BACTERIAL CELLULOSE AS REINFORCEMENT FOR NATURAL FIBRE THERMOSET COMPOSITES

Marta Fortea-Verdejo[1], Koon-Yang Lee[2], Alexander Bismarck[1,3]

[1] Institute for Materials Chemistry & Research, Faculty of Chemistry, Universität Wien, Währinger Straße 42, 1090 Wien, Austria
Email: marta.forteaverdejo@univie.ac.at, alexander.bismarck@univie.ac.at Web Page: http://mc.univie.ac.at/about-pace/
[2] The Composite Centre, Department of Aeronautics, Imperial College London, South Kensington Campus, SW7 2AZ, London, UK.
Email: koonyang.lee@imperial.ac.uk, Web Page: http://www.imperial.ac.uk/materials-and-sustainability
[3] Department of Chemical Engineering, Imperial College London, South Kensington Campus,SW7 2AZ, London, UK
Email: alexander.bismarck@imperial.ac.uk, Web Page:
http://www3.imperial.ac.uk/polymersandcompositesengineering

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Abstract

It has been previously shown that the introduction of nanocellulose as binder for natural fibre thermoplastic and thermoset composites can enhance the mechanical properties of natural fibre composites. We aim to investigate the effect that bacterial cellulose (BC) has as binder, when introduced in different thermoset-flax fibre composites. Two matrices were investigated, epoxy and phenolic resins. To introduce the bacterial cellulose into the composites, a preform using BC as binder for loose flax fibres was manufactured. The preform was then infused with the thermosetting resins and cured. The resins penetrated the preforms during infusion, however, no significant improvement of the tensile properties could be seen when introducing BC for neither of the resins.

1. Introduction

Natural fibre composites have found a wide range of application on many areas. Nowadays, natural fibre composites are being used in the automotive industry, for inside and outside parts of cars [1, 2]; in the packaging and building industry [2-4] and many others [4, 5]. The interest in these composites, comes from the fact that natural fibres are a cheap renewable resource, with low density and high specific stiffness and strength, to enhance the properties of polymeric matrices [6].

Due to this interest, the optimization of natural fibre composites properties is a relevant topic. Apart from the fibre and matrix selection (to achieve the desired performance), the interfacial adhesion between them is important to increase the stress transfer between the fibres and matrix. To achieve an increase of the interfacial strength in natural fibre composites, chemical modification of the fibres/matrix or the use of adhesion promoters has been extensively studied [7, 8]. However, the use of chemicals and their disposal is a problem that has to be taken into account. Another way of improving natural fibre composites is by improving the fibre orientation in the matrix, for example using natural fibre mats or fabrics. Fabrics can be made by weaving natural fibres (in the form of yarns) or by mechanical bonding, such as entanglement/interlocking of the fibres (to produce nonwovens) [9], where the loose fibres are hold together by mechanical means. However, these manufacturing procedures require specific

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equipment. There is another way of holding loose natural fibres to give mechanical stability without machinery involved and enhancing the fibre-fibre stress transfer: producing nonwovens/preforms using nanocellulose as binder [10].

Nanocellulose possesses a high surface area, which leads to high concentration of hydroxyl groups on its surface. Upon the removal of water, nanocellulose forms a network-like structure through hydrogen bonding. This process is known as hornification [11]. Nanocellulose can be obtained from natural fibres/wood following a top-down approach or it can be synthesized by some bacteria, following a bottom-up approach [12, 13].

Bacterial cellulose (BC), as well as nanocellulose from other sources have already been used as reinforcement for thermoplastic and thermoset matrices, as well as for composites [14-16]. Lee et. al reported the use of BC as additional binder for sisal/polyAESO composites [17]. The addition of just 10 wt. % BC doubled the tensile and flexural properties of the composites. To do so, they manufactured a preform from sisal and BC following a paper-making process, which was later infused.

In this work, we explore the use of BC as binder for flax fibres and two different thermoset polymers: an epoxy resin and a phenolic resin. The flax/BC nonwoven preforms were produced following a paper-making process similar to Lee et al. and the resin was introduced in the preforms by vacuum infusion.

2. Experimental

2.1. Materials

Flax fibres with an approximate length of 25mm were kindly supplied by Novalin France (Millam, France). Epoxy resin Araldite ® LY556 and hardener XB 3473 were purchased from Huntsman Advanced Materials Ltd. (Cambridge, UK). Phenolic resin Cellobond J2027L and catalyst Phencat 382 were both purchased from Caleb Technical Products (Bristol, UK). Bacterial Cellulose was extracted from nata de coco (CHAOKOH coconut gel in syrup, Ampol Food Processing Ltd., Nakorn Pathom, Thailand) as previously reported [18].

2.2. Manufacturing of the nonwoven preforms and composites

The manufacturing of the composites consisted of two main steps, the preform consolidation and the infusion of the preform with the thermosetting polymer. The preforms were manufactured following a layer-by-layer filtration that can be found in literature [10]. The grammage of the preforms was set to 800 g m^{-2}. The composites were manufactured using a double bag vacuum assisted resin infusion [19], followed by a thermal curing cycle. The mixing ratio, the infusion and the curing conditions for each thermosetting polymer can be found in table 1.

Table 1. Mixing ration, infusion and curing conditions for epoxy and phenolic resins.

Matrix	Mixing ratio (wt.%)	Degas Conditions	Temperature during Infusion (°C)	Curing Cycle
Cellbond/Phencat	100/4	Room Temperature 30 min	-	1h at 80 °C 4h at 80 °C
Araldite/Hardener	100/23	80 °C 30 min	80	2h at 120 °C 4h 180 °C

2.3. Characterisation of the nonwoven preforms and composites

The tensile properties of the nonwoven preforms and natural fibre composites were investigated using an Instron universal testing machine (Instron 5969, Instron GmbH, Germany) equipped with a 50 kN load cell (natural fibre composites) or a 1kN load cell (nonwoven preforms). The testing speed was 1 mm min^{-1} and the distance between the clamps was set to 60 mm. Additionally, the strain of the samples

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was measured with a video extensometer (iMetrum Ltd, UK). The testing specimens possessed dimensions of 100 × 15 ×1.5 mm^3 and a minimum of 5 samples were measured for every set of samples. The morphology of the natural fibre composites was studied by Scanning Electron Microscopy (JEOL JCM-6000) at an accelerating voltage of 15kV. Relevant parts of the composites were ripped apart and fixed to the sample holder with carbon tabs. The samples were then coated at 30 mA for 40 s on a sputter coater (JFC-1200, JEOL GmbH, Eching, Germany).

3. Results

3.1 Tensile properties of the nonwoven preforms and composites

Prior to the composite manufacturing, flax/BC nonwoven preforms were produced. The mechanical properties of the nonwovens can be found in literature [10]. The nonwovens containing 10 wt.% BC possessed a tensile strength of 12.7 ± 1.1 MPa. Due to the high porosity of the nonwovens (~60 %), no constant area could be assumed, thus no tensile modulus was reported and the tensile strength in MPa was only to ease comparison, since it was originally reported in $kN{\cdot}m^{-1}$. The effect of the introduction of bacterial cellulose (10 wt.% BC in the nonwoven preforms) in the natural fibre composites can be seen in table 2.

Table 2. Tensile properties of the thermoset/flax composites with and without BC as additional binder.

Matrix	BC (nonwoven wt.%)	Nonwoven (composite wt.%)	Tensile Properties	
			σ (MPa)	E (GPa)
Phenolic	0	50	31.0 ± 3.2	8.3 ± 2.4
Phenolic	10	47	35.1 ± 2.7	9.4 ± 0.6
Epoxy	0	50	44.4 ± 4.1	10.3 ± 1.3
Epoxy	10	47	39.4 ± 2.8	9.8 ± 1.2

When using phenolic resin as matrix, the addition of 10 wt.% BC in the nonwoven preforms produced a slight increase of the tensile strength and modulus of the composite. However, when using epoxy resin as matrix, the addition of BC did not enhance the tensile properties of the composites, but slightly reduced them. It has been previously reported that the introduction of nanocellulose into sisal/polyAESO composites following this manufacturing method, doubled the tensile properties of the composites [17]. However, this increase was not seen for flax/epoxy or flax/phenolic resin, probably due to a poor penetration of the matrix through the nanocellulose network, which consolidates the nonwoven preform, impairing impregnation. Since we created a nanostructure, we may introduce in our system nanoporosity, so the space between the nanofibres could be too low for the resin to penetrate [20].

3.1 Morphological characterization of the composites

Figure 1 shows micrographs of the phenolic/flax composites. The composites were ripped apart prior to taking the images, to ensure that the phenolic resin penetrated the nonwoven preforms during the infusion process and not only on its surface.

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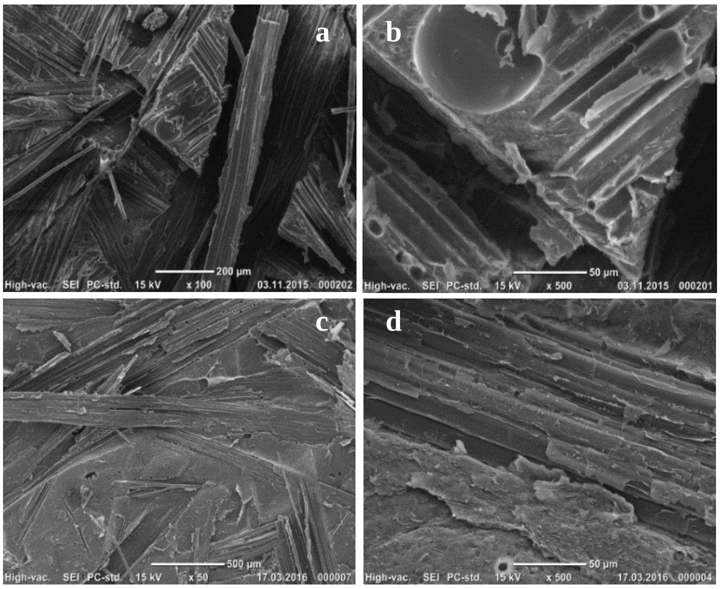

Figure 1. Micrographs of phenolic/flax composites containing 0 wt.%BC (a,b) and 10 wt.% BC (c,d) nonwoven preforms at different magnifications.

In both composites (0 and 10 wt.% BC) the resin penetrated into the preform. It can also be seen in both composites circular shaped voids, probably created by water evaporation, present in the phenolic resin. It can be seen that the adhesion on the phenolic/flax composite (Figure 1 a, b) is poor, since the flax fibres did not break, but detached (fibre tracks in the matrix can be seen on the micrographs). These tracks were not so pronounced on the composite containing 10 wt.% BC (Figure 1 c,d).

In Figure 4, micrographs of the flax/epoxy composites are shown. As for the composites with the phenolic resin, the composite with 10 wt.% BC (Figure 2 c,d) did not show as much fibre tracks in the matrix as the composite without BC (Figure 2 a,b). BC cannot be differentiated from the epoxy resin in the images, so no further assumptions can be made.

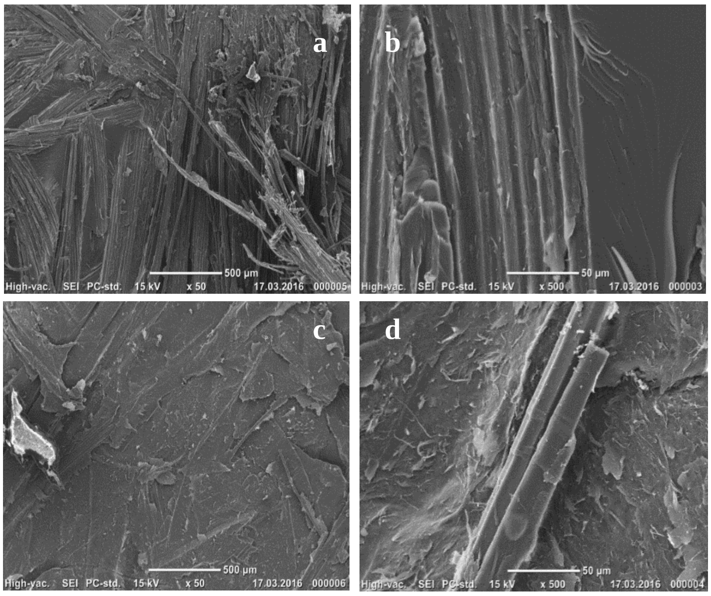

Figure 2. Micrographs of epoxy/flax composites containing 0 wt.%BC (a,b) and 10 wt.% BC (c,d) nonwoven preforms at different magnifications.

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4. Conclusions

The effect of the addition of bacterial cellulose to flax/thermoset composites was investigated. When using a phenolic resin as matrix, the addition of 10 wt.% BC lead only to a slight increase of the tensile strength and modulus of the composites, up to 35.1 ± 2.7 MPa and 9.4 ± 0.6 GPa respectively. The addition of BC to flax/epoxy composites did not improve the properties of the composites. Both thermosetting resins penetrated the nonwoven during the infusion process, for both 0 and 10 wt.% BC content. However, also in both cases, the adhesion was poor, as fibre tracks in the matrix could be observed. These fibre tracks were not so pronounced on the composites containing nonwoven preforms with 10 wt.% BC. The lack of reinforcement of BC when using the different matrices for flax composites could be due to the matrix not being able to totally penetrate the nanocellulose network, impairing impregnation.

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