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Failure mechanisms in composites reinforced with unidirectional *Phormium* leaf fibre

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ABSTRACT

Confocal microscopy, field-emission SEM and acoustic emission experiments were used to investigate fracture mechanisms in composites made from epoxy resin reinforced with unidirectional *Phormium tenax* (harakeke) leaf fibre. Resin filled the lumens of a large proportion of thin-walled vascular cells and bundle sheath cells, and also filled some of the fibre cells in assemblies that had been split during fibre processing, but rarely penetrated intact thick-walled fibre cells. Vascular tissue and cuticular matter were particularly susceptible to brittle fracture. Cell–cell debonding was abundant on fracture surfaces. Low-, medium- and high-energy acoustic events showed transient signals of similar duration, constructed from similar frequencies and differing only in amplitude. The wide distribution of event energies was attributed to the diversity of types of technical fibres, from assemblies of a few thin-walled cells to assemblies of hundreds of thick-walled fibre cells.

1. Introduction

Life cycle assessments of composite materials reinforced with plant fibres have shown advantages relative to composites reinforced with glass fibres [1–3]. The assessments have assumed that replacement of glass fibres with plant fibres will have little influence on the mechanical performance of the composites. Single-fibre tests showed promising performance data, e.g., a mean tensile strength of 1.73 GPa was reported for hemp fibres carefully isolated by hand [4], compared with values of 1.95 or 2.28 GPa reported for glass fibres [5,6]. Those results seemed even more impressive when expressed as specific tensile strengths, since the density of a typical plant fibre is little more than half that of glass. Unfortunately, when plant fibres were used to reinforce thermosetting resins, the composites were much weaker than those reinforced by glass fibres [7–11], even when the comparisons were based on specific tensile strengths. Table 1 summarises published results for thermosetting resins reinforced with unidirectional flax or sisal fibres. Flax and sisal were selected as examples of bast and leaf fibres, respectively. While the results for both species show scatter, all of the tabulated specific tensile strengths are less than those for unidirectional glass fibre. The present study investigates failure mechanisms for composites reinforced with plant fibres, including the consequences of variable mixtures of cell types in plant fibres.

Variable quality is an important feature of plant fibres. We cannot expect the performance of commercial plant fibres to match the performance of fibres carefully extracted by hand. For example, the mean tensile strength of flax fibres was reported as 1.83 GPa for hand decortication and 1.52 GPa for standard mechanical decortication [12]. Even if the fibres are extracted by hand, the tensile strengths can be scattered widely around the mean value. Coefficients of variation of 0.42 and 0.49 were reported for hand-extracted hemp and flax, respectively [4,12]. For comparison, coefficients of variation of 0.32 and 0.20 were reported for glass fibres [5,6]. A high coefficient of variation means that some natural fibres are considerably weaker than others, providing potential crack initiation points.

Technical fibres obtained from plants are aggregates of ultimate cells, sometimes containing mixtures of fibre cells and vascular bundles [11]. Other potentially variable features of plant fibres, but not of glass fibres, include:

(a) Debonding of hydrophilic fibre surfaces from hydrophobic matrices [13].
(b) Variability of cross-sectional shapes and dimensions of technical fibres [14].
(c) Cell–cell debonding within technical fibres [15].
(d) Partial penetration of the matrix polymer into fibre cells [16].

The overall result of all of these features seems likely to be depression of the mechanical performance of composites

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Table 1
Published properties for unidirectional composites. UPE = unsaturated polyester.

Fibre	Resin	Volume fraction	Specific tensile strength (kPa kg^{-1} m^3)	Reference
Glass	Epoxy	0.48	478	[7]
Glass	UPE	0.42	413	[8]
Flax	UPE	0.58	233	[8]
Flax	Vinyl ester	0.37	165	[9]
Flax	Epoxy	0.51	102	[10]
Sisal	Epoxy	0.50	218	[11]
Sisal	Epoxy	0.51	180	[7]

reinforced with commercial plant fibres, relative to composites reinforced with plant fibres carefully selected and processed in a laboratory.

We used confocal microscopy to characterise the diversity of technical fibres in a natural-fibre composite. We then used acoustic emission to characterise the distribution of energies involved in events leading up to fracture [17], and field-emission SEM to characterise the fracture surfaces. We chose to test composites made from epoxy resin reinforced with unidirectional *Phormium tenax* leaf fibre. This choice was based on the availability of published background knowledge concerning *Phormium* technical fibres and their composites [18–21]. In particular, measurements of single-fibre tensile strength have shown coefficients of variation of 0.40, 0.68 and 0.87 for technical fibres from three different *Phormium* cultivars [18]. These coefficients of variation are larger than those observed for glass fibres [5,6], and at least as large as those reported for bast fibres, e.g., hemp and flax [4,12]. We therefore expected to see variability in the resistance offered to crack propagation.

2. Experimental

2.1. Materials

Preparation of the *Phormium*–epoxy composites has been described elsewhere [19,20]. *Phormium* (harakeke) technical fibre was obtained from the Templeton Flax Mill, Riverton, New Zealand. The fibre was extracted from a wild stand, not a cultivar. Unidirectional composites were made from technical fibre that was combed, bound in bundles, cut to 450 mm, and dried at 103 °C for 2 h before impregnation. The bundles were impregnated with epoxy resin (Nuplex R300/H310) in a vacuum bag and compressed in a mould similar to that used to make sisal–epoxy composites [22]. Excess resin escaped from gaps at each end of the mould. Rectangular specimens with nominal dimensions 450 mm × 25 mm × 4 mm were cured in the mould overnight at room temperature, released and post-cured for 48 h at 60 °C.

An additional specimen (UD4) contained fluorescent dyes for use in confocal microscopy. The method of preparation has been described elsewhere [20]. The fibre was dyed by immersion overnight in 10 mg L^{-1} aqueous acriflavine. The matrix was labelled by adding 30 mg safranine to 100 g epoxy resin, stirring at 80 °C for 20 min, then cooling to room temperature prior to mixing with hardener. Since both fibre and matrix fluoresced, any black regions in the images represented voids.

2.2. Methods

Confocal fluorescent microscopic (CFM) images were acquired using a Leica TCS/NT confocal laser microscope. The samples were illuminated with a Kr/Ar laser, and the resulting fluorescence was recorded. All images were recorded with a 16× lens.

Field-emission scanning electron microscope (FESEM) images were obtained using a Jeol JSM-6700F instrument. Each specimen was fastened onto a support with a 12 mm diameter disc of carbon tape. The specimen was coated with chromium under a 10^{-4}/10^{-3} mbar vacuum and scanned at a 3 kV acceleration tension.

Tensile testing was based on ASTM D3039 M-00e1 (Standard Test Method for Tensile Properties of Polymer Matrix Composite Materials), with friction grips and a crosshead speed of 2.0 mm/min. Specimens were cut to 250 mm length and conditioned at 50% relative humidity and 23 °C. Axial strain was measured by a standard Instron clip–gauge extensometer with a 25 mm gauge length. Multiple-loading tests were undertaken by initially loading each specimen to approximately 30 MPa stress, unloading to approximately 1 MPa, loading to 50% of expected ultimate strength, unloading again, loading to 75% of expected ultimate strength, unloading again, then reloading to failure.

Acoustic emission monitoring used a Vallen AMS-3 system with a single sensor. Events were recorded if they exceeded an amplitude of 40 dB relative to an arbitrary limit. This threshold was chosen to exclude the majority of background noise. The use of a servo-hydraulic instrument, rather than an electromechanical instrument, contributed to the exclusion of background noise. The relationship between mechanical energy and electrical energy is poorly defined, mainly due to the non-linear frequency response of a typical transducer. For this reason, energies were presented in arbitrary units and their use should be confined to comparisons between experiments based on this instrument.

3. Results and discussion

3.1. Composite densities

Densities were determined as a test for porosity. The theoretical density of a void-free composite is [23]:

$$1/\rho_c = f_w/\rho_f - (1 - f_w)/\rho_m \quad (1)$$

Here f_w is the fibre weight fraction, and ρ_c, ρ_m and ρ_f are the densities of the composite, the matrix and the fibre, respectively. The density of the unreinforced epoxy resin used in our work was ρ_m = 1138 kg m^{-3} [24]. No data were available for *Phormium* leaf fibres, so we plotted $1/\rho_c$ against f_w for 11 composite specimens. We then used Eq. (1) to obtain a best-fit value of ρ_f = 1180 kg m^{-3}. Using this best-fit value predicted the experimental values of ρ_c with a root-mean-square deviation of just 18 kg m^{-3}, which is similar to our estimate of the uncertainty in measuring density. Similar experiments on epoxy composites reinforced with pulped *Phormium* indicated ρ_f = 1500 kg m^{-3} [24]. The relatively low fibre density for raw *Phormium* fibres was attributed to air-filled cavities, not penetrated by the resin. This point is discussed further below (Section 3.3).

The best-fit fibre density was used to calculate the fibre volume fractions listed in Tables 2 and 3.

Table 2
Results from single–load tensile tests on unidirectional *Phormium*–epoxy composites. Entries are mean values for four specimens.

Property	Mean	Std. dev.
Fibre weight fraction	0.52	0.01
Fibre volume fraction	0.51	0.01
Density (kg m^{-3})	1164	20
Initial tensile modulus (GPa)	14.7	0.8
Strain (%) at maximum stress	1.54	0.11
Tensile strength (MPa)	211	10