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EFFECTS OF ALKALI AND GRAPHENE OXIDE TREATMENT ON MECHANICAL PERFORMANCE OF FLAX-EPOXY COMPOSITES

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ABSTRACT

While natural fiber composites have lower environmental impact and improved lifecycle as well as cost compared to synthetic carbon and glass fiber composites, they have the drawback of lower compatibility and reduced interfacial properties with the polymer matrices, which have detrimental effects on the final composite behavior. To alleviate this problem, natural fiber composite material properties have been shown to improve by surface treatment methods such as graphene oxide (GO) treatments in past studies. This study conducts a parametric analysis on flax fibers and flax fiber composites treated with GO to determine the effect of GO treatment on fibers in the absence of the epoxy matrix. GO characterization was performed with X-ray powder diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). Flax fibers were dip-coated in GO suspensions of concentrations ranging between 0.2-1% at durations between 30 minutes and 2 hours. Tensile tests, as well as microbond tests on treated single fibers, were performed to assess the tensile properties and interfacial shear strength of the fibers. GO-treated flax/epoxy composites were then prepared at characteristic conditions identified through single fiber tests, and tested to failure on an Instron tensile tester to evaluate the degree of improvement attributable to GO interactions with the surface of the fiber. SEM was used for failure analysis to determine the mode and degree of failure, and digital image correlation was used to obtain full-field mapping of the deformation behavior of the produced composites as well as an accurate value for strain after sample breakage. Results indicated that GO treatment significantly increases the tensile strength and modulus of flax-epoxy composites compared to control samples with tensile strength increases of up to 16.6% and tensile modulus increases of up to 7%, but no significant consistent change to the tensile strength, modulus, or interfacial shear strength of individual fibers was observed. This study provides insights into the properties of GO, improving material properties of natural fiber composites through GO pre-treatment, and the mechanisms and interactions that cause GO to be effective in acting as a reinforcer for composites.

1 INTRODUCTION

Fiber reinforced composites are widely used in a variety of industrial sectors, including aerospace, automotive, recreation, and construction. A variety of synthetic and natural fibers are used in the production of fiber reinforced composites, including glass fibers, carbon fibers, and flax fibers. While the performance of synthetic fibers is higher than that of natural fibers by weight, natural fibers are significantly more environmentally sustainable, less toxic, more biodegradable, and cost efficient compared to traditional synthetic fibers. Production of raw flax has a negative global warming indicator due to CO2 sequestration by photosynthesis [1]. However, intrinsic material properties and durability of flax fibers fall behind that of synthetic counterparts, limiting broader applications in more widespread use.

Graphene oxide (GO) is a carbon-based monolayer nanosheet manufactured from graphite with high toughness and stiffness that can be used to improve the interfacial adhesion and tensile strength of fiber composites. The exceptional mechanical strength, large surface area, and chemical functionality of GO provide avenues for composite reinforcement [2]. The hardness of GO originates from its single layered structured lattice network, with high stability due to a very large number of resonance structures. The ability of GO to improve interfacial adhesion arises from a large number of hydroxide, carboxylic acid, and epoxy groups on the surface of the GO which allows it to interact with a variety of groups on the surface of reinforcing fibers. By integrating GO info flax-epoxy composites, the mechanical [3] and thermal [4] properties of biocomposites can be substantially improved, potentially expanding applications of more sustainable fibers in high performance composites.

A multitude of methods for the integration of GO in various forms within composites exist [5], including crosslinking with amines and surface chemical modifications to improve chemical interfacial interactions on the material. The majority of research conducted on GO enhanced polymers mixes GO directly into the epoxy resin itself, with observed results indicating that GO acts to limit crack propagation in epoxy matrix [6]. This study aims to investigate the effects of GO treatment on the interface between natural fibers and epoxy matrix as well as the effects of GO on fibers in the absence of epoxy, removing the effects of inhibiting crack propagation in matrix by “dip coating” single fibers in GO solution.

2 MATERIALS AND METHODS

2.1 Materials

Flax fibers used were single fibers extracted from ampliTex ™ BComp UD 280 gsm flax fiber tows, and composite plates were made on ampliTex ™ BComp 300 gsm twill 2/2 flax fiber tows. Resin for infusion was 2 part EPOKOTE™ MGS™ RIMR 035c Resin and EPIKURE™ Curing Agents RIMH 037 from Westlake Epoxy. GO was provided by Zentek, and characterized through Fourier transform infrared microscopy (FTIR), x-ray diffraction (XRD), and transmission electron microscopy (TEM). Fiber yarns were treated with concentrations of 0.2%, 0.5%, and 1% GO for durations of 30 mins, 1h, and 2h before single fibers were extracted.

2.2 Single fiber tensile testing

Single fiber tensile tests were performed in accordance with ASTM D3822 [7] with some modifications: a gauge length of 14 mm and a fiber elongation rate of 0.2 mm/min. Cyanoacetate based superglue was used to affix fibers to paper frames, and fibers diameter was measured via an optical microscope. Tensile modulus results were taken from the midpoint of the stress-strain curve, in the elastoplastic region [8].

2.3 Microbond testing

Microbond testing was performed to measure interfacial shear strength (IFSS) between treated flax fibers and epoxy microdroplets [9] with a blade micrometer as the shear blades with a gauge length of twice the fiber diameter. Epoxy resin microdroplets are cured to single fibers and pulled via Instron tensile tester until the droplet debonds for the fiber to measure the IFSS of the fiber. An average droplet size of 70 um was maintained, and droplets diameter and fiber embedded area were measured under optical microscope. Fiber length was maintained at 10 mm. IFSS was calculated as per equation (1), where *F* is the force at debond and *SA* is the total embedded fiber surface area.

$$\text{IFSS} = F * SA \quad (1)$$

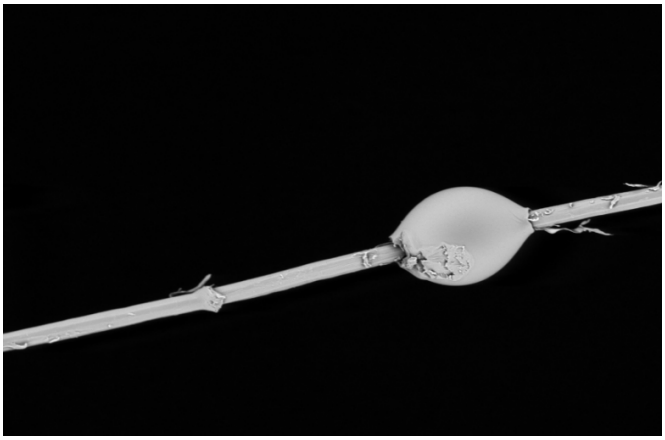

Figure 1: SEM image of a debonded microdroplet cured onto flax fiber following microbond testing.

2.4 Composite tensile testing

Composite tensile tests were performed in accordance with ASTM D3039 [10]. Sandpaper was attached to the sample along where grips contract to minimize sample slippage and lower the probability of the coupon breaking in the grip; grip pressure was controlled to around 100 psi. A gauge length of 60 mm was used, and strain measurements were performed through digital image correlation (DIC). Tensile modulus was calculated using the 0.05-0.2% strain region.

3 RESULTS AND DISCUSSION

3.1 Single fiber tensile

The greatest increase in tensile strength compared to the control value of 738.2 MPa was in the 0.5% GO 30 min treatment for an average of 979.5 MPa, followed by the 0.5% GO 2 h treatment at 898.5 MPa. Other treatments showed slight but ultimately insignificant increases in tensile strength, due to large ranges in the diameters of fibers

tested. A trend is present between the 0.5% and 1% groups in that the 30-minute treatment has the highest tensile strength, followed by a drop in the 1 h group, and recovery in the 2 h group. A likely cause is the interaction between GO and the fiber is complete within the first 30 minutes and that the fibers are negatively impacted by the GO as the duration increases, followed by moderate recovery as the GO has enough time to enter the interior of the hollow flax. The tensile modulus does not significantly change with GO treatment, which is in accordance with expectations as the tensile modulus is primarily dependent on the crystalline structure of cellulose in flax fibers.

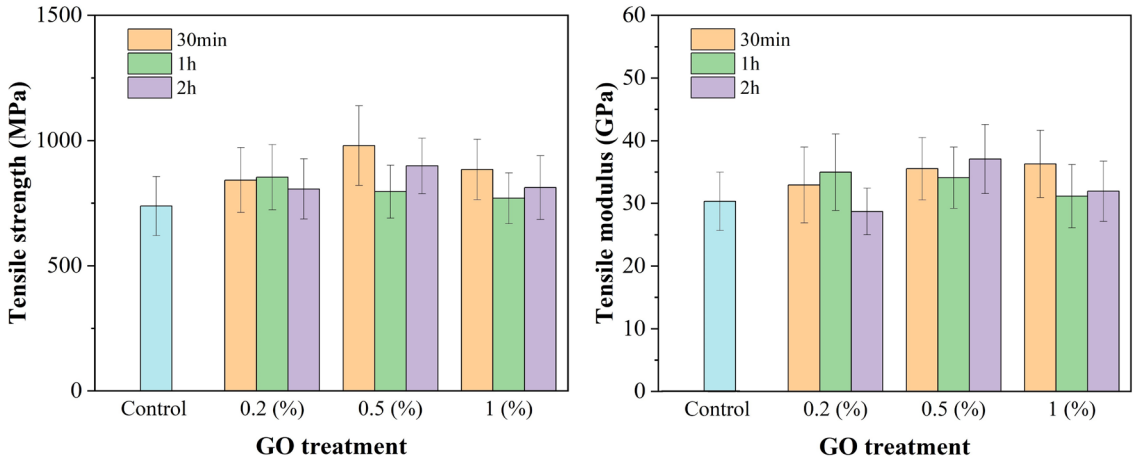

Figure 2: Graphs of tensile strength and modulus of each of the GO treatments by concentration and time.

3.2 Microbond

While the degree of variation induced by the GO treatment to IFSS was lower than expected, the trends followed expected patterns. At higher concentration treatments, the GO has a greater likelihood of aggregating to the fibers over the course of the treatment and interfering with interfacial adhesion of the resin microdroplet. In particular, the 1% GO 2 h treatment lowered the IFSS of the fiber by a significant amount. Lower concentrations of GO work best in promoting fiber IFSS, with the best result being that of the 0.5% 2 h group with an IFSS of 26.4 MPa compared to the control value of 21.3 MPa.

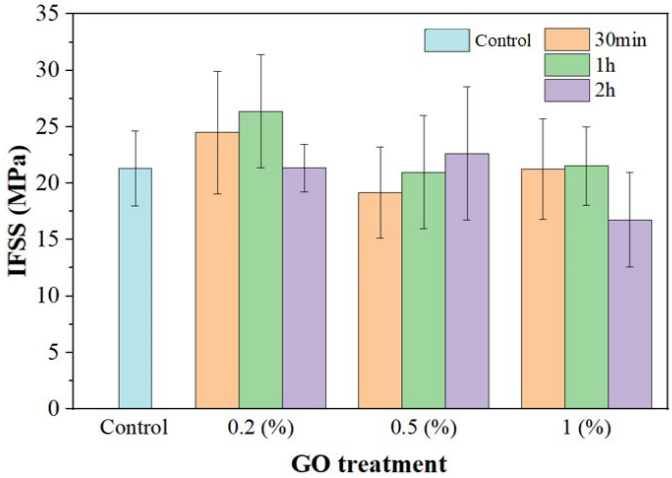

Figure 3: Graph of IFSS obtained through microbond testing of each of the GO treatments by concentration and time.

3.3 Composite coupon tensile

Tensile testing on coupons showed significant improvements in tensile strength and an increase in the modulus of characteristic samples chosen from the treatments compared to the control, from an average stress at break increase from 76.9 MPa as a control sample to 82.6 MPa for the 0.2% GO 2 h treatment and 89.7 MPa for the 1% GO 2 h treatment. Tensile modulus values likewise increased from 7.07 GPa in the control to 7.56 GPa in the 0.2% GO 2 h treatment and 7.47 in the 1% GO 2 h treatment. The approximately equal tensile modulus between the 0.2% and 1% samples combined with an increase of tensile strength by about 7 MPa indicate that the increased properties are likely due to interactions between the GO and the epoxy matrix rather than an increase in interfacial interactions between fiber and GO. The GO particles reduce matrix crack propagation and affect the overall composite rather than just fibers, which causes the modulus to increase while tensile strength remains relative to GO concentration. While the increased tensile strength between the 0.2% and 1% samples suggests that the degree of improvement is concentration dependent, GO concentration past a certain value can interfere with epoxy infusion and cross-linking, resulting a weaker composite overall.

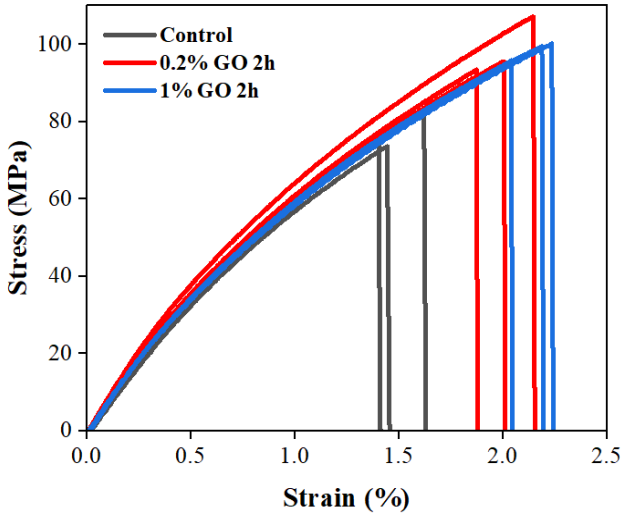

Figure 3: Stress-strain curves of control, 0.2% GO 2 treated, and 1% GO 2 h treated samples.

4 CONCLUSIONS

This study has helped shed light on the mechanisms by which GO treatment improves composite material properties. The relatively low changes to single fiber tensile strength and interfacial shear strength coupled with the more significant increases in composite strength and modulus suggest that the increase in mechanical properties derived from GO treatment does not primarily stem from fiber property improvements from GO coating or interfacial interactions between GO and fiber. GO interfaces with epoxy during curing, improving stress transfer and acting as an inhibitor for crack propagation due to its excellent strength and large surface area to volume ratio. Improved strength and increased modulus suggest that natural flax fibers composites can be effectively reinforced via GO treatment, presenting a promising strategy of developing high performance, sustainable materials, opening up new possibilities for using flax fiber in composites that demand higher performance, such as the automotive and

aerospace industries. Future research should focus on optimizing GO content, GO interactions with epoxy when mixed directly into the resin, and the environmental impact and disposal of GO in composites.

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