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Quasi-static mechanical response and corresponding analytical model of laminates incorporating with nanoweb interlayers

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ABSTRACT

Composite nanofibers of multiwalled carbon nanotubes and polyurethane (CNTs-PU) were prepared and collected on the surfaces of T700/epoxy prepregs, acted as nanoweb interlayers, and then formed unidirectional laminates. Quasi-static mechanical responses were examined through tensile and three-point flexure tests to evaluate the reinforcing effect of CNTs-PU nanoweb interlayers on fiber-dominated mechanical properties of the resulting laminates. Results showed that tensile strength and modulus of the laminates integrated with nanowebs increased up to 4.5–8.4% and 3.1–4.7%, respectively. Also, improvements in flexural strength and modulus were correspondingly up to 12.3–15.6% and 0.6–2.7%. Improvement of nanoweb interlayers on fiber-dominated performance of laminates has been achieved successfully. Furthermore, an analytical model based on the modified classical laminated theory and an inverse approach was deduced to investigate the reinforced mechanism. The proposed model could well explain the mechanical responses and the reinforced mechanism of the laminates integrated with nanowebs.

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1. Introduction

Due to high specific strength and stiffness, superior manufacturability, as well as excellent corrosion resistance and fatigue tolerance, fiber reinforced polymer (FRP) composites have a wide range of applications in aeronautical and astronautical structures, ground vehicles, and sports utilities [1,2]. Nevertheless, polymeric matrix especially epoxy dominated mechanical properties (i.e., shear, transverse and interlaminar properties) are always weaker than those dominated by fibers (i.e., longitudinal properties). This feature somewhat limits possible applications of composites as high performance structural materials [3]. Therefore, toughening brittle epoxy resins using additional phases has always been an important practice for researchers [3–5]. Nanoscale additional phases in composites are of interest due to their potentials for significantly improving the composite material properties. To date, numerous attempts have been carried out to improve matrix dominated properties of FRP composites by dispersing nanoparticles into epoxies, such as clay nanoplatelets [6,7], carbon nanofibers [8,9], and carbon nanotubes [10–13]. Significant improvement has been achieved by using those nano-phases [3–13], however, agglomeration is still one of the major drawbacks in mixing particles into polymer matrices which results limited exploring the super performance of nanoparticles [14].

Another kind of nano-materials, nanowebs formed by electrospinning, has recently been explored for their reinforcing ability in composites. By forming a network of nanofibers, electrospinning secures the uniform planar dispersion of nanofibers that can be further preserved when used in polymeric matrix composites [15]. This innovative idea was firstly explored by Kim and Reneker in 1999 [16], as well as proposed by Bergshoef et al. [17] in the same year. Inspired by this idea, Romo-Uribe et al. [18], Neppall et al. [19], Lee et al. [20], Ozden et al. [21], Han et al. [22], Tang and Liu [23], etc., explored various nanowebs as bulk reinforcements of polymer matrix for applications in reinforcing, functional, and biomedical fields. Meanwhile, some researchers took concerning nanowebs as reinforcements of composite laminates. In line with the works of Dzenis and Reneker [24], Sihn et al. [25], Liu et al. [14,26,27], Shivakumar et al. [28,29], and Zhang et al. [30], the influence of nanoweb interleaving on the tensile first-ply-stress [25], tensile ultimate strength [25], interlaminar fracture toughness [26–30], impact resistance [28,29], flexural properties [14] and interlaminar shear strength [14] of laminated composites

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were investigated. Both matrix-dominated and fiber-dominated mechanical properties were found to be improved to some degree by using this kind of nano-materials [24–30]. A recent paper by Bilge et al. [31] investigated the effects of P(St-co-GMA) copolymer and P(St-co-GMA)/MWCNT fibrous mats as interlayer reinforcing materials on the flexural properties, mode II strain energy release rate, charpy-impact and transverse tensile strength, and increases up to 17%, 70%, 20%, and 27% were recorded, respectively. Another recent article by Palazzetti et al. [32] revealed the enhancement of energy absorbing capability of Nylon 6,6 nanowebs as the laminate mid-plane interleaving on the mode I and mode II fracture strengths. Results from these reports have consistently demonstrated that the strong interfacial bonding has been crucial to benefit from the unique properties of nanofibers for composite reinforcement [15–32].

Besides experimental studies, some attempts have also been carried out from analytical and numerical points of view to investigate the reinforced mechanism of laminates with nanofiber interleaving. Sihn et al. [33] tried to analyze the delaminations in mode I, mode II and the mixed mode failures for nano-interleaved laminates based on a parametric numerical procedure. To predict the delamination failure, interface elements based on a bilinear softening constitutive law were employed in their studies. By combining micrograph observations with the stress–strain behaviors predicted by the numerical analyses, they concluded that nano-interlayers can suppress the stress concentration, the propagation of the microcrack and the delamination damage. While, Sihn et al. [25] used the Weibull statistical theory to analyze the first ply failure (FPF) strength of laminates with the nano-interlayers, and their predictions agreed well with the experimental data. The work of Chamis [34] applied meso mechanics to simulate the enhanced matrices reinforced by nanofibers with different volume ratios. Results showed that matrices reinforced with nanofiber volume ratios of less than 0.1 had useful properties and could be used for conventional fiber reinforced composites. In the analysis conducted by Arai et al. [35], a two-dimensional finite element analysis and a modified Tsai-Wu criterion were introduced to evaluate the critical curve of the interlaminar fracture of laminates with carbon nanofiber interlayer.

Up to now, only limited reports about nanowebs reinforced laminates and the corresponding analytical/numerical models can be found from the published literatures. More efforts, especially theoretical and numerical studies, are needed to explore the toughening mechanism of the nanofibers as laminate interleaving so as to fulfill this relatively young research field. Unlike most available papers which paid more attention to the matrix-dominated properties of the nanoweb interleaved laminates, this research contributed to investigate the reinforced capability and corresponding mechanism of multi-walled carbon nanotubes and polyurethane (CNTs-PU) composite nanofiber webs as laminate interleaving on the fiber-dominated properties. The corresponding results will give some supplementary information for the comprehensive understanding of the reinforcing ability of nanowebs on the bulk mechanical properties of laminates, and contributes to the data generation of nanowebs reinforced composite laminates.

In this study, the chosen CNTs-PU nanofibers were prepared by use of electrospinning [14], which were directly collected on surfaces of T700/epoxy prepregs to form a thin nanoweb interlayer. Then, the prepregs were stacked one by one to form a unidirectional laminate. Tensile and flexural properties were examined for the prepared laminates to evaluate the reinforced capability of CNTs-PU nanowebs. To further understand the strengthening mechanics, an analytical model based on the modified classical laminated theory and an inverse approach was proposed. The effectiveness of CNTs-PU interfaces is verified through the analysis.

2. Experimental part

2.1. Preparation and characterization

2.1.1. Preparation of nanowebs and laminates

Polyurethane (PU, BASF 1185A), which was well compatible with epoxy resins, was chosen to fabricate nanofibers in this research. The electrospinning solutions were prepared by dissolving PU 10 wt% in a mixed solvent of tetrahydrofuran (THF) and *N-N* dimethylformamide (DMF) (THF:DMF = 6:4 by weight ratio). To obtain strengthened nanofibers, multi-walled nanotubes (MWNTs, purchased from Shenzhen Nanotech Port Co., Ltd., with a carboxyl ratio of 2.3 wt%, 10–20 nm in diameter and 5–15 μm in length) were added to the solution at 1 and 5 wt% of the PU in the solution and sonicated for 2–3 h at 60 °C to ensure a homogeneous dispersion. During the electrospinning, the applied high voltage, solution flow rate, inner diameter of spinning needle and tip to ground distance were set at 23 kV, 3.5 mL/h, 0.68 mm and 13 cm, respectively. It is noted that the inner diameter of spinning needle is over 30,000 and 40 times of the diameter and length of CNT, respectively. Thus, CNTs can pass the needle without difficulty. The nanofibers were directly received on the prepreg surface forming the nanoweb interlayer with a thickness of nearly 20 μm (Fig. 1a). Meanwhile, fewer nanofibers were also eletrospun onto the prepreg surface so as to evaluate the wetting between the nanofibers and the epoxy matrix.

The T700/MTM28 epoxy prepregs (made in Advanced Composites Group) with an average thickness 0.22 mm (see Fig. 1b) were employed to fabricate the composite laminates. The weight percentage of T700 carbon fibers in the prepreg was 67%, whereas its areal density was 200 g/m^2. The epoxy in the prepreg, MTM28, is a high performance epoxy resin which provides good adhesive properties. The density of the cured MTM28 epoxy is 1.22 g/cm^3. Three types of laminates, in each of which 6 layers of the T700/epoxy prepregs were arranged unidirectionally (i.e. $[0]_6$), were prepared and investigated. The first type laminate was a base specimen with no CNTs nanowebs arranged into ply interfaces. The second and third laminates denoted two specimens incorporated with CNTs-PU nanoweb interlayers on every interface of two adjacent structural layers. The MWNTs concentrations in CNTs-PU composite nanofibers are 1 wt% and 5 wt%, respectively. The total thickness of the inserted nanowebs was $(0.020 \pm 0.006) \times 5$ mm. After stacking the plies for the intended laminates, these three plates were vacuum bagged and kept under vacuum during the cure cycle. The laminates were heated up to 120 °C at a rate of 1 °C/min, and were hold at 120 °C for 1 h. Afterwards, the laminates were cooled down to room temperature for further characterization.

2.1.2. Characterization

Microscopic views of CNTs-PU nanoweb interlayers on the surface of the prepreg, immediately received from the electrospinning,

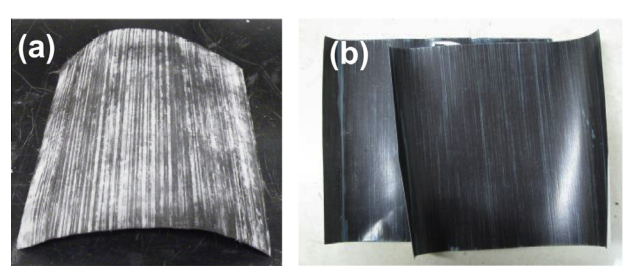

Fig. 1. Blank prepreg (a) and nanofibrous webs attached on the surface of prepreg (b).

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