CARBON PLAIN WEAVE TEXTILE REINFORCED EPOXY MODIFIED WITH CELLULOSE NANO FIBERS: EFFECT OF CNF LENGTH

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Keywords: Cellulose Nano Fibers, Carbon plain weave textile composites; Mechanical properties

ABSTRACT

The present investigation gives a contribution on understanding the effects of hybrid epoxy resins, enhanced with cellulose nano fibers (CNF), on the mechanical performance of carbon fibers plain weave textile composites. The experimental investigation considered two lengths of CNF for the same weight content (0.3%) in the epoxy resin. The effects on the mechanical performance, compared to the composite with pure resin counterpart, were assessed with: quasi-static tensile and bending loading. Moreover, bending and tensile cyclic loading, for some stress levels, were supposed to detect advantages of the hybrid resin in extending the fatigue life of the textile composite. The results show slight variations of the quasi-static properties and a considerable extension of the fatigue life mainly when the longer CNF are considered.

1 INTRODUCTION

Several research efforts were dedicated for the development of hybrid resins to get improved mechanical properties of fiber reinforced polymer (FRP) composites. Modified thermosetting matrix resins were created by the dispersion of a second phase normally consisting of nano- or micro- sized fillers. The more investigated nano fillers to have tougher polymer matrices included different materials such as silica particle, nano clay [1] and carbon nano tube [2]. When a small amount of those fillers is incorporated into the matrix, the initiation and propagation of matrix crack in FRP can be delayed significantly [3].

The identification of nano-sized cellulose micro-fibrils, named cellulose nano fibers (CNF) [4], increased the range of hybrid nano-enhanced composite materials [5]. Cellulose is the most abundant natural homopolymer and one of the most promising renewable and environmentally friendly resources.

CNFs are obtained through a homogenization process, at high shear, producing fibrils with a diameter range of 10-100 nm and a web-like structure. Plant derived cellulose were adopted as either all-cellulose composites [6], or biodegradable natural polymer matrix [7].

The positive effect on some quasi-static mechanical properties and fracture toughness of micro-fibrillated cellulose-based epoxy reinforced carbon textile was demonstrated in [8]. Moreover, the effectiveness of epoxy resin modified with micro fibrillated cellulose on the improvement of the fatigue life and the impact strength of carbon textile composite was measured in [9] and [10].

The present experimental investigation intended to give a contribution on understanding the effects of hybrid epoxy resins, enhanced with cellulose nano fibers, on the mechanical performance of carbon fibers plain weave textile composites. The main scope was to study the influence of the lengths of CNFs on some mechanical properties of the composite.

The effects of the same content of CNFs of two lengths was assessed comparing to the results of the composite with pure epoxy resin. Quasi-static and cyclic loading conditions were considered. Of both, tensile and bending tests were conducted to detect advantages of the hybrid resin on the mechanical behavior of the textile composite.

The results show, for quasi-static loading, an improvement of the average bending strength in the range 4-5% for both CNFs, while the tensile properties did not have considerable variations for both modifications of the matrix, with a slight reduction of the strength recorded with long CNFs.

The main advantage of the CNF hybrid epoxy resin was observed for the cyclic loadings response. The effect of both CNFs was a considerable extension of the fatigue life for both tensile and bending loading conditions, with the best performance recorded for the composite with the longer CNFs. The reason for such enhancement could be connected to the local cracks deviation around the cellulose nano fibers and to the fiber bridging effect of the CNF which delayed the local cracks propagation.

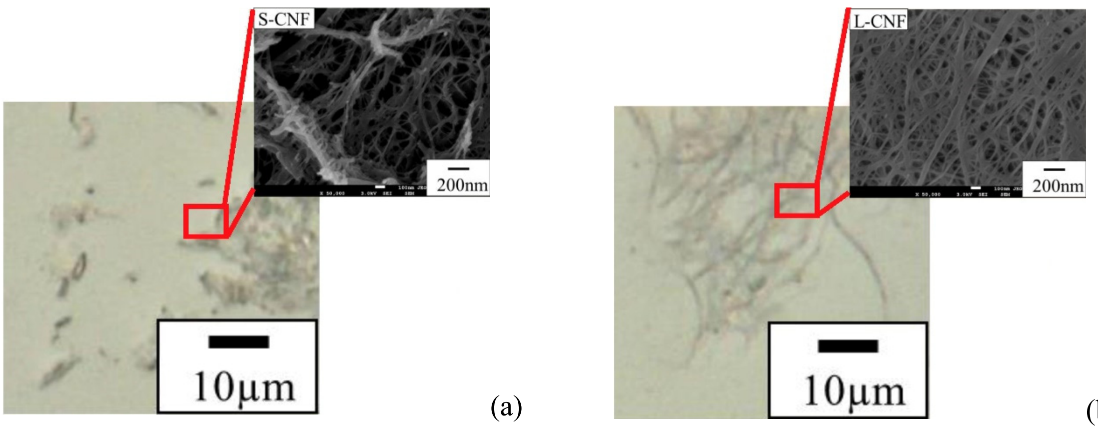
2 MATERIALS AND MANUFACTURING

The considered composite material is an epoxy matrix reinforced with a carbon fabric (Pyrofil TR-3110-MS of areal density 200 g/m^2, and yarns Pyrofil TR30S3L Mitsubishi Rayon Co. Ltd., Japan). The epoxy resin (JER 828 with curing agent JER cure-113, Mitsubishi Chemical Corporation, Japan) properties were enhanced with two cellulose nano fibers (CNF) (Sugino Machine Limited Corporation, Japan) of nominal diameter 20 nm and two lengths: FMa-10002, length about 6μm (Figure 1a, named CNF-Short); IMa-10002, length about 22 μm (Figure 1b, named CNF-Long).

The experimental investigation considers the two lengths of CNF for the same weight content (0.3%) in the epoxy resin, assuming the results of a previous investigations [10].

The water slurry containing CNF nano fibers is firstly treated by solvent exchange with pure ethanol to remove water. CNF containing ethanol is stirred for 30 min at 5000 rpm and then filtered by Büchner funnel to remove water. The filtered CNFs is stirred again with additional ethanol, for the same time and speed, and then mixed with the proper quantity of epoxy resin. The mix is stirred for 30 minutes in a high speed homogenizer (10,000 rpm). In this phase, ethanol is used to decrease the epoxy viscosity and to have uniformly dispersed CNF. The ethanol is removed maintaining the mixture in an electric oven at 80 °C for 7 days.

Eight layers of fabric were stacked and laminated by hand lay-up impregnation. The mold was cured in a hot press at 80°C for 1 h, and then 150°C for 3 h, keeping a pressure of 2 MPa. The resulting laminates had thickness of 1.97 ± 0.06 mm and fiber volume fraction of 44.8 ± 1.1%.



(a) (b)

Figure 1: (a) CNF-Short; (b) CNF-Long.

3 EXPERIMENTAL PROGRAM AND FEATURES

The mechanical performance of CNF modified epoxy resin reinforced with carbon textile was investigated with an experimental campaign encompassing quasi-static and cyclic loadings. In particular, both loading conditions were considered for bending and tension.

Quasi-static three-point bending tests were performed according to the standard JIS K 7074, using a universal testing machine. The prismatic specimens allow using a supports distance of 80 mm. The test speed was set to 1 mm/min

The standard JIS K 7161 was considered for the quasi-static tensile tests, using the same universal testing machine mentioned above for bending tests. The specimen geometry had total length of 200 mm, gage length of 100 mm and width of 25 mm. Aluminum tabs were glued at both ends in the gripping zones. The longitudinal strain was measured with strain gages (10 mm gage length) or by the digital image correlation method. For the latter, the central part of some specimens was speckled with white and black acrylic paints for a length of about 30 mm. The cross head speed was set to 1 mm/min

The same prismatic geometry and supports span were adopted for bending fatigue. Cyclic bending was supposed with constant stress amplitude, sinusoidal wave-form loading and ratio R = 0.1 (ratio of the minimum to the maximum stress in the cycle). The frequency was set to 2 Hz. Three different maximum stress levels were considered in the range 75-85% of the bending quasi-static strength. Four specimens were tested for each stress level up to failure or run out after 100 thousand cycles.

For tensile fatigue, the specimen geometry was as for quasi-static tensile tests. Tests were performed under constant stress amplitude, sinusoidal wave-form tensile-tensile loading and assuming the ratio R = 0.1 (standard JIS K 7083). The frequency was set to 5 Hz. Three different maximum stress levels were considered in the range 65-80% of the ultimate static tensile stress. At least three specimens were tested for each stress level up to failure or run out after 1 million cycles.

4 RESULTS AND COMPARISONS

The effect of the two CNFs on the mechanical properties is detailed distinguishing the quasi-static and the fatigue behavior for both loading conditions, bending and tension.

4.1 Quasi-static response

The bending and tensile mechanical properties are highlighted comparing the elastic modulus (initial secant) and the strength. Figure 2 presents the average value of the bending mechanical features for the considered CNF combinations, compared to the composite with pure epoxy resin (content 0%).

The modified resin does not provide relevant variation of the bending material stiffness. While the average bending strength has an improvement ranging 4-5% for both CNFs (Figure 2), even though results for the three materials can be considered in the same experimental scatter band.

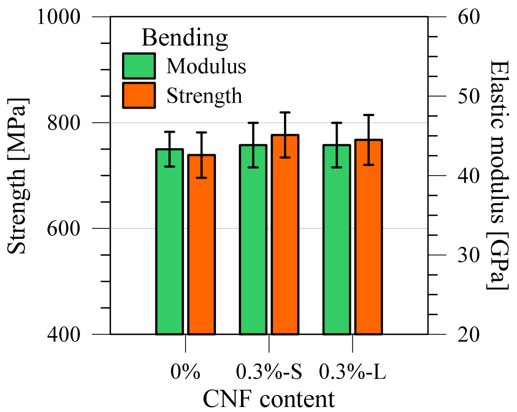


Figure 2: Comparison of bending elastic modulus and strength. Average and standard deviation of five specimens.

Similar to the bending results, the tensile modulus and strength do not have considerable variations for both modifications of the matrix, with a slight reduction of the strength recorded with long CNF (Figure 3). Slight improvement of tensile strength was recorded in [10] for a similar textile composite with different CNF embedded in epoxy matrix.

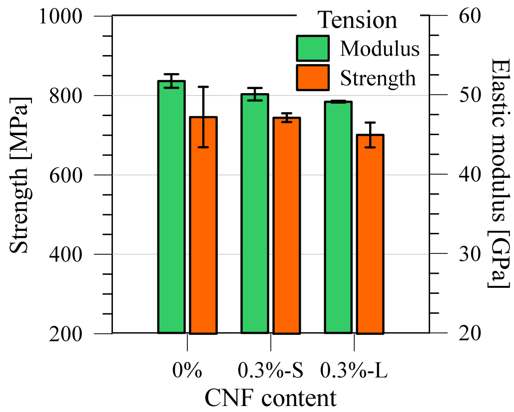


Figure 3: Comparison of quasi-static tensile elastic modulus and strength. Average and standard deviation of four specimens.

4.2 Bending and tensile-tensile fatigue

The fatigue life diagram of bending cyclic loading highlights considerable extension of the endurance varying from short to long CFN reinforcement (Figure 4). The latter increased the fatigue life of about twenty times, while the short of five times compared to the unreinforced material (0%).

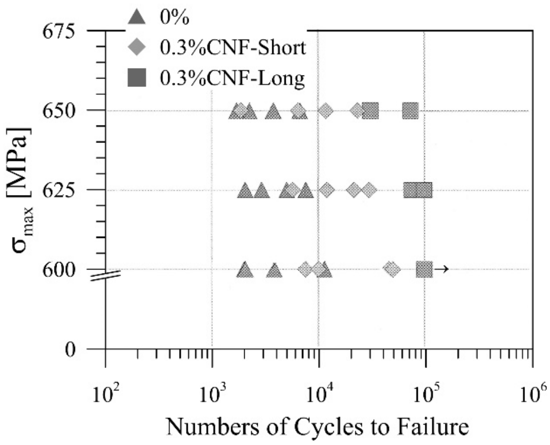


Figure 4: Fatigue bending: maximum stress in the cycle vs. number of cycles to failure ('→' means no failure after 100 thousand cycles).

The tensile-tensile cyclic tests performed in the considered stress range (three stress levels) enabled depicting the fatigue life diagram for each of the considered composites (Figure 5a). The comparison is here depicted considering the number of cycles to failure versus the maximum stresses in the cycle σ_{max} normalized to the tensile strength σ_u.

Fitting of the experimental data enables predicting the fatigue life corresponding to other stress levels, which were not directly determined by testing. The adopted semi-logarithmic ($\sigma_{max} = k \log N + a$) includes the number of cycle to failure (N), and two parameters (k and a) to be defined by the least squares method. The fittings of the experimental results (run-outs are not included) have coefficient of determination (R^2) higher than 0.97, highlighting their reliability. The comparison of the slope of the linear fitting shows the fast reduction of the fatigue life for the composite with pure epoxy resin (content 0% in Figure 5b) and the slowest decrease with the load level is for the composite including the long CNF (Figure 5b).

Similar understand is comparing the average number of cycles to failure for the considered load levels. The diagram in Figure 6 clearly shows the longer fatigue life of the composite enhanced with the long CNF for the highest load level (σ_{max} 80% of σ_u). This is almost forty times higher than the unreinforced material, while the composite with short CNF had an increase of almost 50%.

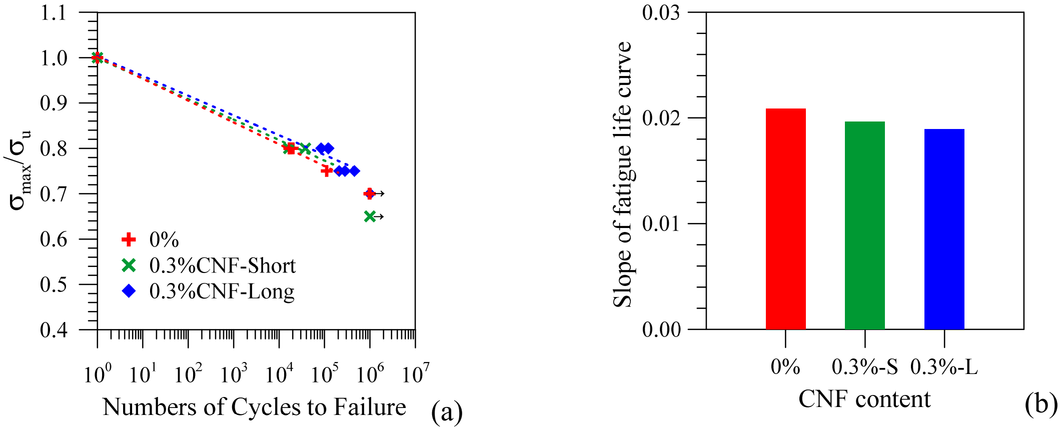


Figure 5: Fatigue tension: (a) Normalized maximum stress in the cycle vs. number of cycles to failure and semi-logarithm fitting for each composite ('→' means no failure after 1 million cycles). (b) Slope of the linear fitting.

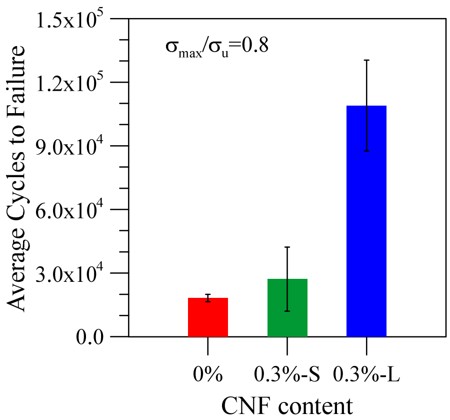
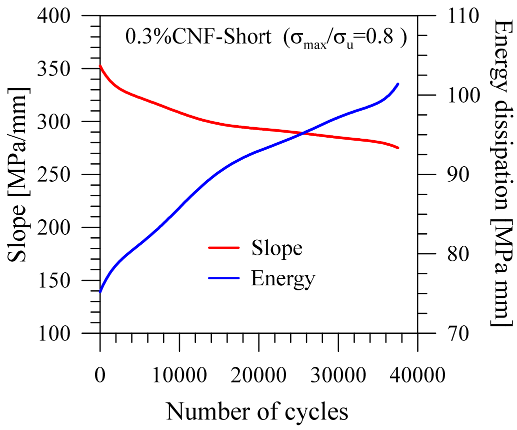


Figure 6: Average fatigue life for maximum stress level 80% of σ_u. Bars indicate standard deviation of three tests.

The fatigue damage development in composite materials may be described at the macroscopic scale by empirical metrics [11]. One of the main adopted damage metric is the stiffness degradation [12]. Here two damage metrics were adopted: the slope of the segment passing through the points of maximum and minimum of the stress-displacement cycle curve (cycle slope), and the energy dissipated in the cyclic loading, i.e. the area contained in the stress-displacement cycle curve. It should be noted that the cycle slope does not coincide with the stiffness of the material, it has not been separated from the compliance of the testing machine. But, as shown in [10], stiffness and cycle slope evolution provide analogous qualitative information on the damage imparted during cyclic loading.

The evolution of the slope and energy of all cycles for the load level σ_{max} / σ_u = 80% is depicted in Figure 7 for two specimens of materials containing the two CNFs. Despite composite with longer CNF had a fatigue life more than double of the one with short CNF, similar trend of the two metrics was observed for both specimens. Diagrams in Figure 7 contain three stage curves [11]. Initial stage with a rapid decrease of cycle slope (a rapid increase of the cycle dissipation) demonstrates a fast development of the damage in almost 10% of the fatigue life. The second stage has a slower reduction of the slope (a slower increase of the dissipation), meaning a slowest diffusion of damage in the composite up to almost 90% of the fatigue life. The third and final stage indicates a fast decrease of the cycle slope (a fast increase of the dissipation) of both materials as consequence of the rapid spread of the damage leading to failure (Figure 7). The main difference of the composites with the two CNF lengths is the rate of reduction and increase of the cycle slope (stiffness) and energy dissipation, respectively, in the three stages. Compared to the composite with the short CNF, the one with long CNF had more considerable drop of the cycle slope in the first phase, showing the initial consistent loss of material stiffness. In the second phase, the long CNF composite had a very slow diffusion of the damage and, as consequence, a very slow degradation of the cycle slope. On the contrary, the composite with short CNF had a less consistent reduction of the stiffness in the first stage, while, in the second one, a continuous and rapid propagation of the damage produced a higher rate of decrease of the cycle slope (increase of the dissipation). The latter must be judged also considering the diffusion of the damage up to failure during the fatigue life that was almost half of that of the composite with long CNF.



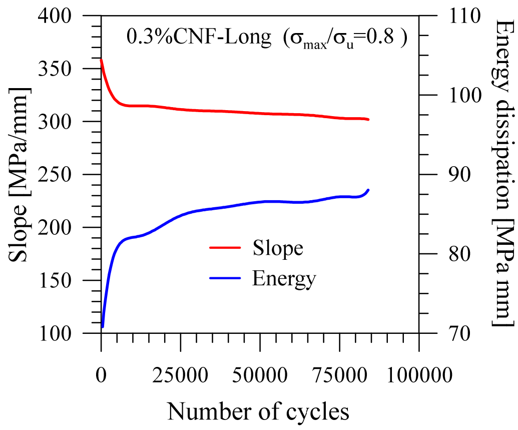


Figure 7: Fatigue tests stress level 80% of σ_u. Cycle slope and energy dissipated vs. number of cycles: (a) 0.3% CNF-Short, (b) 0.3% CNF-Long.

5 CONCLUSIONS

The effects of cellulose nano fibers (CNF) embedded in the epoxy resin of a carbon textile reinforced composite was experimentally investigated. Two lengths of CNF for the same weight content (0.3%) in the epoxy resin were considered.

The effects on the mechanical performance, compared to the composite with pure resin counterpart, were assessed with quasi-static and cyclic tensile and bending loadings.

The quasi-static measurements showed slight variations of the properties, with many results in the same experimental scatter band.

The evidence of the enhancement using the hybrid resin was connected to the fatigue response of the textile composites. The effect of both CNFs was the extension of the fatigue life for both tensile and bending loading conditions. The bending fatigue life was almost twenty times and five times, compared to the unreinforced material, for the long and short CNF, respectively.

The tensile-tensile fatigue tests provided more information related to the effect of the CNF on the diffusion of the damage. The adopted damage metrics highlighted a slowest reduction of the material stiffness during cyclic loading, meaning a slowest propagation of the damage. This led to the considerable extension of the fatigue life compared to the composite with pure epoxy resin. As mentioned in [10], the reason for such enhancement could be connected to the observed extensive 'plastic' deformation of the matrix. This was consequence of the local cracks deviation around the cellulose nano fibers and of the fiber bridging effect of the CNF which delayed the local cracks propagation. The latter requires a more in-depth future investigation with micro-scale observations.

ACKNOWLEDGEMENTS

The research was funded by JSPS KAKENHI Grant Number 26289012.

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