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Mechanical behaviour and practical adhesion at a bamboo composite interface: Physical adhesion and mechanical interlocking

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ABSTRACT

Physical adhesion was experimentally determined by measuring contact angles with different liquids on bamboo and glass fibres, using the Wilhelmy technique, and by applying the acid–base theory for calculating the surface energy components and the theoretical work of adhesion. The mechanical strength of the interfaces was assessed by single fibre pull-out tests. In order to consider the real mechanisms of interfacial failure of natural fibre composites, the fibre matrix interfacial bond strength was characterised by the critical local value of interfacial shear stress, τ_d, and the radial normal stress at the interface, σ_{ult}, at the moment of crack initiation. Both interfacial parameters are used for correlating thermodynamic work of adhesion and practical adhesion. Pull-out tests (taking into account friction), XPS, and profilometry techniques were used to study the influence of rough natural fibre surfaces on the interface between the fibre and a thermoplastic matrix, by comparing the mechanical behaviour at the interface of a smooth optical glass fibre with that of rough natural fibres. The results suggest that the physical and chemical compatibility between the bamboo fibre and the matrix does not improve substantially the composite performance if compared with glass composites. The relatively low off-axis strength of the bamboo fibres is suggested as the main reason for the low stress transfer capability at the fibre–matrix interphase. Furthermore, the pull-out process may be friction-dominated in bamboo fibre systems.

1. Introduction

With the increasing demand for natural fibre reinforced composites, a lot of effort is put in improving their mechanical properties. The weakest part of these composites is often the fibre–matrix interface. This weakness is usually attributed to a bad compatibility between the typically hydrophilic reinforcing fibre and in particular hydrophobic thermoplastic matrices. To achieve a composite with good mechanical properties, a strong fibre–matrix adhesion has to be obtained by interfacial interactions, including mechanical interlocking, chemical bonding and physical adhesion.

A quantitative estimation of physical adhesion is possible by wetting analysis, while mechanical and chemical interactions can only indirectly be estimated from destructive micromechanical tests (micro-indentation, pull-out, etc.). However, micromechanical experiments measure "practical adhesion", which not only represents purely physical and chemical interactions at the interface. Certainly, the load transfer between the fibre and the matrix also depends on the mechanical properties of both, the fibre and the matrix, and can also be affected by local stresses, matrix residual stresses (processing conditions), presence of easy fracture sites, and the mode of applying external stresses [1,2]. Hence, micromechanical tests not only measure surface interactions but interdependent interface characteristics. Moreover, typical tests characterise the interfacial shear stress which is parallel to the fibre surface, while the adhesion strength is defined as the work required to separate the matrix from the fibre under loading perpendicular to fibre surface [1].

The pull out test is a widely used technique to characterise the mechanical behaviour of the interface between a matrix and a reinforcing fibre in a composite. Theoretical analyses have been developed to study the fibre pull-out process, and these can be divided into two different approaches, using different failure criteria: energy-based and stress-based criteria. The first approach considers that debonding is the result of crack propagation along the

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interface, presenting the energy release rate (G_i) for interfacial crack growth as the failure criterion. The second approach is based on failure at interfacial shear stress. This approach has evolved, using the local shear stress (derived from the "kink" force, where debonding starts) instead of the average stress (derived from the peak force), and considers the local adhesional shear strength value (τ_d) as the intrinsic interface property that characterise the strength of the fibre–matrix interface [3,4]. Both approaches have been proved to be practically equivalent, and they both can predict the debond force as a function of the embedded fibre length [5].

As it was mentioned before, τ_d or G_{ic} are measured under shear loading, parallel (mode II) to the fibre surface while the work of adhesion (W_a) is defined under perpendicular loading conditions (mode I). Since it has been demonstrated that during crack initiation in the pull-out test the crack surfaces move directly apart [6,7], Pisanova et al. [1] suggest the possibility to correctly relate W_a with the normal stress at the debond point. The latter can be calculated using the analytical expressions derived by Nairn and Sheer for the analysis of all stresses in a pull-out specimen [8,9], and the results obtained from pull-out tests.

The aim of this paper is to study the influence of physical adhesion and roughness on the mechanical behaviour of interfaces between a bamboo natural fibre and a polypropylene (PP) and a polyvinylidene fluoride (PVDF) matrix. For comparing the mechanical behaviour at the interface of a smooth fibre with that of rough natural fibres, optical glass fibres were used since they possess a very smooth surface, and constant cross section along the fibre direction, which makes it ideal for reducing the effect of mechanical interlocking. Physical adhesion was experimentally determined by measuring contact angles with different liquids using the Wilhelmy technique and by applying the acid–base theory for calculating the surface energy components and the wetting parameters.

2. Materials and methods

2.1. Materials

The silica core (diameter: 200 μm) of optical glass fibres from Thorlabs (FR200UMT) were used in this study. Technical bamboo fibres of the species Guadua angustifolia were mechanically extracted from bamboo culms in the Department of Materials Engineering at KULeuven. Polypropylene (PP) and polyvinylidene fluoride (PVDF, Solef 1008) were obtained from Propex, and Solvay respectively in the form of films. The selection of these matrices is based on the difference of surface energies between PP (non-polar) and PVDF (polar).

2.2. Materials preparation

Only the silica core of the optical glass fibres is needed, and thus the outer layers have to be removed. The cladding layer is removed by submerging the stripped fibres in hot sulphuric acid. The fibres were submerged in piranha solution (mixture of concentrated sulphuric acid and hydrogen peroxide) for 30 min. Finally, the glass fibres were rinsed off with water and stored in ultrapure water (resistivity > 18 MΩ cm) for avoiding environmental organic contamination. A consistent cleaning of the fibre surface guarantees the same surface chemistry for all the samples, avoiding the chemical or physical interaction of unknown elements during the pull-out tests.

Bamboo fibres were cleaned with warm water for 1 h (90 °C), then wiped with ethanol with a piece of cotton tissue before being dried in a vacuum oven at 80 °C for 1 h. The fibres were then stored at room conditions (60% RH, 20 °C).

2.3. Contact angle measurements and surface energy analysis

Advancing and receding contact angles of various test liquids (ultrapure water: 18.2 Ω cm resistivity, diiodomethane: Merck, and ethylene glycol: Sigma–Aldrich) were measured on the polymer films and glass fibres under controlled conditions (temperature of 20 °C and humidity of 60%), with a Krüss K100 tensiometer using the Wilhelmy technique [10,11]. In order to better describe both the low surface energy and the high surface energy components of the analysed surfaces, the average of the cosines of the advancing (θ_{adv}) and receding (θ_{adv}) angles was used for the glass fibres to estimate the cosine of the equilibrium angle (θ_{equ}), as has been suggested by Andrieu et al. [12], and is shown in Eq. (1).

$$\cos\theta_{equ} = 0.5\cos\theta_{adv} + 0.5\cos\theta_{rec} \tag{1}$$

For the case of bamboo fibres, equilibrium contact angles were measured directly by using acoustic vibration, as it was shown in our previous publication [13].

For evaluating the effect of organic molecules from the air on the surface of glass, some contact angles were measured immediately after the optical fibre were cleaned, and some others after being exposed to our lab environment for a certain period of time.

Surface energy components were calculated according to the Van Oss model and by using the SurfTen 4.3 software developed by Claudio Della Volpe [14]. Also, the work of adhesion (W_a), the spreading coefficient (S), the wetting tension (ΔF), and the interfacial energy (γ_{sl}), which are wetting parameters related to the interfacial strength [15], are calculated according to the following equations:

$$W_a = \gamma_s + \gamma_l - \gamma_{sl} = \gamma_l(1+\cos\theta) \tag{2}$$

$$S = \gamma_s - (\gamma_l + \gamma_{sl}) \tag{3}$$

$$\Delta F = \gamma_s - \gamma_{sl} = \gamma_l\cos\theta \tag{4}$$

$$\gamma_{sl} = \left(\sqrt{\gamma_s^{LW}} - \sqrt{\gamma_l^{LW}}\right)^2 + 2\left(\sqrt{\gamma_s^+} - \sqrt{\gamma_l^+}\right)\left(\sqrt{\gamma_s^-} - \sqrt{\gamma_l^-}\right) \tag{5}$$

2.4. Roughness

The roughness of glass and bamboo fibre surface was measured with a WYKO NT3300 profilometer. In combination with a microscope, it allows roughness measurement of surfaces. The spatial sampling interval ranges from approximately 0.1–10 μm.

2.5. Surface characterisation: X-ray photoelectron spectroscopy (XPS)

XPS analyses were performed on a Kratos Axis Ultra spectrometer (Kratos Analytical – Manchester – UK) equipped with a monochromatized aluminium X-ray source (powered at 10 mA and 15 kV). More information regarding the XPS analysis procedure can be found in our previous publication [10].

2.6. Pull-out test

2.6.1. Sample preparation

A block of polymer was put in an aluminium cylindrical container with a radius of 5 mm and heated until melting temperature. When the polymer was completely molten, the fibre (glass and bamboo) is placed perpendicular to the polymer surface and in its centre with the help of an optical microscope to guarantee accuracy. The embedded length was controlled in the following way: when the fibre entered in contact with the molten polymer, it was pushed down to a certain depth driven by a micrometer with