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Apparent interfacial shear strength of short-flax-fiber/starch acetate composites

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ABSTRACT

The paper deals with an indirect industry-friendly method for identification of the interfacial shear strength (IFSS) in a fully bio-based composite. The IFSS of flax fiber/starch acetate is evaluated by a modified Bowyer and Bader method based on an analysis of the stress–strain curve of a short-fiber-reinforced composite in tension. A shear lag model is developed for the tensile stress–strain response of short-fiber-reinforced composites allowing for an elastic-perfectly plastic stress transfer. Composites with different fiber volume fractions and a variable content of plasticizer have been analyzed. The apparent IFSS of flax/starch acetate is within the range of 5.5–20.5 MPa, depending on composition of the material. The IFSS is found to be greater for composites with a higher fiber loading and to decrease with increasing content of plasticizer. The IFSS is equal or greater than the yield strength of the neat polymer, suggesting good adhesion, as expected for the chemically compatible constituents.

1. Introduction

In order to reduce the environmental impact of composite materials, natural fibers and bio-based matrices are considered as a replacement for the traditional, man-made reinforcements and the matrices derived from petrochemical resources. The production methods and mechanical properties of such green, biodegradable composites manufactured from renewable constituents are being studied extensively as reflected in recent reviews [1–4]. The worldwide capacity of bio-based plastics is expected to increase almost tenfold by 2020 compared with that in 2007, with starch-based plastics among the leading products in terms of production volume [1,2]. Among natural, plant-derived fibers, bast fibers arguably possess the highest potential for reinforcement due to their superior specific axial mechanical properties imparted by a combination of the high content of crystalline cellulose and the low microfibril angle with fiber axis [5].

Since the quality of the interface between the matrix and fibers exerts a major effect on the mechanical properties of composite materials, the compatibility of their constituents is also an important issue for green composites. It has been argued in [6] that acetylation of the free hydroxyl groups of starch with a degree of substitution higher than 2 but lower than 3, while making the material melt-processable, still retains a sufficient hydroxyl functionality in starch acetate for hydrogen bonding with cellulosic fibers. Short-bast-fiber composites with a starch acetate matrix (with a degree of acetylation evaluated at 2.6) and different content of plasticizer were produced and characterized [6]. While the stiffness [7] and tensile strength [8] of the composites have been subjected to detailed studies, the evaluation of the fiber/matrix adhesion achieved has been addressed only via an analysis of composite strength [8].

A commonly used parameter characterizing the adhesion between a fiber and matrix is the interfacial shear strength (IFSS). An extensive range of test methods has been developed for the IFSS, see, e.g., [9], which are also applied to natural fibers. Although the values of IFSS obtained by different methods may not coincide exactly, the results of several commonly used test methods, such as pull-out, microbond, and single fiber fragmentation tests showed the same trends for cellulose (flax, kenaf, Lyocell) fibers [10]. In terms of ease of implementation, the evaluation of IFSS based on an analysis of the stress–strain response of short-fiber-reinforced composites in tension appears to be the

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most industry-friendly adhesion test method [11]. As originally proposed in [12,13], the IFSS and the fiber orientation factor are evaluated from experimental measurements of composite stress at two different values of applied strain within the nonlinear deformation range, assuming a linear elastic matrix response. The method was further developed in [11,14–16] by taking into account the nonlinearity of the matrix response in tension. Subsequently, using the whole experimental stress–strain curve of the composite, rather than only two points, was proposed in [17,18] for estimation of its IFSS. The composite-test-based approach has also been applied to the evaluation of the IFSS of natural fibers, such as, e.g., flax [19–21], hemp [22–24], corn stalk [25], stone groundwood [26], old newspaper [27], and orange tree pruning pulp [28] fibers and polypropylene matrices, as well as hemp fibers and a Mater-Bi® thermoplastic starch matrix [29].

The distribution of stress in reinforcing fibers has been modeled in [12–18] assuming that the stress transfer takes place via a rigid-perfectly plastic interfacial shear response [30]. While such an approach is well substantiated for relatively high strains, neglecting the effect of elastic stress transfer appears to be questionable for small to intermediate strain range, which is also taken into account when the whole stress–strain curve is employed for estimation of the IFSS, as, e.g., in [17,18]. More advanced models of elastic–plastic deformation of short-fiber-reinforced composites, allowing for matrix plasticity and interfacial debonding of fibers, have been developed in, e.g., [31–34], but their application for identification of the IFSS is complicated by the necessity of an extensive numerical analysis [31] or identification of a set of damage parameters [32–34]. In the current study, an elementary shear lag model is developed for the tensile stress–strain response of short-fiber-reinforced composites allowing for an elastic-perfectly plastic stress transfer. The model is applied for evaluation of the apparent IFSS of flax/starch acetate composites with a variable content of plasticizer, produced and tested as described in [6].

2. Materials and methods

The constituents, production, morphological analysis, and testing of short-flax-fiber reinforced starch acetate composites has been described in [6,7]; below, for completeness and the ease of reference, we briefly recapitulate the relevant information.

The flax fibers were supplied by Ekotex, Poland, and an amylose-rich corn starch was obtained from Gargill, USA (Cerestar Amylogel 03003: 65 wt% amylose and 35 wt% amylopectin). The processes of fiber pelletizing, starch acetylation, and plasticization, as well as the compounding, post-processing, and injection molding of composites and the manufacturing of tensile specimens are described in detail in [6]. Nine flax/starch composites differing in the content of plasticizer and fibers, as summarized in Table 1, were produced.

Prior to the mechanical testing, the tensile specimens were conditioned at 23 °C and 50% RH for a minimum of five days. The tensile specimens were tested according to the ISO 527 standard on an Instron 4505 Universal Tensile Tester with a 10 kN load cell and a cross-head speed of 5 mm/min. The strain was measured by an Instron 2665 Series High Resolution Digital Automatic Extensometer. The testing was performed at controlled ambient conditions: 23 °C and a relative humidity of 50%. The cross-sectional dimensions of the gauge area section of each tensile specimen were measured with a slide gauge (± 0.01 mm). Young's modulus was evaluated as the slope of the experimental stress strain diagram within the strain range of 0.1–0.3%.

The volume fractions of constituents were determined by using gravimetric measurements of the densities of fibers, matrix, and composites and the known fiber weight fraction in the composites (as described in detail in [6]). Fiber dimensions (length and diameter) were determined from fibers extracted from composites (the matrix was dissolved in hot chloroform) by optical microscopy and image analyses (see [7] for details). The average apparent fiber diameter and the aspect ratio, defined as the ratio of the average fiber length to its average diameter, for the composites considered are provided in Table 1.

3. An elementary model of the non-linear stress–strain response of a short-fiber-reinforced composite

It has been suggested in [12,13] that the non-linear stress–strain diagram in tension of a short-fiber-reinforced composite can be evaluated by the rule-of-mixtures approach, expressing the composite stress σ_c at an applied strain ε as the sum of fiber and matrix contributions. The resulting relation can be recast as follows:

$$\sigma_c = \eta_o \eta_l \nu_f E_f \, \varepsilon + (1-\nu_f)\, \sigma_m, \tag{1}$$

where η_o and η_l stand for the fiber orientation and length efficiency factors, ν_f is the fiber volume fraction in the composite, E_f denotes the longitudinal modulus of linear elastic reinforcing fibers, and σ_m is the axial stress acting in the matrix. The non-linearity of the composite response may stem from the nonlinear deformation of matrix, $\sigma_m = \sigma_m(\varepsilon)$, and the inelastic stress transfer between the constituents. The latter, for the specific case of a rigid-perfectly plastic interfacial shear response, has been considered in [12,13].

In the following sections, we recapitulate the classical linear elastic shear lag relations and consider expressions of the fiber length efficiency factor η_l in Eq. (1) for the elastic-perfectly plastic and rigid-perfectly plastic stress transfer between the fibers and matrix by shear.

Table 1
Characteristics of constituents.

Composite designation	Plasticizer type	Plasticizer fraction in matrix (wt%)	Yield strength of matrix (MPa)	Fiber loading in composite (wt%)	Fiber aspect ratio	Fiber diameter (μm)
C15	PSA2.1	20	22.6 ± 1.2	10	19	18
C16	PSA2.1	20	22.6 ± 1.2	40	8	19
C17	PSA2.2	20	23.2 ± 1.1	40	9	17
C20	PSA3	25	17.1 ± 0.3	10	19	17
C21	PSA3	25	17.1 ± 0.3	40	15	17
C28	PSA5.1	32.5	6.6 ± 0.2	10	18	19
C29	PSA5.1	32.5	6.6 ± 0.2	40	15	19
C37	PSA6	35	3.9 ± 0.3	10	28	18
C38	PSA6	35	3.9 ± 0.3	40	17	17