absorbed water molecules occupy space between the micro-fibrils resulting in expansion of the material, i.e. fibres swell. In the case of flax fibres, they are composed of a very small percentage of lignin (2%), up to 20.6% of hemicellulose and more than 65% of cellulose (Pejic et al. 2008; Yan, Chouw, and Jayaraman 2014). The composition of flax fibre depends on time of flax harvesting and degumming method. Table 1 shows the equilibrium moisture content of some natural fibres at 65% relative humidity (RH) and 21°C (Rowell 2008). The moisture content at a given RH can have a significant effect on the performance of the fibres and their composites. Davies and Bruce (1998) studied the tensile properties of flax and nettle fibres and reported a consistent relationship between the modulus and RH when mechanical damage of fibres was considered. Baley, Morvan, and Grohens (2005) investigated the effect of absorbed water on the tensile strength of elementary flax fibres. They concluded that the fibre drying process affects the tensile strength and reduces the mechanical properties of the fibres. A different study (Joffe, Andersons, and Wallström 2003) of water absorbed elementary flax fibres revealed that the strain at failure is increased with moisture penetration at the fibre cell wall and it is the outcome of plasticizing effect of absorbed water of the flax fibres. Netravali and Chabba (2003) also mentioned that fibre strength can be decreased significantly with continuous moisture absorption and desorption cycles.

Table 1. Equilibrium moisture content at 65% relative humidity (RH) and 21°C (Rowell 2008).

Fibre	Sisal	Hemp	Jute	Flax	Ramie	Bagasse	Bamboo
Equilibrium moisture content (%)	11	9	12	7	9	8.8	8.9

Stamboulis, Baillie, and Peijs (2001) have reported moisture contents in flax fibres of 3%, 15% and 24% when exposed to 20, 66 and 93% RH environments, respectively, and the absorption curves showed typical Fickian behaviour. When the fibres were exposed to 100% RH, they found that the absorption mechanism changed, and Fick's law was not useful. Basically, they observed an absorption curve that increased in a stepped manner, reaching different equilibrium levels (showing a maximum of 42% moisture content). They attributed this behaviour to swelling stresses that develop during moisture absorption and relax after a certain time once the first equilibrium is reached, changing the equilibrium moisture condition. The authors also observed changes in the structure of the fibres as they were exposed to high humidity levels. The surface of dry fibres was smooth and the fibrils were well connected by the organic matrix forming a bundle, while the 100% RH conditioned fibres were swollen and their surface was rougher than that of the dry ones. In addition, the individual fibrils were more separated and some damages in form of kink bands were observed. These observations strongly suggest that properties of composites manufactured with natural fibre fabrics having different moisture contents could be varied significantly.

It has been widely reported that the exposure of natural fibre composites to wet and/or very humid environments cause degradation and is detrimental for their performance, although flax based biocomposites are still environmentally attractive (Le Duigou et al. 2012). However, little information can be found about the effect of moisture present in the fibres prior to the manufacturing stage on the composites' properties. A drying stage is usually performed on plant fibre fabrics conjecturing that it would allow obtaining better composites, since moisture in fibres is generally considered to be detrimental to composite performance. However, Baley et al. (2012) probed that drying the fibres (14 h at 105°C) results in significant loss of strength (44% on average) and failure strain (39%). They also performed tensile tests on unidirectional composites reinforced with dried and "as-received flax fibres" and found that drying did not affect axial stiffness but caused a large drop (36%) in composite strength, in a similar way to the drop found in fibre strength. Besides, they stated that the removal of moisture would also influence the properties of the fibre surfaces and hence the interfacial bonding. Therefore, they suggested that a more detailed study is necessary to optimize the conditioning of plant fibre fabrics in order to obtain the highest quality composites. Recently, Fuentes et al. (2016) studied the influence of moisture content in the environment during manufacture by vacuum infusion of flax fibre-unsaturated polyester (UP) composites on their performance. They compared the performance of composites which were manufactured under dry and 100% RH conditions, and found a reduction in tensile and flexural strength and stiffness, which was related to the decrease of the fibre mechanical properties due to softening and possible dissolution of the interface between elementary fibres. Therefore, this study was carried out to enrich the little existing knowledge regarding the conditioning of plant fibre fabrics prior to the manufacture of thermoset matrix composites. Since the matrix chemistry could be important in the moisture-fibre-matrix interaction, an epoxy resin was chosen to compliment and contrast the results available for an UP matrix, reported by Fuentes et al. (2016). The effect of moisture present in the fabrics on the quality of vacuum infused flax/epoxy composites was studied in terms of overall composite quality, mechanical properties and microstructural observations.

Experimental procedure

Materials

Flax composites have been manufactured by using prepreg flax fibres and epoxy resin. Commercially available FlaxPLY UD 180 (fabric pretreated with 16% epoxy based resin) supplied by Lineo company (France). The areal weight of this flax fabric is 180 gm/m^2. The low density (1.4 gm/cm^3) fabric has a fibre fraction of about 65% by weight before curing. The H180 standard hardener was properly mixed with epoxy resin (R-180 Part A) before infusion. Both were procured from Nuplex industries (Aust) Pty Ltd. TR 102 regular wax (TR industries, USA) was used to release the mould.

Drying of fabrics and moisture absorption tests

The fabrics were stored in our laboratory (65% RH, 23°C) and were cut into five rectangular samples (230 mm × 270 mm), weighted using an analytical scale (precision ± 0.0001 gm) and placed inside a dehumidifier oven (model: TD300F, Thermoline Scientific) at 60°C. The weight loss was monitored every 20 min until equilibrium was reached.

The study on the moisture absorption kinetics was done in a humidity chamber (model: CH700 V, Angelantoni) set to different relative humidity values (10, 35, 50, 70 and 95% RH). The humidity chamber can control the RH within the error limit +/- 1%. Fabrics were first dried and weighted. Weight gain was monitored by means of an analytical scale until equilibrium was reached.

The relative specimen weight was calculated with Eq. (1), where $M_r(t)$ is the relative weight of the specimen at any time, $W(t)$ is the specimen weight at each time, and W_0 is the initial specimen weight.

$$M_r(t) = \frac{W(t)}{W_0} \tag{1}$$

Composites manufacturing

Composites were manufactured by the vacuum infusion technique using ten layers of reinforcement, which were conditioned at 25°C and different relative humidity levels in the humidity chamber (10, 35, 50, 70, 95% RH). To ensure that the composites were manufactured with the correct moisture content in the fabrics, the following procedure was carefully carried out:

The layers of fabric were first cut to the dimensions needed for the composite panels (230 mm × 270 mm) and dried. Then, they were placed inside the humidity chamber at the desired value of RH distributed individually on the shelves (not stacked) for 24 h. In the meantime, an aluminium flat mould was waxed and all the infusion materials (bag, peel ply, flow media, hoses) were prepared and cut to the correct dimensions. Fabrics were withdrawn from the humidity chamber and they were quickly placed on the aluminium mould. The peel ply and the flow media were laid over the fabrics and the vacuum bag was sealed to the mould but only in two sides, where the hoses (resin and vacuum ports) were also attached. In this way, the other two longer sides of the bag remained widely open. Although this procedure takes not more than 15 minutes, the fabrics can lose or gain moisture, depending on the moisture gradient between the fabrics and the environment. Therefore, the mould was placed inside the humidity chamber for another 24 h, giving enough time to the fabrics to reach the corresponding equilibrium value for the set RH. Finally, the mould was removed from the humidity chamber and the remaining two open sides were sealed instantaneously (less than one minute), ensuring that the fabrics were kept at or close to the desired moisture content, and the resin infusion was carried out immediately following the bagging process. The flow media allowed completing the impregnation in less than 10 minutes, therefore the extraction of moisture from the preform by the vacuum system was kept to the (practically achievable) minimum. In addition, once the impregnation was finished the resin inlet and vacuum port were immediately closed.

In addition, composites were manufactured using dry fabrics. In this case, fabrics were dried in the oven for 4 h at 60°C, then placed on the mould, the infusion set up was prepared and the vacuum bag was completely sealed to the mould. Vacuum was applied and the stack of fabrics was dried in the oven for another 3 h under full vacuum at 45°C. Since the preform was under vacuum and there was no risk of moisture absorption, the resin was infused 15 minutes after the mould was withdrawn from the oven, allowing the system to cool down and reach at the laboratory temperature.

All composites were cured under full vacuum (−100 kPa) at room temperature for 24 h and then post cured for 8 h at 60°C. The tests samples were cut from the panels by means of a laser cutter device to the required shape and dimensions given by the standards used for the mechanical characterization. Table 2 represents the nomenclature used throughout this work to refer each composite material.

Table 2. Nomenclature used in this work.

Nomenclature	C-D	C-10	C-35	C-50	C-70	C-95
% RH used in fabric conditioning	0 (dry fabrics)	10	35	50	70	95

Microstructural characterization

Composites microstructure was analysed by optical microscopy. Transverse sections of the laser cut composites were polished with a 2500 grain size sand paper before the microstructural observations. In addition, the fibre-matrix interface was analysed by scanning electron microscopy (SEM), model: JEOL FE-SEM JSM7100F. The specimens were prepared by polishing the surface of the composites and finally by coating the specimens with a thin layer of platinum.

Mechanical properties evaluation

An Instron universal testing machine was used for the mechanical characterization. Three-point bending tests were performed using a span of 60 mm and a cross head displacement speed of 2.4 mm/min, according to the ASTM D 790 (2007) (sample size: $127 \times 12.7 \times 3.2$ mm). Load-displacement curves were obtained from these tests and flexural modulus and strength values were determined. Tensile properties (Tensile strength, modulus and strain at failure) were measured according to the ASTM D 3039 (2008) (sample size: $250 \times 15 \times 3.2$ mm), using a testing speed of 2 mm/min. A static axial clip-on extensometer (INSTRON) was used to measure the elongation of the samples.
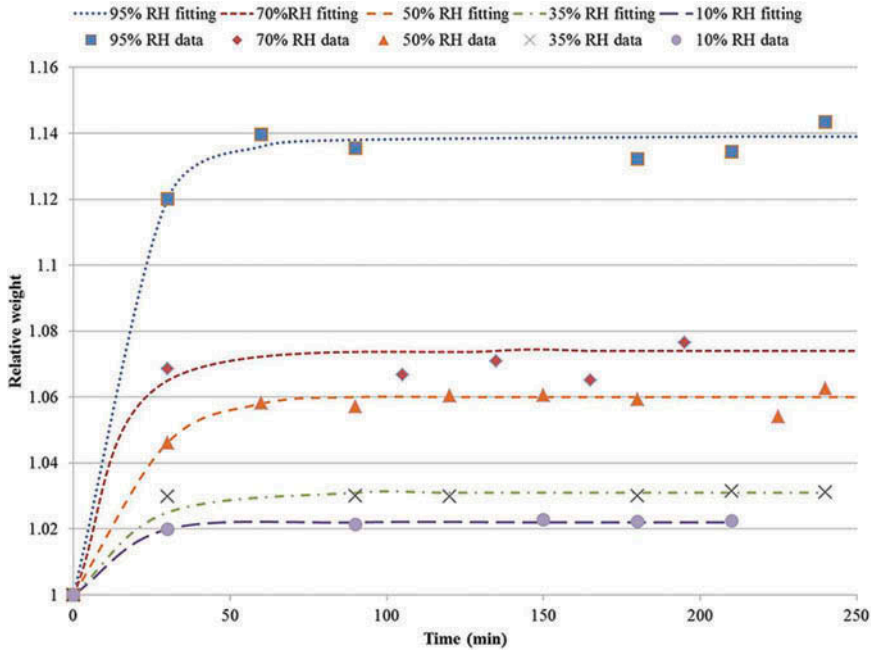
Results and discussion

Moisture absorption

The equilibrium moisture content of flax fibre fabrics exposed to different relative humidity levels is shown in Figure 1. As expected, the moisture content increased with increasing relative humidity
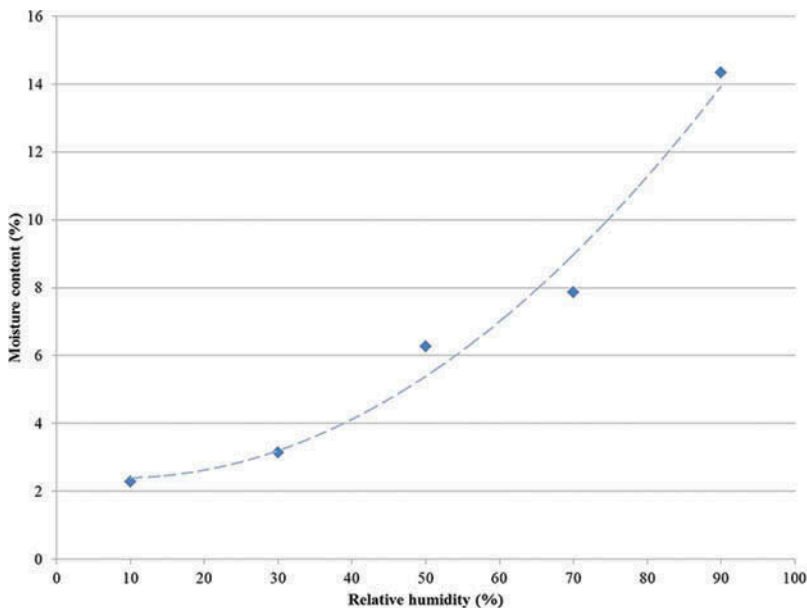
Figure 1. Equilibrium moisture content in the fabrics exposed at different RH.

**Figure 2.** Moisture absorption kinetics for all conditions.

reaching a maximum value of 14.3% at 95% RH. These results are in good agreement with those found by Stamboulis, Baillie, and Peijs (2001) for duralin flax fibres (treated fibres). The equilibrium moisture contents in the fabrics for different relative humidity levels were increased almost linearly with the relative humidity from 10% RH to 70% RH. A sharp increase can be observed between that value and 95% RH where the moisture content was almost double that found at 70% RH. This trend is very similar to that found by other authors on different cellulose fibres, such as cotton (Peirce 1929), bamboo (Chen, Miao, and Ding 2009) and flax (Stamboulis, Baillie, and Peijs 2001). When cellulose fibres are exposed to moisture, at the first stage water molecules are absorbed directly onto the hydrophilic groups of the fibre material. After that, more water molecules are attracted either to other hydrophilic groups or they may form further layers on top of the water molecules already absorbed (Morton and Hearle 2008; Peirce 1929). Therefore, at high humidity levels, liquid water may be held together by the forces of surface tension in capillary spaces available in the material (Chen, Miao, and Ding 2009).

It should be noted that all fabrics reached the equilibrium moisture content in less than 60 minutes (Figure 2), showing that the absorption kinetics is quite fast if compared to the normal processing times required for handling, pre-forming and preparing the infusion. Similar findings were observed in a study about flax and hemp fibres. The absorption-desorption equilibrium for flax fibres at 21°C and 40% RH was 40 minutes and 40–45 minutes for hemp fibres with same conditions (Mustata and Mustata 2013). Nair et al. (2011) also reported that, flax fibres reached the equilibrium after 28.5, 34 and 37 minutes when they were drying at 80°C, 60°C and 40°C respectively.

Visual observations of the manufactured composite panels

Panels manufactured with fabrics conditioned at 95% RH have been shown severe warpage after demoulding, as shown in Figure 3a, and this effect was also observed, in a less extent, on the composite made with fabrics conditioned at 70% RH (Figure 3b). On the other hand, composites made with fabrics conditioned at 50% RH and lower did not suffer any deformation (Figure 3c).
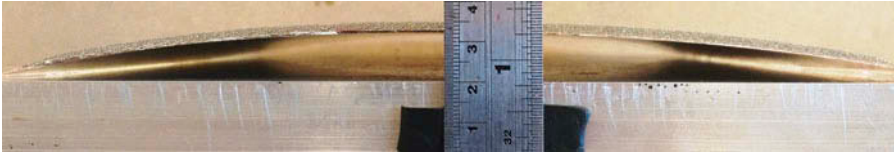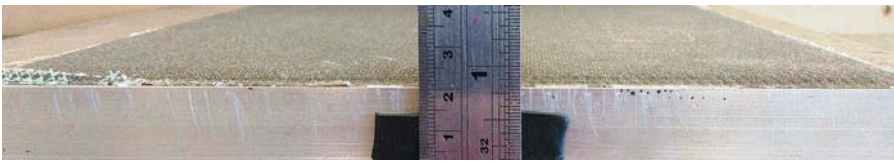
Figure 3a. Severe warpage observed. Fabrics conditioned at 95% RH.

Figure 3b. Moderate warpage observed. Fabrics conditioned at 70% RH.

Figure 3c. No warpage observed. Fabrics conditioned at 50% RH.

Warpage of composite laminates has been observed by many authors (Albert and Fernlund 2002; Ersoy et al. 2005; Fernlund et al. 2002; Potter et al. 2005; Twigg, Poursartip, and Fernlund 2004; Zeng and Raghavan 2010). When the autoclave processing technique is used, the main reported causes for this issue are the residual stresses that develop due to mismatched coefficients of thermal expansion (part-mould) and chemical shrinkage of the resin. These stresses can also develop due to gradients in temperature, fibre volume fraction, and uneven curing throughout the part (Kaushik and Raghavan 2010). This non-uniform stress distribution is locked in as the resin cures and upon removal from the tooling, the resultant bending moment warps the part away from the tooling. When liquid resin infusion processes are used, such as those used in this work where Perrin, D'Acunto, and Martin (2010) suggested that the resin distribution media can be another important source of warpage, explaining that the cure shrinkages are too different in the part (high fibre to resin ratio) than in the flow media (high resin to nylon mesh ratio).

In the present work, the composites were manufactured and cured at room temperature, so temperature was uniform throughout the part (no temperature or resin conversion gradients existed) and no significant thermal expansions/contractions could have occurred in the mould and the part. In addition, no chemical treatment was carried out to reduce the hydrophilic nature of the fibres. Therefore, the only significant interaction between the part and the mould must have been caused by part shrinkage. It is important to note that all composites were manufactured using the same infusion materials (same peel ply, flow media, and vacuum bag) and that the samples made with dry fabrics and those conditioned at 35% RH and 50% RH did not show any warpage. These facts strongly suggest that the high moisture content in the fabrics conditioned at 70 and 95% RH was the main reason for the post processing deformation of the panels.

A possible cause for part shrinkage and residual stress development is the swelling and shrinkage of flax fibres exposed to high % of RH. It is well known that plant fibres swell and shrink as they absorb and desorb moisture, respectively. In addition, moisture-induced swelling is considerably larger in the radial or transverse direction compared with that of the axial direction (Gamstedt 2016),

which is consistent with the warpage direction observed in the composites. The fibrous preform was in a swollen state when compacted by the vacuum bag, and part of the moisture could have been gradually extracted during the infusion, drawn away by the vacuum and dissolved in the resin, causing fibre shrinkage and therefore, compressive stresses were developed in the part. This suggestion is consistent with the optical microscopies where significant number of voids were found when the composites were manufactured with high moisture content fabrics. Water molecules dissolved in the resin could have form water vapour as the pressure decreased due to the applied vacuum and the temperature of the system increased due to the exothermic reaction, forming the voids that remained in the cured composite.

Summarizing, it seems to be a moisture content over which residual stresses caused by part shrinkage are significant enough to originate deformation on the manufactured part. In this work, that limit was found to be at some point between 7% (C-70) and 14% (C-95).

During the post curing stage, composite panels were flattened by compressing them between two heavy steel plates. In this way, the post curing cycle was also used to relieve the residual stresses in the parts trying to restore their original shape. Figures 4a and 4b show pictures of the two samples that suffered significant warpage after the post curing stage. It can be seen that C-70 composite flattened almost completely, while the composite made with the most humid fabrics (C-95) still presented significant warpage, although it was much less severe than that observed immediately after demoulding.

Microstructural analysis

Moisture present in the fabrics led to the formation of voids in the composites microstructure. Figures 5a, b and c shows optical micrographs of C-D, C-50 and C-95 composites. It can be seen that the composite produced with dry fabrics presented almost a void-free microstructure, the one manufactured with the 50% RH conditioned fabrics had some small voids, and the sample produced with the most humid fabrics exhibited at least 40–50 interconnected voids as seen by the optical microscope. As suggested in the previous section, water molecules attached to the fibres could have been dissolved by the resin and formed a water gaseous phase as the pressure decreased due to the applied vacuum, and the temperature of the system increased due to the exothermic reaction, forming the voids found in the cured composite. It is also interesting to note that the number of voids was higher close to the bag side of the panel. This is consistent with the visual observation of the manufactured composites, which presented a smooth and quasi-perfect surface on the mould side, but a more porous surface at the bag side (only for composites made with 95% RH fabrics).

Figure 4a. Warpage almost disappeared after the post curing stage. Fabrics conditioned at 70% RH.

Figure 4b. Warpage decreased but still significant. Fabrics conditioned at 95% RH.

Figure 5a. Optical micrograph of C-D showing an almost void-free microstructure.


Figure 5b. Optical micrograph of C-50 showing a few amount of small pores.


Figure 5c. Optical micrograph of C-90 showing a significant amount of large interconnected voids.

These observations suggest that low-density gaseous water bubbles had enough time to migrate to the upper surface, before gelation point was reached (3–4 h).

SEM images provided more details of the composites microstructure (Figure 6a, 6b, 6c). It can be seen that composites manufactured with dry fabrics showed a healthy microstructure and good cohesion between fibres and matrix (Figure 6a). The black dots which can be seen in the picture are not pores but the tip of some elementary fibres overcharged by the electron beam. When 50% RH conditioned fabrics were used, the interface between fibres and the matrix looked similar to that of the dry composite, but some voids were visible in the matrix region (Figure 6b). As expected, the poorest microstructural quality was shown by the composites made with 95% RH conditioned fabrics, where matrix cracking was detected and poor interfacial adhesion between the flax yarns and the epoxy matrix was observed (Figure 6c). Matrix cracks could have been created during panel warpage due to the intense residual stresses developed during the curing stage, as explained in the previous section. Chen, Miao, and Ding (2009) also found a poor fibre-matrix adhesion in their
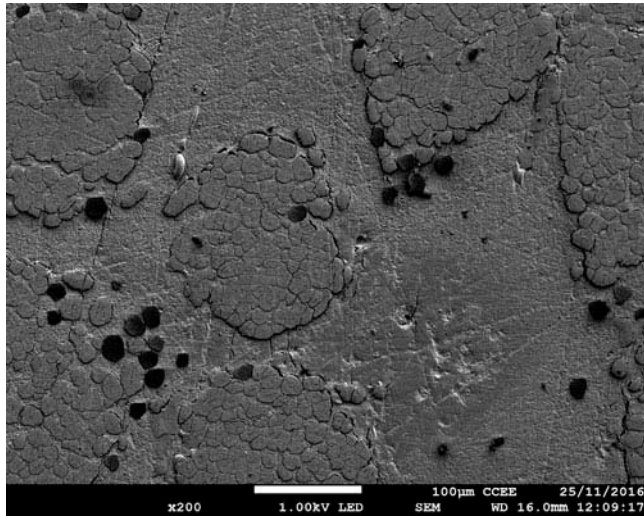
Figure 6a. SEM image of the surface of C-D composite.
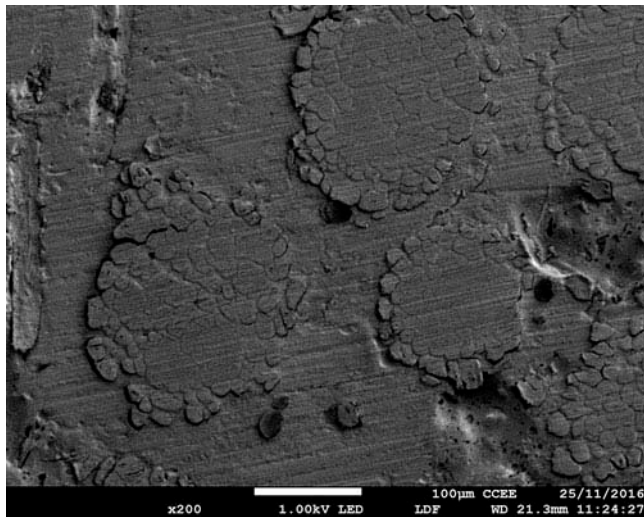
Figure 6b. SEM image of the surface of C-50 composite.

bamboo/vinyl ester composites when high moisture content fibres were used, and suggested that water became a physical barrier between the fibres and the matrix. It should be noted that the macroscopic pores shown in Figure 5c cannot be seen in SEM images due to the high magnification.

Mechanical properties

Figure 7 shows typical stress-strain curves for all the composites. It can be seen that after a first linear region, the slope of the curve decreased substantially and the relation between the stress and strain remained fairly linear until a sudden failure occurred. This shows that after a certain point, the materials lose some rigidity. This behaviour can be attributed to the complex microstructure of plant fibres. During a tensile test carried out in the direction of the fibre axis, the cellulose fibrils orientate
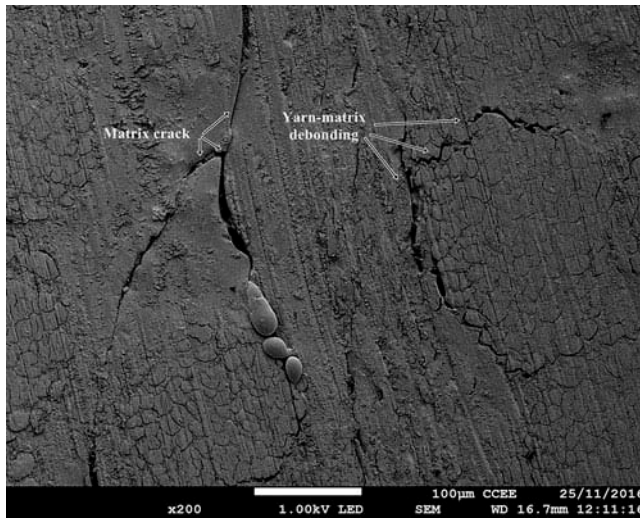
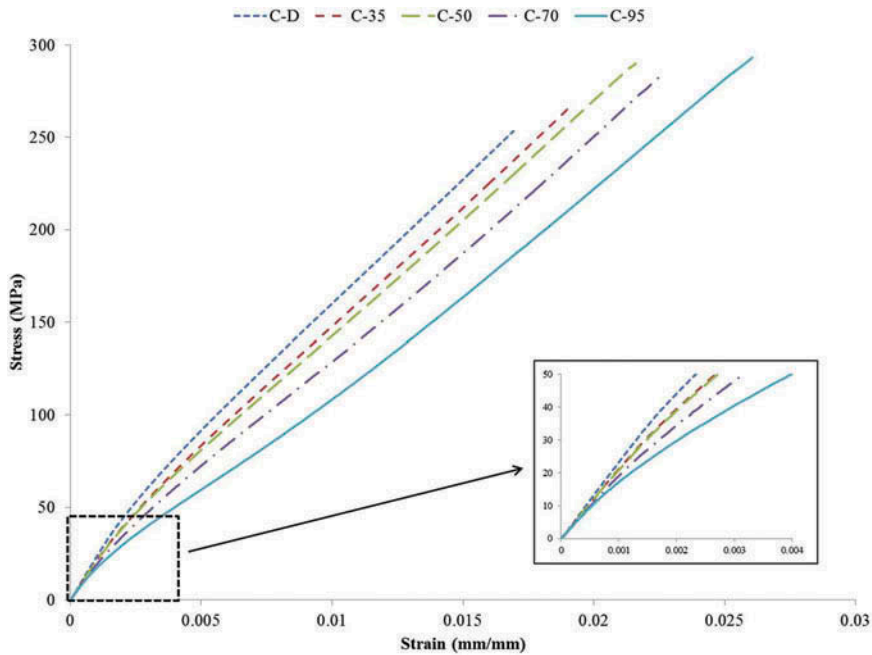**Figure 6c.** SEM image of the surface of C-95 composite.

Figure 7. Stress-strain behaviour of the composites under tensile loading.

towards the direction of load, which can cause an increase in stiffness (although not observed in the composites) and then they start sliding one with respect to the others (Bossuyt 1941), and the primary cell walls collapse leading to delamination between fibre cells (Mukherjee and Satyanarayana 1984). These phenomena are believed to be responsible for the decrease in the rigidity of the materials during tensile loading. It is interesting to note that, as the moisture content of the fabrics increased, the inflection point at which the rigidity change was occurred at a lower stress value due to the plasticizing effect of water that enhanced sliding and delamination of cellulose fibrils. As it was

explained by Stamboulis, Baillie, and Peijs (2001), water molecules penetrate the cellulose network and they may attach by chemical links to groups in the cellulose molecules forcing them apart and allowing them to move more freely, destroying some of the rigidity of the cellulose structure.

Tensile properties of the composites are summarized in Figure 8. Young's modulus decreased continuously as the moisture content in the fibres increased. In addition, the tensile strain at break found for the composites was, in order of increasing moisture content: 1.87% (Dry), 1.93%, 2.09%, 2.23% and 2.64% (95% RH). The softening effect of water on these kinds of fibres produce an increase in the breaking strain and a decrease in the tensile modulus of the composites as it was found by Stamboulis, Baillie, and Peijs (2001) in tensile tests of single flax fibres. Furthermore, moisture can also plasticize the polymer matrix and interact with it chemically (hydrolysis) (Tsenoglou, Pavlidou, and Papaspyrides 2006; Won et al. 2008), usually decreasing its mechanical performance. Although the difference in tensile strength between the best and worst material was small (6%), an optimum tensile strength was found for the composite manufactured with fabrics conditioned to 50% RH.

The overall performance of the composites depends on the properties of the fibre, the matrix, the fibre-matrix interface, and the composite microstructure given by the fibre volume fraction and void content. To understand the effect of moisture content in the fibres on the composites overall mechanical behaviour, it is necessary to identify the influence of water molecules on each individual component. In the first place, moisture increases the tensile strength of plant fibres, as it was reported by many authors (Chen, Miao, and Ding 2009; Mukherjee and Satyanarayana 1984; Stamboulis, Baillie, and Peijs 2001), which was mainly attributed to a plasticizing effect advantageous to the strength of cellulose fibres generated by the availability of free water molecules. For example, Stamboulis, Baillie, and Peijs (2001) reported that after being humidified at 90% RH, green flax fibres increased their tensile strength by about 20%. Chen, Miao, and Ding (2009) and Chung and Yu (2002) also found a small increase of the tensile strength as the moisture content increased, on bamboo strips and moso bamboo, respectively.
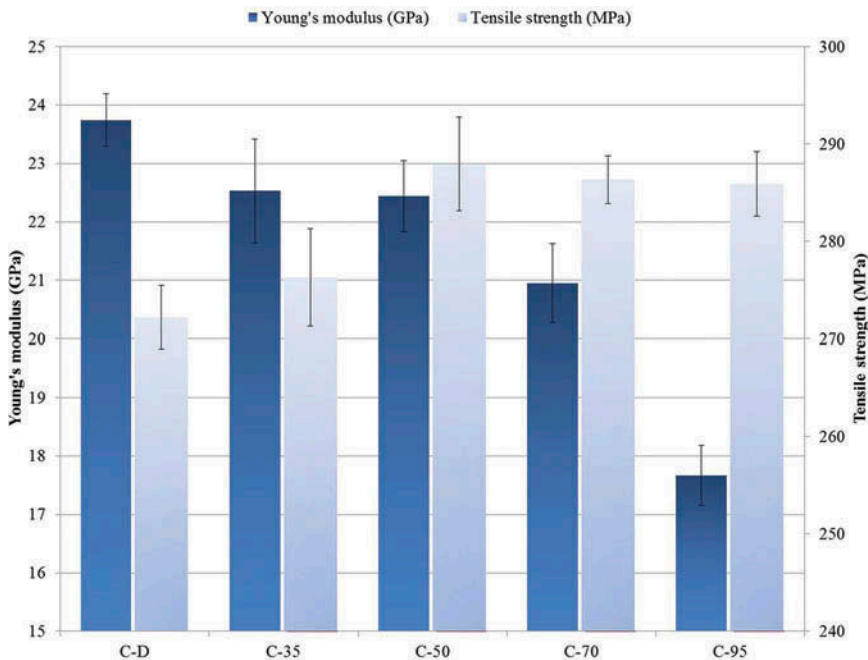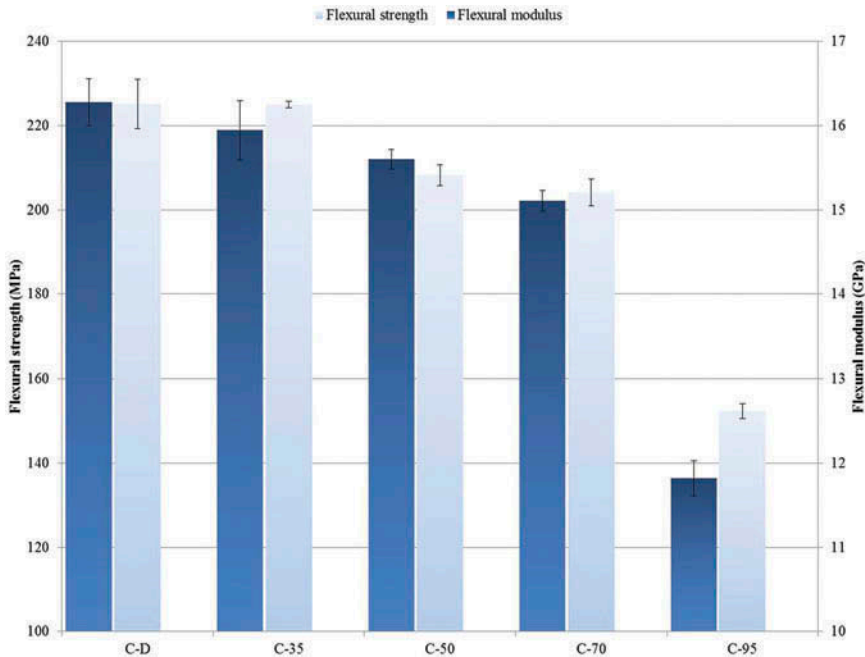

Figure 8. Tensile properties of the composites.

Figure 9. Flexural properties of the composites.

Finally, moisture present in the fabrics led to the formation of voids in the final composite, especially in composites made with fabrics conditioned to more than 50% RH as it was shown in the optical microscopes (Figure 5a, 5b, 5c). The detrimental effect of voids on the mechanical performance of composites was widely probed and can be found in literatures (Costa, De Almeida, and Rezende 2001; Li, Li, and Ma 2015; Varna et al. 1995). In addition, some cracks were found in the epoxy matrix in the C-95 composites, which can also weaken the material.

Summarizing, moisture increases flax fibre tensile strength but it reduces the fibre-matrix adhesion, plasticizes the matrix and is detrimental for the composite microstructural quality as it enhances void formation and matrix cracking. Therefore an optimum moisture content in the fibres exists in which the balance between all these phenomena give the maximum tensile strength for the composite.

Flexural properties of the composites are presented in Figure 9. Flexural strength of the composites slightly decreased with the moisture content in the fabrics from "dry" to 70% RH while the composite manufactured with the most humid fabrics showed a significant loss in flexural strength. Apparently, the gain in tensile strength of the flax fibres due to their increased moisture content has a less significant effect on the overall flexural properties of the composites, and it is surpassed by the negative effects of moisture on the fibre-matrix interface and microstructural quality. In addition, in this type of loading configuration just half of the fibres are loaded under tension, while the other half suffer under compressive loading. In accordance with the results obtained for the Young's modulus, the flexural modulus decreased as the moisture in the fabrics increased, and a steep drop in this property was observed for the C-95 composite. The steep drop of properties found can be correlated to the steep moisture uptake observed in the fabrics conditioned at 95% RH where they showed a moisture gain almost double that shown by the fabrics conditioned at 70% RH.

Conclusions

This work presented a study on the effect of moisture content in flax fibre fabrics prior to manufacturing epoxy reinforced composites, on the overall quality of the composites. Dimensional

stability of flat panels was greatly affected, since severe warpage was observed for parts made with fabrics taken from 70% RH and 95% RH environments. The microstructure of the composites was also affected by the high moisture content in the fabrics and thus the fibre-matrix interface became weaker and porosity was increased. Water molecules present in the fibres caused positive and negative effects on the tensile properties of the composites. The plasticizing effect of water molecules on flax fibre microstructure and deformation mechanisms significantly increased the strain at break and decreased Young's modulus. In addition, an optimum strength was found for the composites made with fabrics conditioned at 50% RH, given that water molecules increased the tensile strength of the fibres but at the same time they had a detrimental effect on the fibre-matrix interface and overall microstructural condition. In terms of flexural properties, a continuous decreasing trend was observed in both strength and modulus which suggests that, for this loading configuration, the positive effect of water in fibre tensile strength could not compensate for the negative effects resulting from microstructural changes in the composites. A steep drop in properties (Young's modulus, flexural strength and modulus) was found when the composites were manufactured with fabrics conditioned at 95% RH. This was consistent with the moisture absorption data, which showed that moisture uptake of the fabrics was almost linear from 10% RH to 70% RH, but beyond the 70% threshold (at 95% RH), the moisture uptake almost doubled.

Acknowledgments

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