Interfacial Architecture Constructed Using Functionalized MWNT Resulting in Enhanced EMI Shielding in Epoxy/Carbon Fiber Composites

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ABSTRACT: In this work, we have attempted to improve electromagnetic interference (EMI) shielding and mechanical behavior of epoxy/carbon fiber (CF) composite, simultaneously, in the presence of functionalized carbon nanotubes. It is well understood that properties of composite depend on the interface between the filler and matrix. Considering this basic understanding, functionalized carbon nanotubes/epoxy nanocomposites were impregnated into a bidirectional carbon fiber (CF) mat and, further, various mechanical and EMI shielding behaviors were studied. Multiwalled carbon nanotubes were functionalized with branched poly(ethyleneimine) (b-MWNT) to tailor the interface of epoxy/CF composites. Laminates with two layers of CF were fabricated with functional MWNT modified epoxy. Scanning electron microscopy was used to analyze the microstructure of epoxy/CF laminates. Lap shear test was performed to analyze adhesion between the modified epoxy and carbon fiber. Further dynamic mechanical analysis in the temperature range of 30−160 °C was performed. Thermal degradation of composites was studied using a thermogravimetric analyzer. Electrical conductivity of laminates was measured using a four-point method on an Agilent probe station. EMI shielding effectiveness (SE) was measured for 0.5 mm-thin laminates in the Ku band. The b-MWNT modified epoxy/CF composites showed excellent SE_E of ca. −60 dB and SE_A of ca. −50 dB, which are of commercial importance. Compared to unmodified epoxy/CF, b-MWNTs/epoxy/CF exhibited 200% increment in EMI SE_E and 35% enhancement in storage modulus due to the improved interface between the epoxy matrix and carbon fiber.

1. INTRODUCTION

Epoxy (Ep)/carbon fiber (CF) composites have brought extensive technological advancements in different sectors due to their versatile properties.1 Epoxy/carbon fiber composites provide a combination of strength and modulus of matrix and filler, ease of fabrication, low density, high strength to weight ratio, and so on.2 These composites are nonisotropic in nature and hence design of components is difficult to compare to that of pure metals or polymers. However, this also provides the additional benefit of tailoring the properties depending on requirements. The replacement of metals with composites has led to reduced weight of aircraft and hence extensively improved efficiency has been achieved. In these composites, in-plane load is carried by the directional fiber, whereas out-of-plane load is transferred by the matrix and interface.3 Hence, developing a better interface between the filler and matrix remains an active research area among the industry and academia. Improved interface reduces the possibility of debonding and hence increases the load-bearing property of the composite.4 Considering these requisites, researchers have continuously attempted to functionalize carbon fiber or epoxy resin with various nanoparticles, such as clay, graphene, montmorillonite, and carbon nanotubes (CNTs), to obtain enhanced properties depending on a wide range of applications.1−14 Owing to the exceptional mechanical, electrical, and thermal behaviors, CNT have always attracted the attention of researchers.15,16 In one of the studies by Kepple and co-workers, multiwalled carbon nanotube (MWNCT) was grown on carbon fiber using the chemical vapor deposition technique and four layered composites were prepared using structural epoxy. These composites were subjected to fracture testing, and fracture toughness was increased by 50% with no structural stiffness loss.17 Bekarova et al.18 developed multi-scale CNT/CF preform by the electrophoresis technique, followed by epoxy composite fabrication using vacuum-assisted resin transfer molding (VARTM). They reported 30% increment in interlaminar shear strength and higher out-of-plane electrical conductivity. Godara et al.19 studied the effects of functionalized and nonfunctionalized CNTs on the mechanical properties and processing of epoxy/carbon fiber composites.

Nowadays, epoxy/carbon fiber composites are the preferred materials for aircraft structure. Current technologies involved in aircraft/satellite design include a highly efficient on-board electronics control system, which leads to electromagnetic perturbations. It can also be caused due to the gadgets or electronic devices used by the boarders.20 For example, during launch and orbiting of satellites, their communication system is
prone to electromagnetic interference (EMI) and hence EMI shielding is required. These disturbances can also lead to the loss of data in the communication system. The improper functioning or performance of any electronic device or system arises due to the interference of electromagnetic waves coming out from nearby devices. Hence, to avoid or minimize the effect of electromagnetic interference, various shielding materials have been employed. The ability of a material of shielding EM waves is expressed in terms of EMI shielding effectiveness (SE), given in terms of decibel (dB). There has been very little study carried out to understand the EMI shielding behavior of epoxy/CF composites. Shah et al. reported reflection loss of -26.8 dB at a frequency of 4.9 GHz for arrayed CF/gradiently dispersed Fe nanoparticle-filled epoxy matrix. Apart from EMI shielding behavior, it is important to understand the mechanical and thermal responses of shielding materials. Because during the service life of shield materials, they have to undergo both mechanical and thermal cycles, there is the possibility of performance deterioration. Hence, it becomes important to study the overall behavior of EMI shielding materials. In the current work, we have attempted to simultaneously improve EMI shielding as well as mechanical and thermal properties of epoxy composites; such materials have potential for use in a wide range of applications.

In our previous work, we studied the functionalization of carboxylic acid terminated MWNTs (c-MWNTs) with branched poly(ethyleneimine) (BPEI) and their effects on mechanical and thermal behaviors of epoxy matrix. In the current study, we have attempted to simultaneously improve the dynamic mechanical and EMI shielding behavior of epoxy/CF composites. To this end, first the epoxy matrix was modified with BPEI functionalized MWNT (b-MWNT), followed by CF composite fabrication. We modified the epoxy with b-MWNT, rather than functionalizing the CF mat itself, which adds to the mechanical disintegration of the mat on account of the harsh chemical treatment involved. Poly(ethyleneimine) has the ability of forming cross-bridges with CF to improve the interface with the matrix epoxy. From our previous study, we understood that amine-terminated BPEI helped in tailoring the interface between the epoxy and MWNT. Hence, this work addressed the concerns often encountered in laminates/sheets designed to shield a specific device from incoming EM radiation, especially concerning the fate of the material (thermal cycles). The heat-map of the laminate on interaction with the incoming EM radiation is studied systematically along with the dynamic mechanical properties. Improved interface together with good dissipation makes these laminates/composites an alternate solution in the quest of lightweight and effective shielding materials.

2. EXPERIMENTAL DETAILS

2.1. Material and Composite Fabrication. Bisphenol-F-based epoxy resin (prepolymer) of molecular weight ≤7000, along with diamine hardener was used as the matrix, provided by Axson Technologies. MWNT was obtained from Nanocyl, Belgium, branched poly(ethyleneimine) (BPEI) (Mn = 25 000) was procured from Sigma-Aldrich, and bidirectional woven carbon fiber mat (average diameter 7 μm) was bought from Hindoostan Technical fabrics.

In our previous work, synthesis and characterization of BPEI functionalized MWNTs from carboxylic acid terminated MWNTs have been explained. In this work, first, epoxy nanocomposites with 1 wt % of c-MWNT and b-MWNT were prepared using the sonication-mechanical mixing method. For this work, we used a low-weight fraction of MWNTs, that is, 1 wt %, because higher concentration of MWNTs leads to the formation of agglomerates. Presence of such agglomerates hinders the cross-linking of epoxy polymers and hence can lead to premature failure of the material. Vacuum-assisted resin transfer molding (VARTM) technique was used to prepare two-layered CF laminates. CF mats were kept at 0° angle, followed by the infusion of b-MWNT/epoxy and c-MWNT/epoxy. Table 1 shows the details of the sample prepared and the code used throughout the article. Scheme 1 shows the different steps involved in fabrication of CF laminates, which include synthesis and dispersion of functionalized MWNTs, preparation of epoxy nanocomposites, impregnation of epoxy nanocomposites into CF mat, followed by vacuum bagging and oven curing to obtain the cured CF laminates.

2.2. Characterization. JPK atomic force microscopy (AFM) was used to image the surface of c-MWNT and b-MWNT in tapping mode. The morphology and cross section of different epoxy/CF composites were observed under an FEI ESEM Quanta 200 scanning electron microscope (SEM). Lap shear test was carried out at 0.5 mm min⁻¹ under tensile loading to measure the adhesion strength of different epoxy nanocomposites with a CF surface. Electrical conductivity of
thin laminates was measured using the four-probe method on Agilent probe station. EMI shielding effectiveness was measured using a vector network analyzer in the range 12–18 GHz on samples of dimension 15 × 8 × 0.5 mm³. Thermomechanical behavior was analyzed using Dynamic Mechanical Analyzer from TA instruments. Thermal degradation studies were carried out on Netzsch thermogravimetric analyzer (TGA) in the temperature range of 30–900 °C in air. Thermal mapping of various CF laminates was carried out by irradiating EM wave of frequency 18 GHz for 10 min.

3. RESULTS AND DISCUSSION

3.1. Morphology of Functionalized MWNTs. AFM is an effective tool to image variation in the surface topography of nanoparticles. Tapping mode AFM was employed to characterize the morphology of different MWNTs. Samples were dispersed quite well in ethanol and spin-coated on silicon wafer. Figure 1a,b shows the AFM micrographs and height image of c-MWNTs and b-MWNTs. Surface undulations of MWNTs increased on functionalization with BPEI, which suggests the presence of polymeric chains on their surface. Again from the height image, the increase in the circumference of c-MWNTs on covalent functionalization was clearly evident. The average diameter of c-MWNTs was 10 nm, whereas b-MWNTs showed increased diameters of 20–22 nm.

3.2. Lap Shear Strength Measurement. Adhesive joints were prepared using neat epoxy, c-MWNT/Ep, and b-MWNT/Ep as adhesives and a carbon fiber reinforced polymer composite-based substrate. Substrates were abraded up to lap length to remove polymer (epoxy) so that a bare CF surface is obtained. It is important to remove surface polymers from the substrate because for our studies we need to understand the adherence behavior of different modified epoxy nanocomposites with a CF surface. Furthermore, different epoxy adhesives were applied on clean substrates. Both substrates were held using a spring clamp to ensure uniform spread of adhesive on the CF surface, and excess adhesive material was removed. Later, these specimens were cured in an oven. Scheme 2 shows the different steps involved in the sample preparation for a single lap shear test. Lap shear experiment was conducted under tensile loading at room temperature according to standard ASTM D1002. Nanoparticles are known to increase adhesive strength due to the improved dispersion state in the matrix, which enlarges the interaction area between the adhesive and adherent. It has been found that CNTs can show strong binding in the direction of applied shear. The force curve as a function of strain was obtained (see Figure 2), and Table 2 enlists the lap shear strength measured for various epoxy nanocomposites. Lap shear strength for b-MWNT/Ep composites was found to be 19.7 MPa, nearly 100 and 40% increment compared to neat epoxy and c-MWNT/Ep nano-
composites, respectively. From the force–strain graph, it can be suggested that the presence of b-MWNTs has increased the failure strain of epoxy along with the lap shear strength of epoxy. It can be concluded that b-MWNT can bind better with the adherent under shear. It also explains the improvement in interface between CF and epoxy due to the presence of functionalized MWNTs.

### 3.3. Microstructure of Laminates

Figure 3 shows the microstructure of different Ep/CF composites with and without functionalized MWNTs. It is important to understand the role of epoxy nanocomposites with c-MWNTs and b-MWNTs on wettability of the CF surface. From Figure 3a, it can be clearly seen that there is debonding between CF and matrix (red highlight) due to the weak interface. These regions were minimized in case of c-MWNT/Ep/CF composites, which can be attributed to the presence of c-MWNTs in epoxy (Figure 3b). In case of b-MWNTs/Ep/CF, the adherence between the adjacent fibers was improved. The presence of BPEI in b-MWNTs/Ep assisted in bridging individual CFs; this can further improve load transfer between the filler and matrix. Such morphologies can enhance the overall mechanical and thermal behavior of composites. Furthermore, we obtained highly magnified SEM micrographs of b-MWNTs/Ep/CF laminates to evaluate the localization of functionalized MWNTs in the laminate. From the SEM micrographs, it was clearly evident that b-MWNTs were also present on the edges of epoxy matrix and closer to the surface of CF. From the lap shear experiment, it was seen that b-MWNTs/Ep has better adherence to the CF surface compared to that of neat epoxy and c-MWNTs/Ep. This can be attributed to the enhanced interface between epoxy and CF due to the presence of b-MWNTs.

### 3.4. Electrical Conductivity

Electrical sheet resistance ($R_s$) of CF laminates was performed using four-probe measurements at room temperature (ASTM F76). Further, electrical resistivity was calculated from the van der Pauw technique. It provides the average resistivity of an arbitrary-shaped sample. For our work, rectangular-shaped CF laminates of thickness ($t$) 0.5 mm were cut and silver paste was applied on the corners to ensure proper contact of probes with the sample surface. Electrical resistivity, $\rho$, and conductivity for CF laminates were calculated.

From Table 3, it was observed that electrical resistance possessed by b-MWNT/Ep/CF was the highest among composites. Similarly, its electrical conductivity was the lowest compared to that of c-MWNT/Ep/CF and Ep/CF composites. This can be explained from the fact that BPEI is an insulating polymer that forms an interface between epoxy and CF. There was no significant change in resistance value, which suggests that the contribution of CF is more prominent than that of various modified epoxies. These composites are conducting enough to create a Faraday cage to shield devices against incident EM waves.

### 3.5. EMI Shielding Effectiveness

As discussed earlier, EMI SE provides information about attenuation of the propagating EM waves caused by the shielding material. It is obtained using a two-port vector network analyzer (VNA) in a given frequency range in terms of S-parameters, scattering parameters ($S_{11}, S_{12}, S_{21}, S_{22}$). Shielding is mainly governed by three mechanisms, namely, reflection ($R$), absorption ($A$), and multiple reflection ($M$). Total shielding effectiveness ($SE_T$) can be calculated from the following equation:

$$S_{21}^2 = \frac{P_{out}}{P_{in}}$$

$$SE_T = 10 \log \left( \frac{P_{out}}{P_{in}} \right)$$

$$SE_T = SE_R + SE_A + SE_M$$

Figure 4 shows $SE_T$ as a function of frequency for c-MWNT/Ep/CF and b-MWNT/Ep/CF composites in the range of 12–18 GHz. Two-port VNA was coupled with waveguide setup using an SMA type of connector. Before the measurement, short-open-load-through calibration was carried out. It was observed that c-MWNT/Ep/CF could shield up to −50 dB, whereas b-MWNT/Ep/CF showed EMI SE of −60 dB. Polymer composites are inhomogeneous in nature, unlike metals or conducting polymers. In the present study, heterogeneous junctions were formed consisting of functionalized MWNTs, epoxy, and CF. These types of junctions interact with EM waves differently and decrease the energy of EM waves by absorption and multiple reflection. Reflection mechanism can be explained from the high electrical conductivity shown by the composites. $SE_R$ was similar for all of the three composites because reflection contribution comes mainly from the CF surface due to high electrical conductivity. $SE_A$ shown by different composites was nearly $−10$ dB (Figure 5) and contributes little to the overall SE of the composites. On further investigation, we found that the major shielding effectiveness contribution is from the absorption mechanism. Ep/CF composites showed the lowest $SE_A$ value of approx. $−15$ dB. Interestingly, addition of 1 wt % of functional MWNTs, that is, c-MWNTs and b-MWNTs in Ep/CF composites, exhibited excellent absorption of EM waves. $SE_A$ for c-MWNTs/Ep/CF and b-MWNTs/Ep/CF composites was found to be approx $−39$ and $−50$ dB, respectively, at various frequencies (Figure 6). This can be further explained by the heterogeneity generated in Ep/CF composites by the inclusion...
of c-MWNTs and b-MWNTs. These functional MWNTs form an interface or nonuniform interfacial junction between the epoxy matrix and CF. It is well known that MWNTs possess free electron cloud on their surface. These functional MWNTs
retain charge cloud to a certain extent even after functionalization. Furthermore, the charge accumulation at the interface also contributes to the higher absorption of EM waves in Ep/CF.\(^{28}\) The relatively higher value of \(\sigma_{\text{A}}\) for b-MWNTs/Ep/CF can be explained by the better dispersion of b-MWNTs in the epoxy matrix and hence leading to better and multiple interfacial junctions with CF. In our work, we observed that there were certain undulations in SE values at higher frequencies of 16–18 GHz. This can be attributed to mainly the nonuniform formation of a conductive mesh between the epoxy nanocomposite and CF.\(^{29–33}\) It can also be explained by the fact that at higher frequencies, these conductive networks formed in the modified laminates effectively convert EM energy into heat and leakage current.\(^{34}\)

3.6. Thermal Mapping. Thermal mapping of various CF laminates was carried out by irradiating EM waves of frequency 18 GHz for 10 min. Further thermal response of laminates was scanned using a Fluke Ti 25 IR (fusion technology) thermal sensor (Figure 7a). High-frequency EM wave irradiated on the Ep/CF laminate does not show significant increase in temperature, as seen in Figure 7b. Composites with 1 wt % of c-MWNTs and b-MWNTs showed a slight increase in temperature by 1–2 °C, as observed from thermal maps. This is due to the presence of residual functional groups attached to the surface of carboxylic MWNTs and BPEI functionalized MWNTs (Figure 7c,d).\(^{35}\) CF and MWNTs show excellent thermal conductivity due to which heat dissipation in CF composite is quite efficient.\(^{36–38}\) This can further be attributed to the thermal stability of CF laminates while irradiation and absorption of EM waves.

3.7. Dynamic Mechanical Analysis. Dynamic mechanical analyzer is an efficient tool to analyze the mechanical behavior of a material as a function of temperature.\(^{39}\) Figure 8 represents the storage modulus of CF laminates as a function of temperature. The experiment was performed in the temperature range of 30–160 °C, frequency 10 Hz, and amplitude 15 \(\mu\)m. Ep/CF showed a storage modulus of 17 GPa, whereas c-MWNT/Ep/CF showed an increment of approx. 18% in storage modulus with 20 GPa. b-MWNT/Ep/CF exhibited the highest storage modulus of 24 GPa. This can be attributed to the enhanced interface between the modified epoxy and CF mat.\(^{40}\) b-MWNTs form a better interface due to the enhanced dispersion in the epoxy matrix.\(^{35}\) Furthermore, b-MWNTs hinder the mobility of Ep/CF under mechanical loading and hence improve the stiffness of the composite. The plastic behavior in b-MWNTs/Ep/CF composites initiates only after 110 °C, which explains that these composites can be used at higher temperatures without material failure.

3.8. Thermal Degradation. In case of polymer composites, it becomes important to analyze the thermal degradation temperature. On degradation, polymer breaks down into small molecules CO, CO\(_2\), NO, and so on, which are harmful in nature. Figure 9 represents the percent weight loss of CF composites as a function of temperature. TGA analysis for

| sample          | sheet resistance \((R_s)\) (Ω) | resistivity \((\Omega \text{ cm})\) | conductivity \((\text{S cm}^{-1})\) |
|-----------------|-------------------------------|----------------------------------|----------------------------------|
| Ep/CF           | 1.32                          | 0.07                             | 14                               |
| c-MWNT/Ep/CF    | 1.07                          | 0.05                             | 20                               |
| b-MWNT/Ep/CF    | 4.12                          | 0.21                             | 4                                |

Figure 4. \(\sigma_{\text{E}}\) as a function of frequency.

Figure 5. \(\sigma_{\text{R}}\) as a function of frequency.

Figure 6. \(\sigma_{\text{A}}\) as a function of frequency.

Table 3. Electrical Conductivity of Various Composites

Figure 7. Thermal mapping of CF laminates.

Figure 8. Storage modulus of CF laminates as a function of temperature.

Figure 9. Percent weight loss of CF composites as a function of temperature.
Composites was performed between 30 and 900 °C. CF composites showed only one slope at 350 °C, which is the degradation temperature of the epoxy matrix. There was no observable degradation shown by carbon fiber.

4. CONCLUSIONS

In this work, we studied the effects of b-MWNTs on EMI shielding as well as the thermal and mechanical behaviors of...
Ep/CF composites. The morphology of functionalized MWNTs was obtained from AFM. CF laminates were fabricated with different modified epoxy systems using the VARTM technique. The microstructure of laminates was examined under SEM, and it was observed that b-MWNTs/Ep formed a better interface with the CF surface. Lap shear strength was measured for different epoxy nanocomposites on the CF substrate. It was found that b-MWNT/Ep showed a lap shear strength of 19 MPa, which was 110% higher than that of neat epoxy. EMI SE was measured in the frequency range of 12–18 GHz. SE for b-MWNT/Ep was found to be ~60 dB, and the storage modulus was as high as 24 GPa. b-MWNTs/Ep/CF composites exhibited excellent absorption behavior, having an SE value of maximum ~49 dB at 17 GHz. Such materials with excellent EMI shielding along with mechanical and thermal characteristics are of commercial importance to aircraft industries where the communication system works in the Ku band.

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Notes
The authors declare no competing financial interest.

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