AN EXPERIMENTAL STUDY ON CRACK PROPAGATION IN GREEN COMPOSITES MADE FROM CELLULOSE NANOFIBERS AND EPOXY

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ABSTRACT

The current examples of natural-fiber based polymer composites, because of their biobased and biodegradable properties, offer an alternative as a 'green' material available for automotive and other engineering applications. However such composites suffer from low strength and fatigue properties compared to the carbon- or glass-fiber based polymer composites. Cellulose nanofibers (CNFs), a new type of nanofibers made entirely from cellulose molecules, have very high mechanical properties compared to other natural fibers, even approaching that of inorganic reinforcing fibers. The composites made from cellulose nanofiber films are likely to have very good strength compared to other kinds of biocomposites. However such an assertion about the mechanical properties of composites made from CNF films is yet to be investigated through experiments.

This paper presents the results from a crack-propagation test conducted using a double cantilever beam made using two types of resins and reinforcements. The resins used were a traditional epoxy and a bio-based epoxy. For reinforcement, cellulose nanofiber (CNF) films were used. The samples were tested against a reference case made from fiberglass to study and compare crack propagation patterns in the composites. Various crack resistance characteristics are observed in the two systems. The study shows the need to investigate methods for improving the fracture toughness of polymer composites fabricated using CNF films.

1. INTRODUCTION

In recent years, demand for biodegradable, plant-based, composite, also called 'green composites' or 'biocomposites', is increasing in various sectors (especially the automotive sector) of industry [Masoodi and Pillai, 2011]. Various kinds of natural fibers have been used in green composites; such reinforcements can be classified into two categories of plant and animal fibers. Plant fibers may be produced from different parts of the plants, such as seed, leave, skin, bast, and fruit. Examples of such fibers are cotton, jute, kenaf, coir, and flax. Examples of animal fibers include silk, sinew, wool, catgut, angora, mohair and alpaca. Natural fibers have lower mechanical properties as compared to carbon and glass fibers. As a result, the current breed of

biocomposites suffers from the twin drawbacks of lower strength and fatigue properties compared with the carbon- or glass-fiber based polymer composites.

Cellulose nanofibers (CNFs), a new type of nanofibers made purely of cellulose molecules, have very good mechanical property compared to other natural fibers, and even carbon or glass fibers [Saito et al., 2009; Henriksson et al., 2008]. Recent studies showed that the films or "nanopaper" of CNFs are the strongest man-made, cellulose-based materials [Lindström and Ankerfors, 2009]. However, the full reinforcing potential of these materials has yet to be realized partly because of issues related to manufacturing processes. Recently, cellulose nanofibers have begun receiving serious consideration as potential reinforcement materials. The big problem with using CNF fibers was the energy requirements for breaking down cellulose fibers in nanofibers were prohibitively high until recently [Siró and Plackett, 2010]. Recent advances in chemical and mechanical technologies have drastically reduced the energy requirements for producing cellulose nanofibers [Lindström and Ankerfors, 2009].

Although the composites made from CNF films are likely to have better mechanical properties compared with other kinds of natural-fiber based composites, several mechanical properties of the former have still not been measured. One such property is the mode-I fracture toughness test which reveal crack propagation patterns in composites made from CNF films. This study presents the results from a test on tearing apart a double cantilever beam made using two types of resins and reinforcements. The two types of thermosetting resins used were a traditional epoxy resin and a bio-based epoxy resin. For reinforcement, the CNF films and fiber-glass mats were used. The samples were tested and the results were compared to yield a comparison of crack propagation patterns in these different composites.

2. EXPERIMENTATION

2.1 Test Materials

The CNF films used in this study were produced at the USDA Forest Product Laboratory in Madison, Wisconsin. Figure 1 shows one of the clear films made out of cellulose nanofibers. To make composites, we used two different epoxies as thermosetting resins: a bio-based epoxy and a traditional epoxy. The traditional epoxy that was used here was System 2000 Epoxy Resin from Fibre Glast Development Corp. The bio-based epoxy was Super Sap 100/1000, which is a two-part liquid epoxy system designed by Entropy Bio-Resins Co. Unlike traditional epoxies that are composed primarily of petroleum-based materials, Super Sap 100/1000 is an epoxy resin that is claimed to be up to 75% bio-based. In this paper, 'Epoxy' refers to 'System 2000 Epoxy Resin', and 'Bioepoxy' refers to Super Sap 100/1000 bio-based epoxy rein.

Figure 1: Transparent CNF film produced at the USDA Forest Product Laboratory

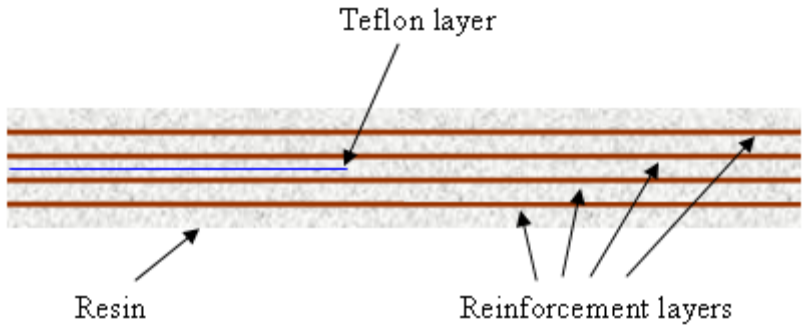
2.2 Test Method

We made samples of composite materials by using the cellulose nanofiber (CNF) films and the two epoxies. Since no standard for the fracture toughness and crack propagations test applicable to CNF composites was available, so we adapted the ASTM D5528-01 for our experiments. The specimens' size was 120x20x3 mm. Four different materials made for this experiment are mentioned in Table 1.

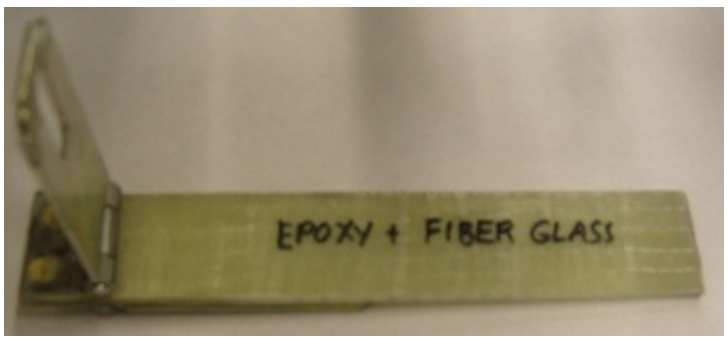
Table 1: Different composites made for experiments.

Sample #	Sample description	Layering
1	Fiber glass and epoxy	Four layers of woven fibermats
2	Fiber glass and bioepoxy	Four layers of woven fibermats
3	CNF and epoxy	Four layers of CNF One layer of woven fibermats on each side
4	CNF and bioepoxy	Four layers of CNF One layer of woven fibermats on each side

The test specimens were made using the hand lay-up technique for making composites. Figure 2a shows the layering of reinforcements in the specimens. For the fiberglass specimens, we used four layers of an E glass woven fabric as reinforcements and epoxy as the matrix. As shown, a thin layer of Teflon film (of length 50 mm) was placed at one side within the middle layer. (Note that the Teflon layer provides the crack starting point for the tests.) Note also that for CNF composites, we added one extra layer of fiberglass on each side of the specimens to increase the thickness and also to prevent rupture of specimen during the cutting of samples to the desired dimensions. Two piano hinges were attached on the outer sides of the specimen to enable gripping of the specimens by the test machine (See photo 2b). The load was applied using an electromechanical test system (see Figure 3). The experiments were performed in a displacement control mode at a rate of 0.5 cm/min. During the tests, the cross-head displacement, load and crack location were recorded.



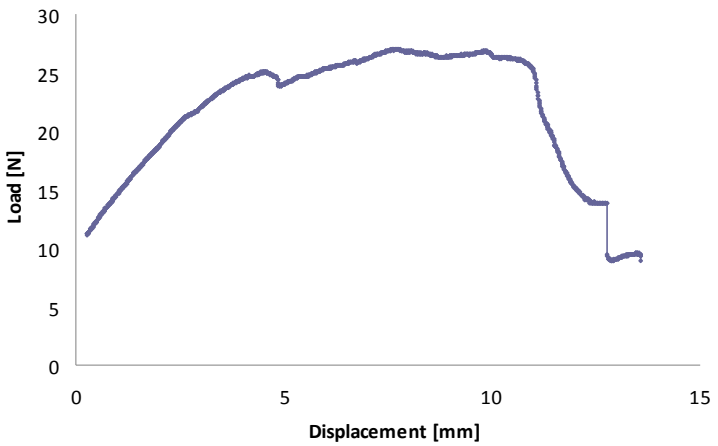
a) A schematic of the longitudinal cross-section of a specimen showing the layering of reinforcements. Note that for the CNF based composites, one extra layer of fiberglass was added on each side.



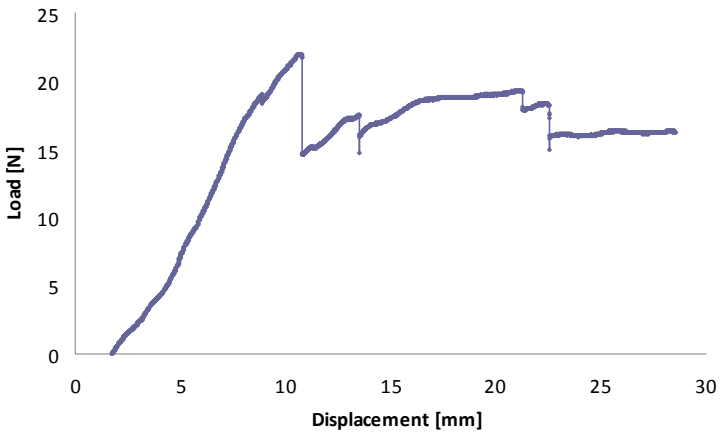
b) A photo of a specimen made from fiberglass and epoxy after the test.

Figure 2: The specimen preparation for the test.

Figure 3: Mode-I Fracture Test for a CNF based composites specimen using an electromechanical test system.

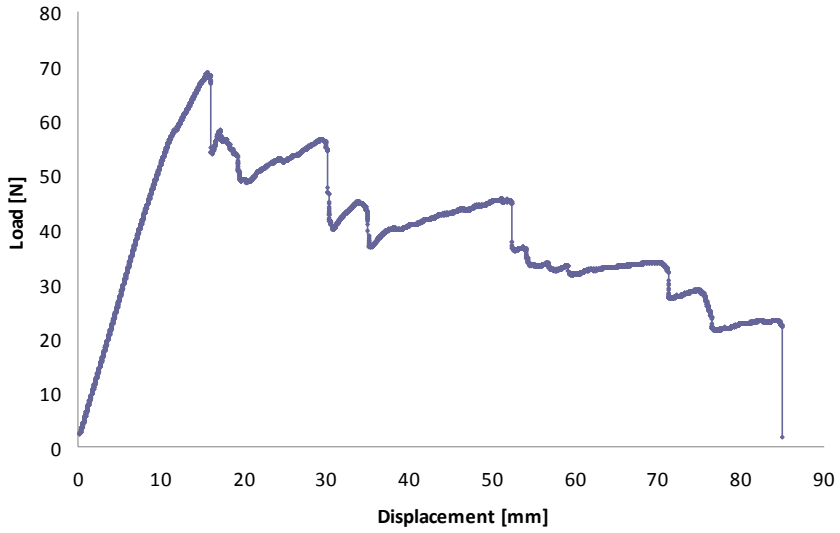


a) Standard Epoxy

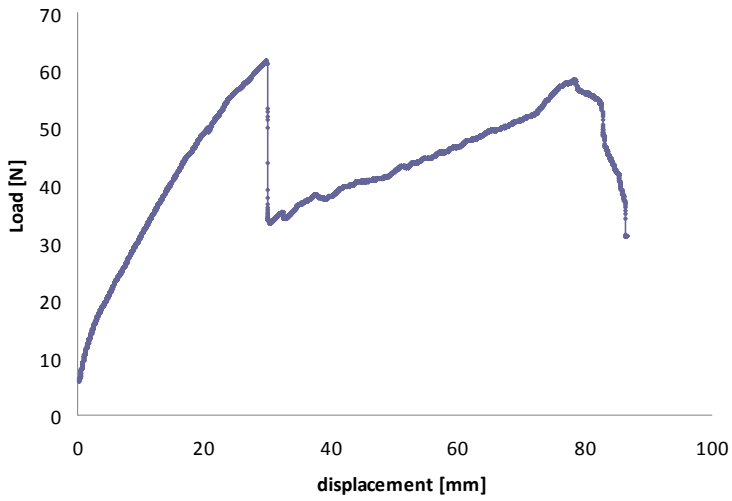


b) Bio-based Epoxy

Figure 4: Fracture toughness test results for the composite sample made from CNF



a) Standard Epoxy



b) Bio-based Epoxy

Figure 5: Fracture toughness tests with composites made from fiberglass

3. TEST RESULTS

Figure 4 shows the relatively similar load displacement behavior when the bioepoxy and epoxy resins are used with CNF. The load displacement behavior seen in Figure 5 shows a higher crack resistance value in the fiberglass composite samples compared to the values obtained when the CNF films are used. In other words, these preliminary results indicate that the interlaminar strength in the fiberglass samples is almost double the strength for the CNF samples[1]. This is explained by the increased fiber bridging involved in the manufacturing process that increases

[1] However in order to establish this fact conclusively, several more tests are needed. We were unable to conduct that many tests due to a very limited number of CNF sheets available for the tests.

the crack resistance behavior. There is also no resin interlock behavior present in the CNF films because of the smoothness and continuity of the CNF layers. Figure 6 shows a double cantilever beam test on a CNC composite manufactured with CNF films. Note the crack jumps observed in the tests after the propagation behavior. In bioresins, the crack propagation is more characterized by crack jumps compared to a more gradual crack growth seen when the standard resin is used. Crack bridging was not observed in the CNC specimen and no similar crack retardation features were found.

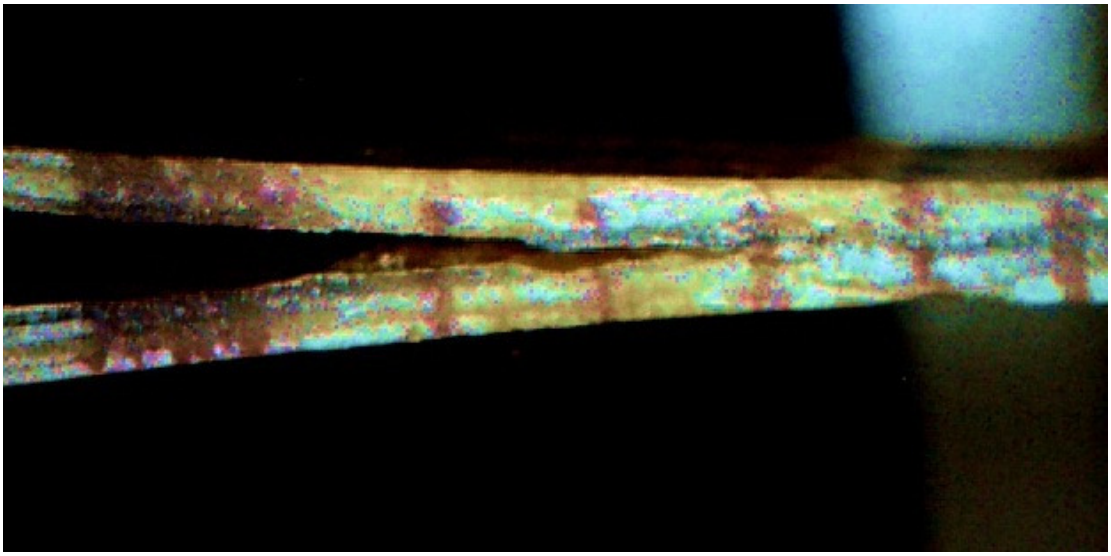


Figure 6 Crack propagation behavior in a CNF composites sample showing minimal fiber bridging

4. CONCLUSIONS

This study presents the results from the Mode-I fracture tests conducted with double cantilever beam specimens made using two types of resins and reinforcements. The thermosetting resins used were a traditional epoxy and a bio-based epoxy. The wet layup manufacturing process was employed to manufacture the double cantilever beam specimens. The biobased epoxy shows similar performance characteristics for fracture toughness when compared to the standard epoxy and does not show reductions at the room temperature test conditions.

Reduced crack resistance is observed when using CNF films compared to those when fiberglass alone is used. This is due to the lack of fiber bridging in the CNC composite and the absence of significant mechanical interlock. Incorporation of CNF films as a reinforcement layer will require methods to improve the fracture toughness. This will be investigated in future studies by perforating the layers or by inducing ripple patterns in the CNF films.

5. REFERENCES

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