Interlaminar Shear and Tensile Strengths of the Polymer Composites Based on CNT Films

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Abstract: Carbon nanotubes demonstrate excellent electrical, mechanical and thermal properties due to their unique structure. To bring this astonish properties into macroscale, CNTs were assembled into large scale film via FCCVD method. However, the obtained CNT film mechanical properties remained unsatisfactory and leaves room for further improvement. Herein, different mixtures of Epoxy and PVA polymer matrix were incorporated via vacuum assisted resin transfer molding (VARTM) technique into CNT film and successfully enhanced its mechanical properties. The obtained CNT film/Epoxy and CNT film/PVA tensile strength increased by **3.5** and **4.7** times compared to pristine CNT film tensile strength. In addition, polymer matrix compatibility with CNT film was investigated by interlaminar strength method. CNT films/epoxy composite exhibited significantly higher peeling strength (633.7 N/m) compared to those with PVA (70.8 N/m) demonstrating the good affinity of epoxy with CNT film.

Keywords: CNT film composite, PVA, Epoxy, Interlaminar shear strength, Tensile strength.

1. INTRODUCTION

Researchers have been always seeking for lightweight strong materials with low energy production, easy to mold and costless which is difficult to find in convenient materials such as metals and glasses. Carbon nanotubes showed good satisfactory to those criteria in addition to their excellent mechanical [1], electrical [2] and thermal properties [3]. They are mainly assembled into fibers [4-6], yarns [7, 8], and films [9, 10] for advanced materials such as smart electronic textiles [11, 12], multifunctional composites [13] and energy storage materials [14]. However, the assembled CNTs properties are limited due to the weak van der wales interaction between CNTs. Chemical bridging [15, 16], mechanical condensation [17] and physical treatment [18] was intensively studied to enhance the electro-mechanical properties of CNT materials. Nevertheless, hybridizing CNT based material with polymers significantly improved the properties of the resultant composites for functional and structural application.

Soft polymers such as, polyethylene, polyamide, polyurethane, PDMS have been used as matrices for CNT based composite for soft electronics, sensors and actuators due to their flexibility and stretchability [19, 20]. In the other hand, matrices with high mechanical properties, such as epoxy resin, are mainly used as structural materials for aerospace, and automobile parts [21]. However, the weak interfacial bonding between CNTs and polymer matrix is the challenge in manufacturing CNT-reinforced nanocomposites which have to be improved for a better composite. In this line, Liu at all [22]created a strong interfacial bonding between CNT film and epoxy by functionalization of the CNT film surface using plasma treatment. The interfacial bonding between the CNT film and epoxy increased significantly by 156.6%. Shao and co-workers [8] also investigated the interfacial shear strength between an aerogel spun CNT yarn and polyphenylene sulfide using the microdroplet test method. This method pulls a single fiber out of a microdroplet of polyphenylene sulfide matrix to measure the interfacial shear strength between CNT yarn and the matrix. The effective interfacial shear strength between the carbon nano-tubes and polyphenylene sulfide resin was reported 13.1 MPa. Further still, Hang *et al*. [23] described that the ability of the nanotubes to improve composite properties primarily based on their compatibility with the polymer matrix.

In this work, epoxy resin and PVA were reinforced with CNT films separately to achieve composites with high mechanical and interfacial properties. The studied tensile strength and Interlaminar shear strength (ILSS) of CNT films/epoxy and CNT films/PVA composites proved that both polymer matrixes possess excellent mechanical properties unlike each other. It was found that CNT films/epoxy composites have higher peeling strength but CNT films/PVA composites have high the tensile strength with low peeling strength.

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2. EXPERIMENTAL

2.1. Materials

CNT film was produced by floating catalyst chemical vapor deposition (FCCVD) method and was supplied from Suzhou Institute of Nano-Tech and Nano-Bionics (SINANO). The prepared film exhibits high purity (>90%), low density (0.89 g/cm3), and a thickness of ~ 13 μm. Epoxy resin (JL-235) and curing agent (JH-242) were purchased from Changshu Jiafa Chemical Company Ltd, Suzhou, China. PVA-1750 with 98% alcoholysis degree was provided by Sinopec Shanghai Petrochemical Co., Ltd, China.

2.2. Methods

Fabrications of CNT Film Based Composites

CNT film/Epoxy and CNT film/PVA composites were fabricated by VARTM process as illustrated in Figure **1**. The epoxy resin mixture was diluted by the addition of ethanol with weight ratio 1:1 to improve the infiltration of resin in the CNT films. After 15 min immersion in the mixture, CNT film/Epoxy was placed in VARTM mold and vacuumed to squeeze out the excessive resin. Afterward, the mold was pre-cured in the oven at 50 ºC for 3 h and post-cured at 70 ºC for 7 h. Finally, a solid state CNT/Epoxy composite was released from the mold. In the other hand, CNT film/PVA composite was fabricated by immersing CNT film in a mixture of PVA for 1h. Dimethyl sulfoxide (DMSO) was used to dissolve and prepare PVA resin mixture at 65 ºC for 4 h. The resulted CNT film/PVA was transferred to VARTM mold and placed in an oven at 100 ºC for 10 h to dry CNT films. At this temperature, DMSO evaporates and leaves PVA within CNT films to finally fabricate the CNT/PVA composites.

2.3. Characterization

Tensile Tests

CNT film, CNT film/Epoxy and CNT/PVA composites tensile properties were systematically tested. Samples dimension were 15 mm length and 1 mm width as illustrated in Figure **1a**. The tensile tests were conducted on XQ-2 tensile tester (Shanghai Xusai Instrument Co., China) at 0.5 mm/min crosshead speed with a gauge length of 10 mm (ASTM D882). The tensile force (F) was recorded and the tensile stress (σ) was calculated following the expression (1) where A is the surface cross sectional area and L is the length between the grippers (10 mm).

$$\sigma = FA.L \quad (1)$$

Figure **1b** shows the configuration setup on the tensile machine. In addition, a polarized light microscope (ECLIPSELV100POL) was used to take the images of the specimens which were analyzed using Image-J software to calculate the correct width of each sample. Fracture morphologies after tensile stress and composites thickness were observed using Scanning Electron Microscopy HITACHI S-4800 (SEM).

Peeling Tests

Peeling behavior of CNT film/Epoxy and CNT film/PVA composites were performed according to T-Peel test method (ASTM D1876). Samples dimension were 60 mm length, 5 mm width and 10 mm pre-crack length as illustrated in Figure **2a**. The test was conducted on XQ-2 tensile tester (Shanghai Xusai Instrument Co., China) at a crosshead speed of 2 mm/min with a gauge length of 15 mm as shown in Figure **2b**.

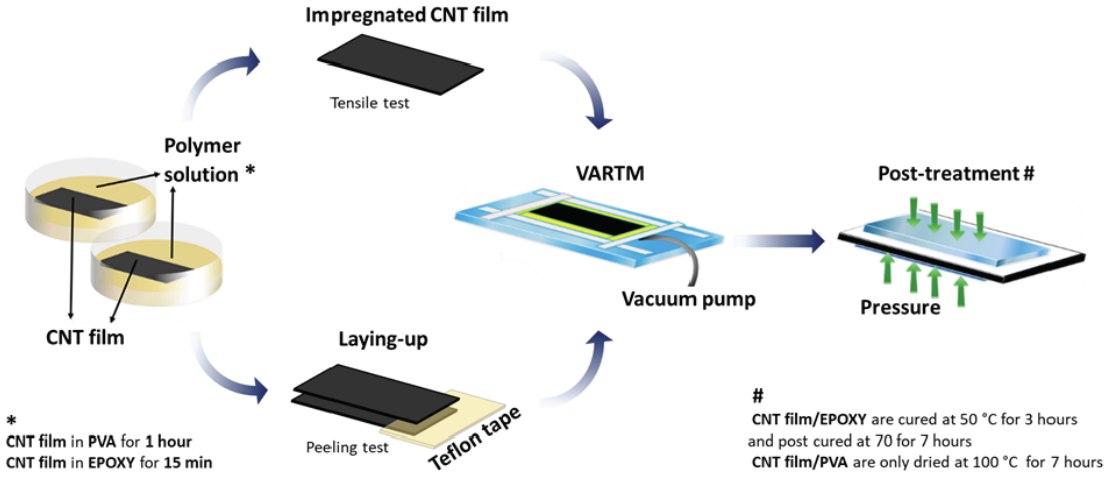

Figure 1: Fabrication methods of CNT film/Epoxy and CNT film/PVA composites.

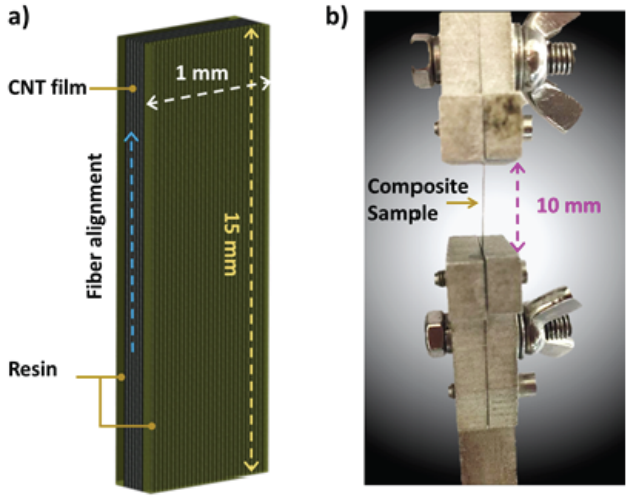

Figure 2: a) Composite dimension for tensile test and **b)** the optical image of the specimen setup.

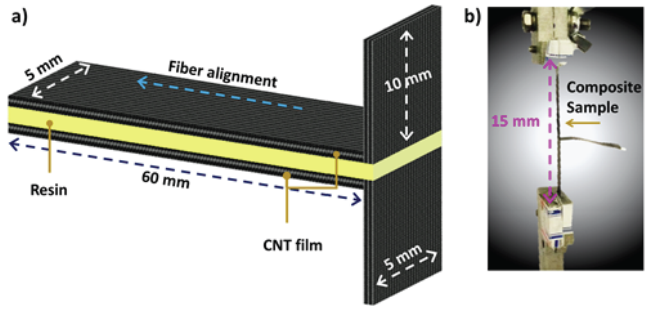

Figure 3: a) Composite dimension for peeling test and **b)** the optical image of the specimen setup.

3. RESULTS AND DISCUSSION

As shown from the SEM cross section images in Figure **4**, the studied polymer matrix have different effects on the CNT film structure. The CNT film and CNT film/epoxy composite thickness was 13.44 and 20.28 µm as shown in Figure **4a** and Figure **4b** respectively. the thickness increasement was due to the coating of epoxy resin on CNT film surfaces. Interestingly, the CNT film/PVA thickness decreased to 10.08 µm as shown in Figure **4c**. PVA is a long chain thermoplastic polymer and it is dissolved in DMSO which is a highly volatile solubilizing agent and caused shrinkage within the composite.

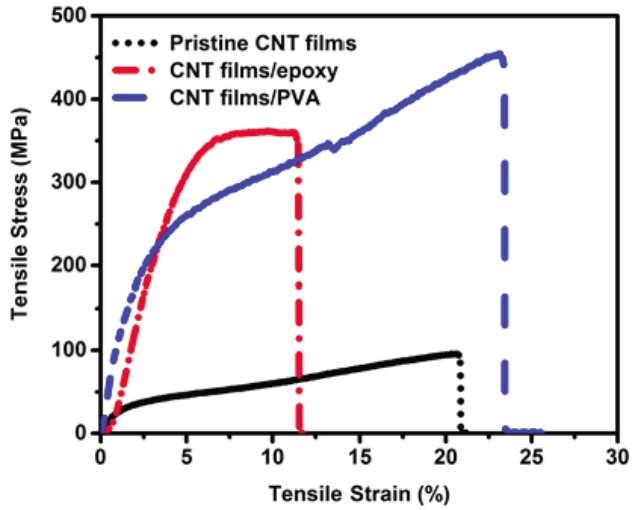

Figure 5: Tensile strength of CNT film, CNT film/Epoxy and CNT film/PVA.

CNT film tensile property increased significantly after immersion in polymer resin as observed in Figure **5**. In addition, the tensile result shows two distinct regions referred to elastic and plastic behavior of the materials studied before rupture.

The tensile strength of CNT film/Epoxy and CNT film/PVA was 332.2 and 452.24 MPa, superior 3.5 and 4.7 times to CNT film tensile strength respectively (Figure **6a**). This increasement demonstrate the effectiveness of composites to enhance CNT film tensile strength and be utilized as structural material for applications such as aerospace and automotive composite parts. In addition, CNT film/PVA composite shows high strain to break due to the thermoplastic nature of PVA (Figure **6b**). In other hand, due to

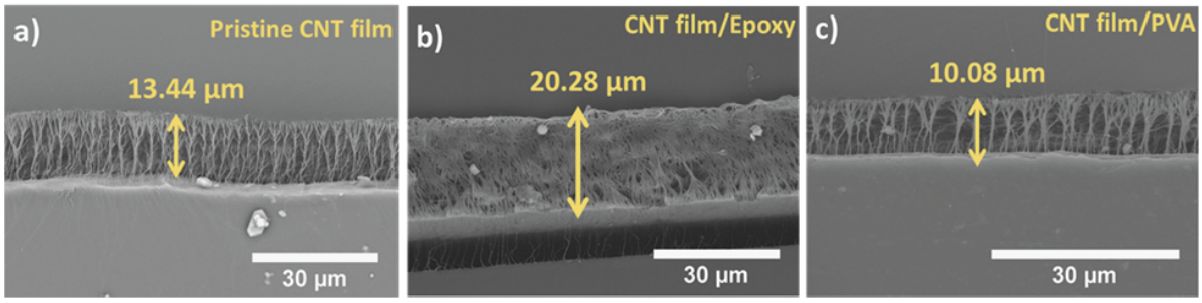

Figure 4: SEM images of **a)** CNT films, **b)** CNT film/Epoxy and **c)** CNT films/PVA composites cross section.

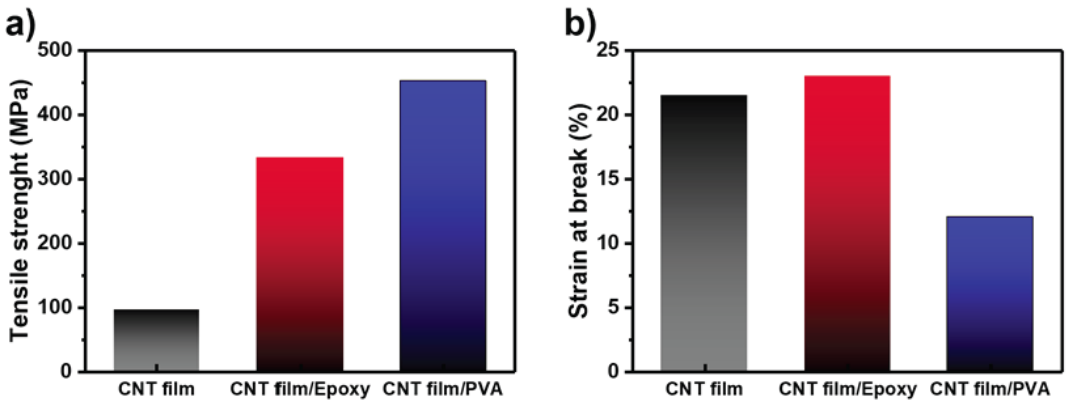

Figure 6: a) Tensile strength and **b)** strain at break of CNT film, CNT film/Epoxy and CNT film/PVA.

toughness of the thermoset epoxy resin, CNT film/Epoxy composite exhibits low strain at breakage. However, CNT film exhibits high strain at break due to rearrangement and friction of CNT fibers and bundles before breakage.

Composites fracture morphology was further investigated with SEM micrographs after tensile test as shown in Figure **6**. CNT films fracture reveals the fibrillary structure of the reinforcement as shown in Figure **6a.b**. SEM images of CNT film / Epoxy fracture was observed at the epoxy matrix level due to the brittleness of epoxy (Figure **6c.d**). Cheng *et al*. [24] reported that the CNTs were broken within the matrix after tensile testing of aligned CNT/epoxy composites. In the other hand, the fractured surface of CNT films/PVA composite showed visible pull-out of CNTs (Figure **6e.f**). Toshio *et al*. [25] reported that the pull-out of CNTs with slightly longer length might be attributed to weaker interfacial shear strength.

Resin selection has also a great effect on composite peeling strenght. As shown in Figure **7**, the peeling load-displacement curves of CNT film/ Epoxy and CNT/PVA composites reveals different behavior. The load needed to peel CNT film/epoxy (Figure **7a**) was more important than CNT film/PVA composite (Figure **7b**). In addition, a noticeable fluctuation when the CNT films/epoxy composite opening was peeled-off which was due to the resin-rich region forming near the opening and therefore, caused defects in the composite and stress concentration [6].

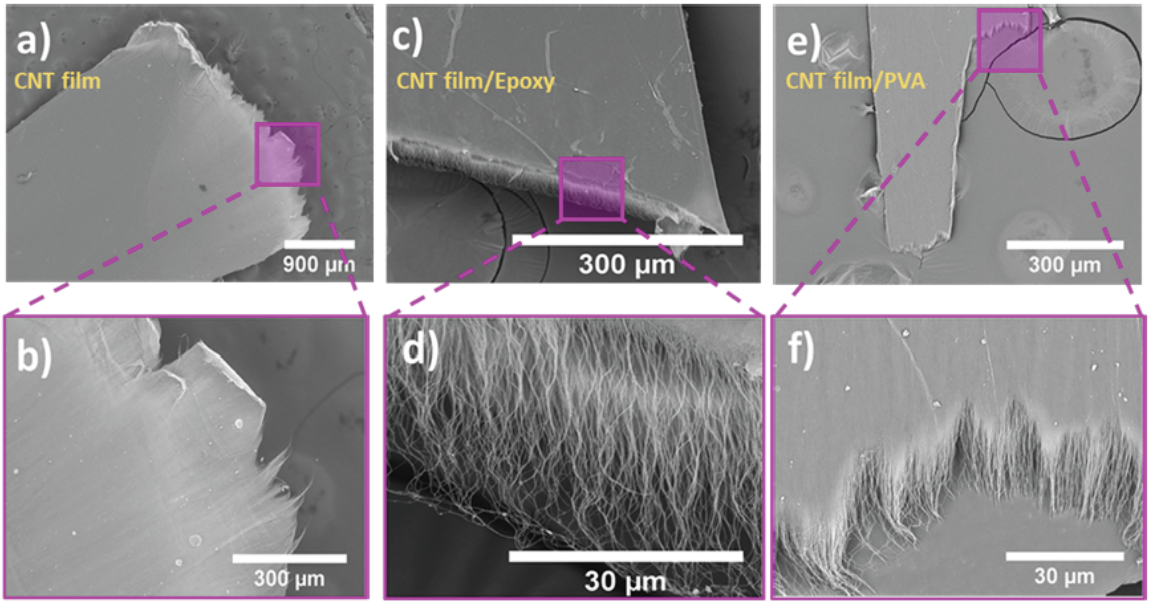

Figure 6: SEM images of the fractured surface after tensile test: **(a** and **b)** CNT films, **(c** and **d)** CNT films/Epoxy and **(e** and **f)** CNT films/PVA composites.

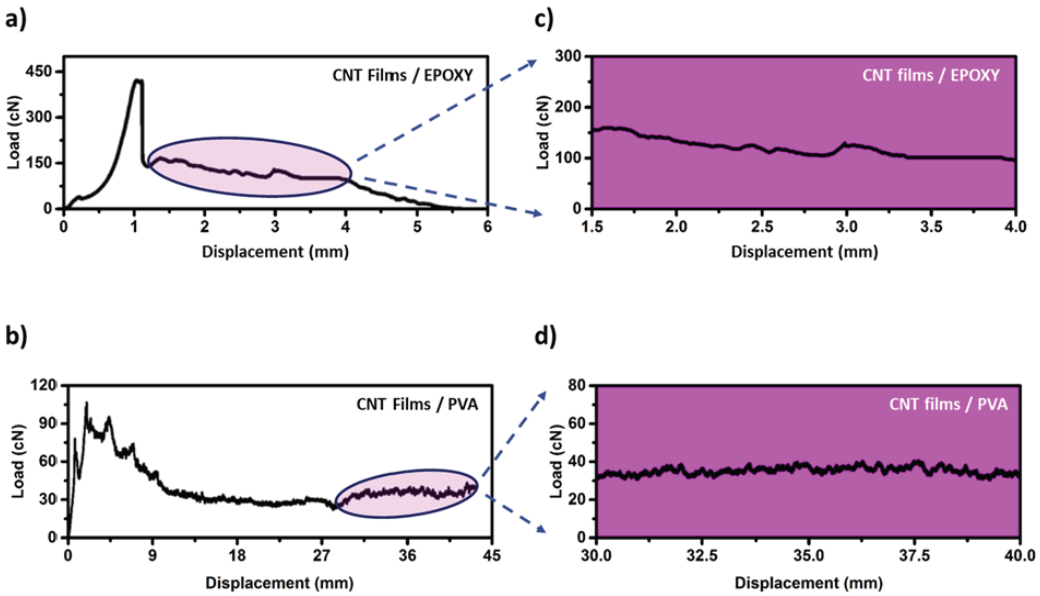

Figure 7: Load-displacement curves of **a)** CNT films/Epoxy and **b)** CNT films/PVA.

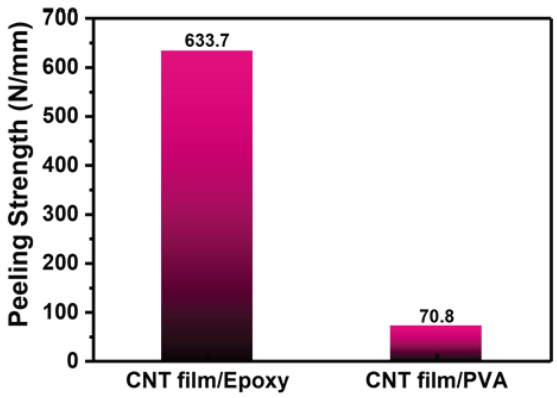

Figure 8: Peeling strength of CNT film/Epoxy and CNT film/PVA composites.

To quantify the peeling strength for both composites, displacement ranging from 1.5 to 4 mm (Figure **7c)** and from 30 to 40 mm (Figure **7d**) was used to calculate interface bond strength of CNT film/Epoxy and CNT films/PVA. The peeling strength of CNT film/Epoxy (633.7 N/m) was **~9** times high than the peeling strength of CNT film/PVA composite (70.8 N/m) as show in Figure **8**. This increasement was due to the strong bonding of CNT films with the epoxy matrix while [26] while the thermoplastic PVA has weak bonding with CNT film, thus carbon nanotube fibers were pulled out smoothly when peeled-off.

CONCLUSION

In summary, carbon nanotube films mechanical properties were enhanced by resin infiltration via VARTM. The tensile strength and interlaminar properties of CNT films/epoxy and CNT films/PVA composites were studied. It was found that peeling strength of CNT films/epoxy composites (633.7 N/m) was significantly improved as compared to CNT films/PVA composites. The CNT films/PVA composites showed peeling strength only as high as 70.8 N/m. However, the CNT films/PVA showed better tensile strength of 452.24 Mpa as compared to that of CNT films/epoxy composites. Such composites can find use in applications where strength is paramount such as aerospace and automobile bodies.

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