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Improving the strength and service life of jute/epoxy laminar composites for structural applications

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ABSTRACT

Natural fibers are lighter, less expensive, and less abrasive than synthetic fibers such as glass or carbon, and provide a resource that is both renewable and biodegradable. This study uses surface treatments and flock fiber based *Z*-axis reinforcement technology to improve the strength and fracture toughness properties of natural fiber composites (NFCs), thus increasing their ability to bear loads. The enhancement of performance properties will expand applications of NFCs in industries such as automotive and housing/construction industries. NFCs will also provide a more sustainable, environmentally friendly, and inexpensive structural material alternative over traditional resin reinforced glass or carbon fiber composite materials.

1. Introduction

Natural fiber composites (NFCs) are materials in which at least the reinforcing fibers are derived from renewable and carbon dioxide neutral resources such as wood or plants. NFCs are used in molded articles that require moderate strength for acceptable performance such as equipment housings, roofing for low-cost housing, and in large diameter piping [1]. Last decade automotive, construction, furniture and packaging industries are demanding a shift of their design and material selection from oil-derived polymers and mineral reinforcement materials to natural materials to exploit the recyclability and/or biodegradability of "green" products at the end of life. However, current building products and automotive components are non-structural applications, which limit the product spectrum and market size.

For NFC components to become major source in load-bearing applications, a significant amount of research is required to overcome the shortcomings of biocomposites: natural fiber uniformity and strength and modulus; interfacial adhesion and lessen moisture susceptibility; bio-based thermoset and thermoplastic matrices of programmed biodegradability; fabrication processes to increase fiber content and fiber alignment, delamination resistance behavior to maximize performance of the NFCs [2].

The focus of this study is to overcome these drawbacks and manufacture reasonably performing natural fiber reinforced structural composites for automotive, construction and other applications. Natural fibers such as flax, ramie and hemp can be used for replacement of glass fiber reinforcement as structural composites for automotive components and building material applications. Biagiotti et al. pointed out that these fibers have comparable tensile strength and specific modulus with E-glass fibers [3].

These natural cellulosic fibers show irregular cross-section geometry, varying surface chemical composition and fiber length distribution, which cause poor adhesion with traditional matrix resin materials. These characteristics make these natural fiber reinforcements prone to dimensional instability due to swelling and microbial attack [4]. In this study we will explore feasibility of mitigating these shortcomings of natural fibers using fiber surface treatments such as alkali treatment and silane coupling.

All laminated composite materials suffer from service life limiting delamination failure. This poor inter-laminar shear stems from the lack of *Z*-direction inter-ply fiber reinforcement between the assembled numbers of composite laminar plies. Traditional methods to impart the *Z*-direction inter-ply fiber reinforcement were special pre-form fabrics fabricated using technologies such as multi-directional knitting, 3-D weaving or through-the-fabric stitching and *Z*-pinning processes [5,6]. While these methods were found to be improving delamination resistance, but they were not cost-effective and design specific restriction [7–9]. To overcome drawbacks of these cumbersome, high cost, design limited techniques, the University of Massachusetts at Dartmouth (UMD) developed a novel *Z*-axis fiber reinforced resin laminar composite materials processing technique [10–14]. The principle of this inter-laminar strength improvement technique involves the electro-static pre-placement of perpendicularly oriented short fibers between the laminar plies during a composite's

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fabrication/lay-up stage. Short fiber placement is accomplished by electro-statically propelling short fiber elements at an uncured polymer (fluid) matrix coated fibrous ply layer of the composite. This electrostatic fiber coating process is called "flocking".

This study aims to address current gaps in understanding by characterizing the influence of surface treatments, reinforcement architecture, and applied through-thickness microfiber reinforcement on the interlaminar fracture and delamination resistance of jute fiber/epoxy laminated composite materials. Tensile and in-plane shear tests were also conducted to gain a more holistic understanding of the effect of through-thickness reinforcement on mechanical properties.

2. Experimental

2.1. Materials

Plain weave jute fabric with areal weight of 330 g/m^2 was procured from Rose Brand in Secaucus, NJ. Unidirectional jute yarn preforms were fabricated with a linear density of 1440 tex, provided by Stuart C. Hurlbert & Co. Inc. Framingham, MA. Two part epoxy resin system 2000/2120 series amine-cure supplied by FiberGlast (Brookville, OH) was used. Formulation of stoichiometric ratio (27 parts currant per 100 parts resin) was used. Sodium Hydroxide pellets (NaOH), acetic acid, 3-Triethoxysilylpropylamine (APTES), phenolphthalein indicator were purchased from Fisher Scientific (Pittsburg, PA) and used as received. Dow Corning Z-6173 silane coupling agent was obtained from Univar Inc., Providence, RI. The flock fibers for *Z*-axis reinforcement (nylon fibers 3-denier, 1.3 mm length) was provided by Spectro Coating, Leominster, MA.

2.2. Fiber surface treatment

Jute fabrics were held taut to prevent shrinkage on a square frame and soaked for 2 h in a 5% (w/w) aqueous NaOH alkali solution. Fabrics were then rinsed with distilled water and soaked in a 2% (w/w) aqueous acetic acid solution for an additional 1 h. Neutrality of the treated fibers was verified using a phenolphthalein solution followed by thorough rinsing with distilled water and drying at 80 °C overnight. The procedure for silane coupling was adapted from that used by Seki [15] and Sarikanat [16]. A solution of 1% (w/w) Dow Corning Z-6173 silane agent in methanol was prepared and mixed with a magnetic stirrer for 5 min. Alkali treated jute fabrics and unidirectional were soaked in the prepare silane treatment solution for 1 h. The silane treated the preforms were rinsed well with distilled water and dried overnight at 100 °C.

2.3. NFC panel fabrication

The vacuum infusion (VI) procedure as optimized for use with natural fiber reinforcement in previous work [5]. The desired number of 260 mm × 260 mm plies was cut from the fabric roll. These fabric plies were laid up in a [0/90]*n* sequence and pre-compacted in a hydraulic press at an applied load of 1240 kPa. Following compaction, the stack was centered on a glass plate coated with silicone release film. Peel-ply sheeting was placed over the laminate stack, followed by a layer of nonwoven nylon mesh (Enkafusion). Spiral tubing was attached to two vacuum tubes, one attached to the vacuum pump, and one inserted in the resin container. These tubes were placed on the edges of the laminate stack, and the entire set-up was sealed under Mylar bagging film using vacuum sealant tape. A vacuum of 3 kPa (absolute) was gently drawn using a vacuum pump (Vacmobile, Auckland, New Zealand).

The laminate stack was left under vacuum for at least 1 h prior to proceeding to insure the evaporation of absorbed moisture. The epoxy resin system was mixed at a 24:100 hardener-to-resin ratio and degassed in a sealed desiccator. The resin was then drawn through the laminate stack via the resin inlet tube by the pressure differential created by the vacuum, fully wetting the stacked fabric plies. Following complete wet-out of the laminates, the vacuum was reduced to 35 kPa (absolute), a 14 kg mass was placed on top of the stack, and the part was left to cure overnight. The panel was then disconnected from the vacuum and post-cured for 3 h at 50 °C and 3 h at 80 °C as recommended for the given epoxy resin system.

For *Z*-axis inter-ply reinforcement nylon flock fibers were applied to the individual laminate plies using the University of Massachusetts Dartmouth's patented *Z*-axis reinforcement "wet-flocking" technology [8]. Reinforcement was applied to the surface of each ply individually, after which the laminate were laid up and infused with resin following the standard vacuum infusion procedure outlined above.

2.4. Double cantilever beam testing

The double cantilever beam (DCB) test was conducted using the procedure outlined in ASTM D5528. To introduce the initial crack, a thin Teflon film is inserted in the mid-plane of the laminar panel stack during the fabrication of the composite. Following curing of the composite panel, the test coupons were then cut to the desired dimensions according to ASTM D5528, and piano hinges were adhered to the ends of the delaminated "arms" using a quick-set epoxy. The final step in sample preparation was to apply measured tick marks on the sample sides by first applying correction fluid and then marking every 1 mm for the first 25 mm past the initial delamination with black ink.

3. Results and discussion

3.1. Surface treatment

Ansell and Mwaikambo [17] reported that chemical composition of jute fibers: cellulose (51–84%), hemicellulose (12–20%), lignin (5–13%), and pectin (0.2%). The purpose of alkali treatment is that (1) removing impurity and hemicellulose on the fiber surface, and (2) imparting chemical accessibility to hydroxyl group of cellulose to promote silane coupling agent reactivity. The average percentage weight loss for alkali treated jute yarns, defined as the percent change in dry-mass as a result of treatment, was found to be (11 ± 1.7)%. Moreover, FTIR analysis of the alkali treated yarns showed a loss of peaks at 1735 and 1240 cm^{-1}, which are representative of carboxylic acid and ester groups, respectively. The loss of these peaks implies that a portion of the hemicellulose was removed by this treatment, since both of these are constituents of hemicellulose. This finding supports the successful partial removal of hemicellulose and other impurities. The alkali treatment with 5% NaOH was suitable for optimal balance between preserving mechanical properties and achieving the goals [18].

FTIR analysis of the silane treated yarns showed that a similar spectral peak near 1100 cm^{-1} (1099 cm^{-1} for Z-6173 and 1040 cm^{-1} for APTES) for yarns treated with APTES and Z-6173. Peaks near 1100 cm^{-1} wavenumber indicate that Si–O–C bonds are formed, which suggests that the silane coupling agent reacted to cellulose hydroxyl groups. This implies that the agent has chemically coupled itself to the fiber structure rather than simply being physically adhered to the surface.

3.2. NFC composite panels fabricated

Square (254 × 254 mm^2) composite panels were fabricated with plain weave and unidirectional jute fabric reinforcement and with or without *Z*-axis reinforcement fibers. The thicknesses of the manufactured composites are listed in Table 1.

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