



Tailoring the vibration damping behaviour of flax fibre-reinforced epoxy composite laminates via polyol additions



Marie-Joo Le Guen^{a,*}, Roger H. Newman^a, Alan Fernyhough^a, Mark P. Staiger^b

^a Scion, Private Bag 3020, Rotorua 3010, New Zealand

^b MacDiarmid Institute for Advanced Materials and Nanotechnology, Department of Mechanical Engineering, University of Canterbury, Private Bag 4800, Christchurch 8140, New Zealand

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ABSTRACT

In the present work, the addition of glycerol and polyglycerol was found to increase the damping coefficient of flax composites by 10–13% and 21–25%, respectively, according to dynamic mechanical thermal analysis and acoustic testing. The damping coefficients of the polyol-treated flax composites were larger than those of the non-treated flax and the aramid- or carbon-fibre composites prepared under similar conditions. The relatively high damping behaviour of polyol-treated flax fibre reinforced composite is attributed to a stick–slip mechanism that involves the cyclic breaking and reformation of hydrogen bonds between the polyols and lamellae of the cell walls or microfibrils within the lamellae.

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1. Introduction

Plant fibres are promising materials for the reinforcement of polymers, particularly where the absorption of mechanical energy is a desirable property [1–3]. The ability of natural fibres to absorb energy is attributed to (a) the interaction between various biopolymers found within the cell walls of plant fibres [3,4], (b) microfibril angle of the cellulose within the cell wall [5,6], (c) presence of kink bands that disrupt the orientation of the microfibrils along the fibre [7]. Vibration damping in flax fibre under stress is attributed to friction between the cellulose microfibrils and the polysaccharide matrix (e.g. hemicellulose, pectin) within the cell walls and between adjacent cells in fibre bundles [3]. The friction within the cell wall of the fibre is caused by hydroxyl groups capable of forming multiple hydrogen bonds between adjacent molecules of cellulose and/or hemicellulose and pectin and therefore incapable of slippage without energy dissipation [8].

The mechanisms of energy absorption in cellulosic materials such as wood have been investigated. The capacity of wood to damp vibrations is influenced by the chemical composition, particularly the presence of hemicellulose [9,10]. The hydrogen bonds between cellulose and hemicellulose molecules are thought to break and reform upon mechanical loading and unloading, respectively. The reversible nature of hydrogen bonding in this context is referred

to as the stick–slip mechanism [8,11] and has been compared to Velcro® at the nanoscale [12]. The stick–slip mechanism is the basis for the elastic–plastic response of wood that exhibits the retention of high stiffness following plastic deformation [8,11,12]. Additionally, the presence of compounds that take part in hydrogen bonding (e.g. water) can alter the elastic–plastic response including the mechanical damping of the system [13–16]. For example, Obataya et al. reported that the vibration damping of wood is at a local maximum when the moisture content is ~1 wt.% due to the motion of the adsorbed water molecules [9]. Then the vibration damping of wood passes through a second maximum when the moisture content is ~20 wt.% that is attributed to increased molecular motion of the hydrated hemicellulose [9].

In the field of wood preservation, polyethylene glycol and glycerol are routinely used as bulking compounds to prevent wood from swelling or shrinking due to environmental moisture variation [17–19]. Following impregnation and drying of the wood, and when considered at atmospheric pressure, these compounds are retained in the cell wall due to their high boiling point (e.g. 290 °C for glycerol). They substitute the water within the cell wall when applied to never-dried wood potentially decreasing water absorption in equilibrium conditions [17,19]. Furthermore, they increase the strength and strain to failure of wood by minimising the shrinkage stresses that would normally accompany moisture removal during the wood drying process [19]. The hydrophilic nature of glycerol assists with its penetration into the cell wall of wood, but does not affect the equilibrated moisture uptake. For

* Corresponding author. Tel.: +64 73435490.

E-mail address: mariejoo.leguen@scionresearch.com (M.-J. Le Guen).

example, a loading of 50 wt.% glycerol in wood reduces the diffusion rate of moisture, although the final moisture content is similar to the untreated wood [18]. Polyglycerols are less hydrophilic than the monomer and due to their larger molecular weight, they can block pores and opening in wood cells decreasing water diffusion and/or absorption in the cell wall [18]. Yet they do not penetrate the cell wall to the same extent, for example, the penetration of wood by polyethylene glycol is not possible if the degree of polymerisation is greater than 10,000 [17].

In the present work, the use of oligomeric or polymeric bulking molecules is investigated as a means of increasing the vibration damping of the composite reinforcement. Specifically, the impregnation of flax fibre (*Linum usitatissimum*) with polyols prior to consolidation of the fibres in resinous matrix is investigated. At low polyol concentrations, it is expected that such a fibre treatment will contribute to energy dissipation through the stick-slip mechanism. However, the treatment of fibres with polyols should not increase moisture uptake of the final composite material, or decrease dramatically the elastic modulus and strength if it is to be viable as a commercial process. An underlying objective of this work was to deepen the current understanding of the mechanisms of mechanical damping and role of friction at the fibre-matrix interface in bio-based polymer composites by comparing flax, carbon and aramid fibre-reinforced epoxy composites.

2. Experimental procedures

2.1. Materials

The epoxy resin system was based on diglycidyl ether of bisphenol A (EPON 828, Shell Chemicals), and methyl tetrahydrophthalic anhydride (Lindride 52, Lindau Chemicals) was used as the curing agent. 1-methylimidazole was used as a catalyst (Sigma Aldrich). The ratio of resin:curing agent:catalyst was 100:87.5:1.5 [20]. All chemicals were used as received.

A linen flax 2/2 twill fabric (Biotex, Composites Evolution Ltd.), marketed specifically as a reinforcement for bio-based composites, was used to fabricate flax-epoxy composite laminates. Plain-weave aramid fabric (Kevlar[®] K49 from Dupont) with a areal weight of 170 g m⁻², and 2/2 twill carbon fabric (Made from Toray 300 yarn) with a areal weight of 240 g m⁻² were purchased from Fibretech Solutions Ltd in Rotorua. The aramid and carbon fibre-reinforced composites were used for comparisons with the flax-reinforced laminate.

The flax fabric was woven from twistless yarn double-wrapped with polyethylene terephthalate filament that accounted for 13 wt.% of the fabric following oven-drying at 105 °C for 2 h to remove any residual moisture. The linear density of the linen yarn was 250 tex, and the areal weight of the fabric was 420 g m⁻². Prior to use the as-received flax textile layers were cleaned of dust particles by boiling in water (75 g oven-dried textile to 1 L of water), drying at room temperature overnight, and finally drying at 105 °C for 2 h, leading to an overall mass loss of 2.01 wt.% based on the oven-dried textiles.

Glycerol and polyglycerol were used as-received (Solvay chemicals). The dynamic viscosity of glycerol and polyglycerol were 1 and 41 Pa s, respectively. The molecular weight of glycerol is 92.1 g mol⁻¹. The average molecular weight of the polyglycerol was 250 g mol⁻¹. The mixture included triglycerol at 47 wt.%, 33 wt.% of di- and tetramer and 20 wt.% (or less) of oligomers larger than the tetramer. The designation of PG3 was used for the polyglycerol as the distribution of oligomers peaked at the trimer. Aqueous solutions of glycerol and PG3 were prepared at a concentration of 2 wt.% using distilled water. The flax textiles were then immersed in the solutions at room temperature for 24 h, using a

fabric to solution weight ratio of 2:100. The flax textiles were then removed from the solutions and dried overnight at 45 °C and then at 105 °C for 2 h.

2.2. Preparation of composite laminates

The epoxy resin was mixed at room temperature and degassed for 1 h in a desiccator under vacuum. The textile layers were individually preimpregnated with the epoxy resin by hand lay-up and pre-cured at 105 °C for 10 min. The prepreg laminae were then stacked on an aluminium plate to give a symmetric [0,90] cross-ply spacers and placed in a vacuum bag. The laminate stack was kept under vacuum at room temperature for 1 h and then placed in a Siempelkamp hydraulic hot press to be consolidated and cured (Fig. 1). The composite laminates were allowed to cool overnight under vacuum at room temperature, and then post-cured in 2 steps at 105 °C for 24 h and 150 °C for 6 h. The carbon, aramid and flax fibre-reinforced epoxy laminates had final thicknesses of 1.4, 1.7 and 2.8–2.9 mm, respectively. Each flax-fibre composite was made in duplicate.

2.3. Physical properties of the composite laminates

Fibre weight fractions in the composite laminates were obtained by dividing the oven-dried mass of the reinforcement after impregnation by the mass of the post-cured laminate panel. Conversion to a volume fraction required a value of density for the impregnated fibre. The density of the fibre was calculated based on the density of the composite, density of the epoxy (1160 kg m⁻³ as determined by pycnometry), and the rule of mixtures, neglecting the presence of voids. The moisture content was determined by measuring the conditioned weight of the fibres or the composites stored two to 4 weeks at 23 °C, 50% RH. Then, the materials were dried at 105 °C for 2 h or overnight for the fibres or the composites respectively and re-weighed after cooling them down to room temperature in a desiccator containing silica gel.

2.4. Vibration testing

The modulus of elasticity (E) and loss coefficient (η) of each composite was determined in longitudinal and flexural modes of vibration [21]. Each specimen was excited by a manual impulse and the resulting vibration decay was recorded with a microphone (THS 130P10 SN6156) plugged to an amplifier from National Instruments (NI USB-4431) as described in the ASTM E1876-09.

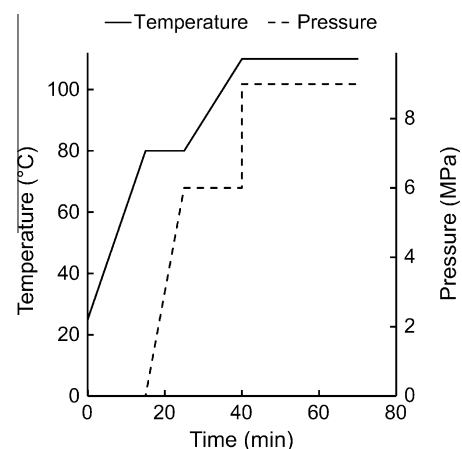


Fig. 1. Curing and compaction profile used during fabrication of the composite laminates.

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