APPARENT IFSS IN MISORIENTED FLAX/PP COMPOSITES: THE EFFECT OF FIBER VOLUME FRACTION

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Abstract

Adhesion of the constituents in fiber-reinforced polymers, frequently characterized by the interfacial shear strength (IFSS), is an important characteristic of the composite material determining to a large extent its strength and toughness. A number of experimental methods have been developed for evaluation of the IFSS. However, it has been argued that adhesion should be determined by tests reflecting the actual stress state the fiber/matrix interface is subjected to, rather than that achieved in model single-fiber composites. Such an adhesion evaluation method for short-fiber composites has been proposed by Bowyer and Bader based on the mechanical response of composite material in tension. In the current study, we apply this method to short-flax-fiber reinforced polypropylene to evaluate the effect of fiber volume fraction on the apparent IFSS. Experimental tension curves of unmodified and modified polypropylene matrix composites reinforced with short flax fibers with three different volume fractions of fibers were used to obtain the orientation factor and IFSS. The estimated IFSS and orientation factors are analyzed to determine tendencies and their causes.

1 Introduction

Adhesion of the constituents in fiber-reinforced polymers, frequently characterized by the interfacial shear strength (IFSS), is an important characteristic of the composite material determining to a large extent its strength and toughness. A number of experimental methods have been developed for evaluation of the IFSS [1]. However, it has been argued that adhesion should be determined by tests reflecting the actual stress state the fiber/matrix interface is subjected to, rather than that achieved in model single-fiber composites [2]. Such an adhesion evaluation method for short-fiber composites has been proposed by Bowyer and Bader in [3, 4] based on the mechanical response of composite material in tension.

The method employs a relation of composite stress σ_c and strain ε_c in axial tension [3, 4], that can be expressed as follows

$$\sigma_c = \eta_o \eta_l \sigma_f v_f + \left(1 - v_f\right)\sigma_m \tag{1}$$

where σ_f and σ_m are fiber and matrix stresses, η_l denotes fiber length efficiency factor, η_o stands for fiber orientation factor, and v_f is fiber volume fraction. The factor η_l depends on fiber lengths and the IFSS τ, as detailed in [5]. For linear elastic fibers, $\sigma_f = E_{fL}\varepsilon_c$. If matrix response is linear elastic, $\sigma_m = E_m \varepsilon_c$, the non-linearity in (1) stems only from the growth of the critical fiber length with the applied strain. In [3, 4], a method of estimation of η_o and τ was proposed, applying Eq. (1) to experimental composite stress measurements at two different applied strain values and solving the obtained system of equations for the two unknowns.

The method was further developed in [6-9] by taking into account the non-linearity of the matrix response in tension $\sigma_m(\varepsilon_c)$. Subsequently, fitting the theoretical relation Eq. (1) to the experimental stress-strain curve (rather than just two points) was proposed in [10, 11].

IFSS is usually found to decrease with increasing fiber content [6, 7, 9, 12, 13], (although a few cases have been reported when IFSS was not affected by fiber loading [14] or even increased with it [15]). Such an effect can be explained assuming that the apparent IFSS is due to Coulomb friction

$$\tau = \mu \sigma_r \tag{2}$$

where μ is the coefficient of friction and σ_r designates the (compressive) residual radial stress acting on a fiber. Since the coefficient of thermal expansion (CTE) of polymer matrices is larger than that of fibers and manufacturing processes take place at elevated temperature, compressive internal stress is exerted on the fibers by the matrix at RT. The magnitude of the stress σ_r decreases with increasing fiber volume fraction, thus, according to Eq. (2), explaining apparent reduction in IFSS. It was demonstrated that reasonable agreement with experimental IFSS data can be achieved using Eq. (2) with μ of ca. 0.6 [6, 7, 9, 12, 13].

A modified relation for IFSS, accounting for a stress-independent component, has been proposed [16]

$$\tau = \tau_0 + \mu \sigma_r \, . \tag{3}$$

It should be noted though that there is an inherent contradiction in interpreting the apparent IFSS values obtained via Eq. (1) (assuming a constant, stress-state-independent τ as the interface characteristic) as originating due to radial stress-dependent, see Eqs. (2) and (3), interfacial friction. Such an approach would be strictly valid only in the case of mechanical radial stresses at the interface being negligible compared to σ_r during loading.

The method of Bowyer and Bader has been applied to obtain the IFSS not only of man-made fibers but also such natural fibers as flax [17] and hemp [18]. In the current study, we apply the method to short-flax-fiber reinforced polypropylene to evaluate the effect of fiber volume fraction on the apparent IFSS [19] and to relate the IFSS to the residual stresses.

2 Materials and tests

The constituents and production of the short-flax-fiber reinforced polypropylene (PP) composites has been described in [20], and their deformation diagrams in tension presented in [21]. Both neat PP (Adstif 770 ADXP Basell polypropylene) and a mixture of the PP and maleic anhydride grafted PP (denoted as PPM in the following) were used as matrices of flax fiber (FinFlax, Finland) composites. Composite plates were produced from flax fiber/PP or PPM compound by pressing it under stiff profile. The compound was manufactured by co-extrusion of the polymer and flax fibers. Composites of weight fractions of fibers 20%, 30%, and 40% were produced. Respective fiber volume fractions were evaluated based on constituent densities.

Rectangular specimens of ca. 250 mm length and 25 mm width were tested in tension with the loading rate of 2 mm/min. The axial strain was measured by an extensometer with 50 mm long base. At least five specimens for each material were loaded until failure. Typical stress-strain diagrams are shown in Fig. 1.

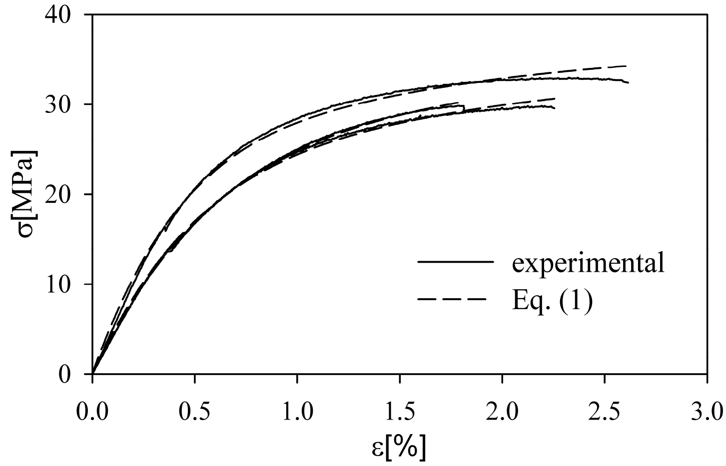

Figure 1. Typical stress-strain curves of short-flax-fiber composites and their approximation by Eq. (1) [19]

3 Evaluation of IFSS

To apply Eq. (1) for estimation of η_o and τ, the rest of the material characteristics entering it have to be specified. For fiber length efficiency factor η_l we use the approach developed in [22, 10], based on an analytical representation of the length distribution of reinforcing fibers. For the Weibull two-parameter distribution of fiber length with scale parameter β and shape parameter α,

$$P(l) = 1 - \exp\left[-\left(\frac{l}{\beta}\right)^{\alpha}\right] \tag{4}$$

the length efficiency factor takes the form [10, 11, 22]

$$\eta_l = \frac{1}{2}\left(\exp\left[-\left(\frac{l_c}{\beta}\right)^{\alpha}\right] - \frac{\beta}{\alpha\, l_c}\left[\Gamma\left(\frac{1}{\alpha}, \left(\frac{l_c}{\beta}\right)^{\alpha}\right) - \Gamma\left(\frac{1}{\alpha}\right)\right] - \frac{l_c}{\beta}\Gamma\left(1 - \frac{1}{\alpha}, \left(\frac{l_c}{\beta}\right)^{\alpha}\right)\right) \tag{5}$$

where the critical fiber length l_c is given by

$$l_c = \frac{E_{fL}\varepsilon_c}{\tau} r_f$$

$\Gamma(x)$ and $\Gamma(x, y)$ are the gamma and the incomplete gamma functions, respectively, and r_f is the fiber radius.

The nonlinear relation between stress and strain in matrix was approximated using a fourth degree polynomial:

$$\sigma_m = C_1\varepsilon + C_2\varepsilon^2 + C_3\varepsilon^3 + C_4\varepsilon^4 \tag{6}$$

4 Results and discussion

FinFlax fibers, used as reinforcement, separate into elementary flax fibers during extrusion [20, 23]. The elementary fibers are characterized by the average axial modulus of E_{fL} = 69 GPa and apparent radius r_f = 8 µm [20, 21]. The parameters of the reinforcing fiber length distribution Eq. (4) were evaluated by the method of moments, using estimates of the average value and coefficient of variation of length obtained from fiber length histogram [20], leading to α = 1.98 and β = 1.37 mm.

The parameters of the fourth degree polynomial describing nonlinear behavior of matrix were evaluated from tension stress-strain curves of neat PP. The following values were obtained: C_1 = 1881 MPa, C_2 = -6.415·10^4 MPa, C_3 = 1.068·10^6 MPa and C_4 = -6.262·10^6 MPa.

The fiber orientation factor and IFSS were determined for each composite specimen tested, as the η_o and τ values ensuring best approximation of the experimental stress-strain diagram by Eq. (1) [19]. The expression Eq. (5) was used for length efficiency factor and matrix stress as a function of the applied strain evaluated using Eq. (6). This way, a reasonably close fit of the test results was obtained as shown in Fig. 1. The average values of the obtained parameters are shown in Fig. 2 as a function of fiber volume fraction.

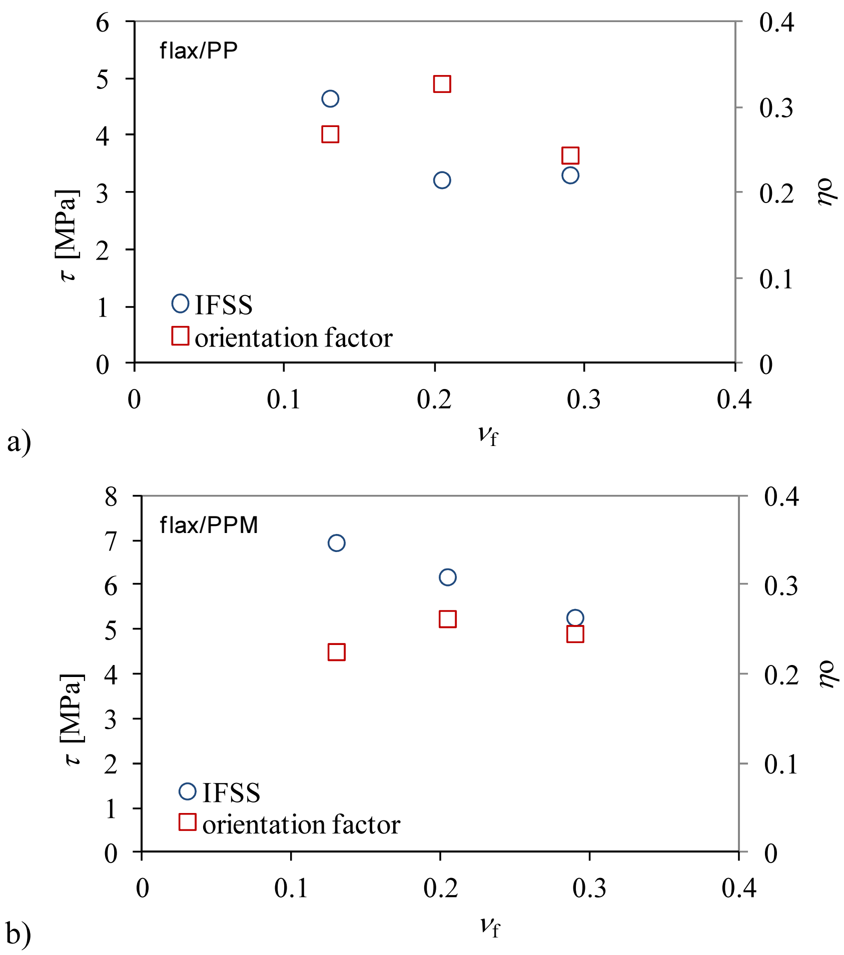

Figure 2. IFSS and orientation factor as a function of fiber volume fraction in flax/PP (a) and flax/PPM (b) composites

Theoretical estimates for fiber orientation factor related to Young's modulus of short-fiber composites, η_{oE}, have been derived in [24]. For random 3D arrangement of fibers, the

maximum theoretical value of η_{oE}, equal to 0.2, is reached when Poisson contraction of the composite is neglected. It is seen in Fig. 2 that the η_o estimates obtained slightly but consistently exceed this value. It is consistent with the presence of a preferential alignment of fibers along the specimen length revealed by optical microscopy [20].
A reduction in IFSS with increasing v_f is seen in Fig. 2. IFSS estimates of flax/PP and MPP obtained by single-fiber tests are in the range of 1.6 to 25 MPa, depending on the fiber retting stage and, possibly, test method applied [25-33]. Model composite tests mostly yield rather high IFSS values, roughly comparable or exceeding those determined in this work by the Bowyer and Bader method using short-fiber composites with the lowest fiber volume fraction. The likely reasons for such a discrepancy between IFSS obtained from single-fiber and short-fiber composite tests are discussed in [2].
The dependence of the apparent IFSS on the fiber volume fraction in a composite material can be related to the variation of the residual stresses with v_f [6, 7, 9, 12, 13]. In such a case, the interfacial interaction of the fibers and matrix is characterized by the coefficient of friction, and the IFSS under given residual stress predicted by Eqs. (2) or (3). In order to evaluate the respective interface parameters, the radial compressive stress acting on fibers has to be determined. The stress arises do to the mismatch in thermal expansion of the fibers and matrix, hence it depends on the radial stiffness and CTE of the fibers.
Neither of these parameters has been measured for the flax fibers used in the composites considered. However, a rough estimate of σ_r can be obtained using the fiber transverse modulus E_{fT}= 7 GPa [34] and substituting for the transverse CTE the value obtained for different bast fibers, those of jute [35]: CTE = $7.72\cdot10^{-5}$ K^{-1}. The radial thermal stress σ_r was calculated as described in [36], considering the dependence on temperature of CTE of PP. Fig. 3 shows the apparent IFSS as a function of the residual stress. It is seen that the observed dependence of IFSS on σ_r can be reasonably well approximated by Eq. (2) both for PP and PPM matrices. Hence Eq. (2) can be applied to predict the apparent IFSS at other fiber volume fractions or test temperatures, leading to different residual stresses.

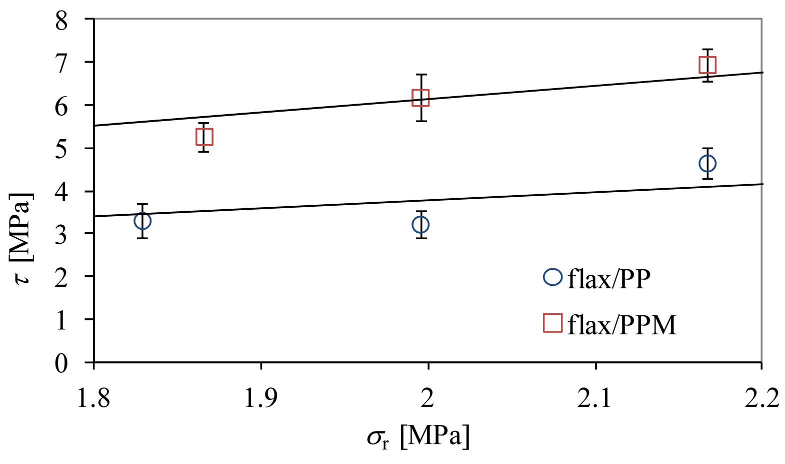

Figure 3. The dependence of IFSS on the residual (compressive) stress acting on the fibers of flax/PP and flax/PPM composites. Lines show approximation of the data by Eq. (2)

Conclusions
The apparent IFSS and fiber orientation factor have been determined by a modified Bowyer and Bader method for short-flax-fiber composites with PP and PPM matrices. The IFSS was observed to decrease with increasing fiber volume fraction in the composites. This finding can

tentatively be explained by the friction-dominated stress transfer to the fibers, which is sensitive to the residual stresses.

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