Interfacial shear strength of flax fiber/thermoset polymers estimated by fiber fragmentation tests

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A number of natural fibers are being evaluated as an environmentally friendly alternatives to glass reinforcement in short fiber reinforced polymers, including flax fibers. Adhesion between reinforcing fibers and polymer matrix is crucial for composite strength. A mechanical parameter commonly used to characterize the adhesion is interfacial shear strength (ISS), measured for flax/polymer systems by pull-out [1–3], microdebond [4], and single fiber fragmentation (SFF) tests [5–8] (see also review [9]). Several fiber surface treatments for improving the adhesion with thermoplastic (particularly polypropylene) matrices have been considered, while less research is dedicated to thermoset polymers. We study flax/thermoset polymer ISS for most common types of thermosets: vinylester, polyester, and epoxy. The effect of fiber surface treatment on ISS in such systems reinforced with flax fibers is investigated. Apparent ISS is evaluated from SFF tests by the Kelly–Tyson approach utilizing fiber strength at the critical length (see e.g., [10]). Fiber strength at such small lengths is hardly accessible to direct testing, therefore it is usually extrapolated from test results at larger gauge lengths. We present ISS of flax fibers subjected to different treatments and thermoset polymers, derived from SFF tests using comprehensive fiber strength data [11].

Enzyme-retted flax fibers delivered by Finflax Oy (Finland) were used. The fibers were stored and tested at ambient conditions. Both flax fibers without any additional coating and treated fibers were used. The resins considered were vinylester (VE), unsaturated polyester (UP), and epoxy (EP). Two different types of surface treatment were applied in the case of VE and UP matrices: fibers were treated with acrylic acid (AA) and vinyl trimethoxy silane (VTMO). Two different intensities of treatment (in terms of the active ingredient concentration) were considered for each treatment referred to by indices 1 and 2 in the following (higher index stands for higher concentration). Maleic anhydride (MA) treatment was applied to fibers only in the case of epoxy resin.

Two types of single fiber composite (SFC) specimens were manufactured and tested differing in fiber and hence SFC gauge length. Short-fiber SFC specimens were prepared by mounting elementary flax fibers on a 1-mm thick steel frame, using double-sided adhesive tape. Small rectangular steel pieces of the same thickness were placed on the frame in order to position fiber in the middle. The frame was then placed between two flat Teflon-coated aluminum mould plates, separated by spacers of 2-mm thickness and provided with a silicon tube sealing. After the resin had solidified, the mould was placed in the oven for postcuring. VE and EP were postcured at 50 °C for 2 hr and 80 °C for 5 hr, UP was postcured at 50 °C for 2 hr. The plates were cut and polished into specimens with the dimensions $L \times W \times H$: $L = 20$–30 mm, $W = 3$–4 mm, $H = 2$–2.5 mm. The gauge length was 10–20 mm depending on the specimen size. Long-fiber SFC preparation procedure differed only in that elementary flax fibers were modified by gluing fiber extensions on both ends of the flax filament by fast drying glue. Thin fishing line of 90-μm diameter was used as fiber extensions. This made it possible to increase the SFC specimen length L to about 100 mm and gauge length to 40–50 mm. Long-fiber SFCs were manufactured with untreated and AA-treated fibers in VE matrix.

The SFF test was performed in a MINIMAT miniature mechanical test machine from Polymer Laboratories Ltd. (UK). The test machine was mounted on the $x - y$ table of a Zeiss optical microscope. The MINIMAT machine was manually controlled. Load was measured by the MINIMAT's built-in load cell (1000 N) and the displacement was registered by the electronic unit of the tensile stage. Fragmentation of the fibers was observed during the loading. For long-fiber SFCs, an extensometer was used to measure applied strain, while for short-fiber SFCs the strain was evaluated from grip displacement corrected for setup compliance. In order to avoid pausing the machine to count fiber cracks, the test was carried out at a rather low loading rate of 0.1 mm/min. Loading was stopped if the specimen failed, or when the fragmentation saturation level was achieved. The latter was defined as occurring when no new fiber breaks appeared during a

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TABLE I Interfacial shear strength obtained by SFF

Matrix	Modifying factor	Number of SFF tests	Average fibre diameter d (μm)	Average fragment length $\langle l \rangle$ (μm)	Interfacial shear strength τ (standard deviation) (MPa)
Vinylester	–	13	19.4	407	28 (11)
	AA_1	4	15.7	338	25 (4)
	AA_2	10	18	326	31 (6)
	$VTMO_1$	4	19.7	494	20 (2)
	$VTMO_2$	3	18.7	450	21 (3)
Polyester	–	4	19.2	703	13 (2)
	AA_1	3	19.4	610	15 (3)
	AA_2	4	19.5	821	13 (5)
	$VTMO_2$	5	18.8	657	14 (2)
Epoxy	–	3	18.1	299	33 (7)
	MA	3	16.4	364	24 (3)

strain increase by 0.5%. For some specimens, loading was continued up to specimen failure (at strain >7%) thus checking that the saturation had indeed occurred. The saturation strain did not exceed 6%. Only the data from specimens that reached saturation were used for interfacial shear strength estimation. As long-fiber SFC fragmentation behavior was the same as for short-fiber SFCs, no distinction between SFC types is made below.

During the experiment the data were transferred to the PC. In order to measure the fiber diameter, digital pictures of the fibers were made before the loading. Images were made by the CCD camera attached to the microscope and then transferred to the PC for further processing. Fiber diameter was evaluated from analysis of digital images as the average of five apparent diameter measurements taken along the fiber.

The apparent interfacial shear strength, τ, is estimated as

$$\tau = \frac{\langle \sigma \rangle d}{2 l_c} \tag{1}$$

where d is the fiber diameter, l_c is the critical length related to the average fiber length at saturation of the fragmentation process, $\langle l \rangle$, as $l_c = \frac{4}{3}\langle l \rangle$ and $\langle \sigma \rangle$ is the average fiber strength at critical length.

Testing of the elementary flax fibers at several gauge lengths revealed that fiber strength comply with the modified Weibull distribution [11]

$$P(\sigma) = 1 - \exp\left(-\left(\frac{l}{l_0}\right)^{\gamma}\left[\frac{\sigma}{\beta}\right]^{\alpha}\right) \tag{2}$$

The parameters of the distribution (2) determined by the maximum likelihood method from strength data in 5–20 mm gauge length interval are as follows: $\gamma = 0.46, \alpha = 2.8, \beta = 1400$ MPa [11] (taking $l_0 =$ 1 mm). It follows from Equation 2 that the average fiber strength as a function of fiber length is given by

$$\langle \sigma \rangle = \beta (l/l_0)^{-\gamma/\alpha} \Gamma(1 + 1/\alpha) \tag{3}$$

The ISS values determined by Equations 1 and 3 from SFF tests are summarized in Table I. Note that ISS estimates for UP and epoxy are in rough agreement with pull-out test results for similar systems [2, 9]. The ISS for epoxy and VE matrices is somewhat higher than that for UP. The surface treatments considered do not lead to significant variation of ISS, the effect, if any, being within the ISS scatter band. Hence, it can be concluded that the adhesion of the enzyme-retted elementary flax fibers and thermoset matrices does not benefit from the common surface treatments.

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