Poly(ether ether ketone)-Grafted Graphene Oxide “Interconnects” Enhance Mechanical, Dynamic Mechanical, and Flame-Retardant Properties in Epoxy Laminates

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ABSTRACT: Herein, the interface of epoxy and graphene oxide (GO) is tailored through hydroxylated poly(ether ether ketone) (HPEEK). The resultant modification (HPEEK-g-GO) improved the interfacial adhesion between epoxy and carbon fiber (CF) in the laminates. This strategy resulted in improved tensile strength, modulus, and storage modulus by 8, 10, and 26%, respectively, with respect to control samples (epoxy/CF laminates). The HPEEK-g-GO was thoroughly characterized using spectroscopic techniques and was infused using vacuum-enhanced resin infusion technology into the epoxy/CF laminates. To address the challenges involved with primary agglomeration, the composite formulation was subjected to mechanical stirring coupled with bath sonication throughout the mixing process. The improved structural properties in epoxy/CF laminates were attributed to HPEEK-g-GO “interconnects”, which provided the necessary reinforcement owing to better interfacial adhesion with the CF mat as inferred from the fracture surface morphology assessed using SEM. In addition, the epoxy laminates containing HPEEK-g-GO also showed flame-retardant properties along with good thermal stability. The electromagnetic interference shielding capability of the modified laminates was also evaluated in the frequency range of 12−18 GHz. It was observed that the laminates exhibited a shielding effectiveness of −50 dB. Thus, this strategy offers some promise in fabricating epoxy/CF laminates with multifunctional properties through HPEEK-g-GO “interconnects”.

INTRODUCTION

The use of carbon fiber (CF)-reinforced polymer matrix composites has seen substantial growth in aerospace, transportation, and automotive because of their high specific strength and modulus and their ability to be tailored for myriad applications.1−3 At the same time, scientific efforts have been concentrated on the improvement in interfacial properties as this significantly impacts the final properties of composites.4−6 Interfacial adhesion is crucial, and interfacial adhesion strength can, in general, be dominated by two phenomena, namely, chemical bonding and mechanical interlocking.7,8 Thus, this interfacial adhesion can be tailored by modifying the matrix using various methods such as incorporation of nanofillers or “interconnects” that leads to an improved interface and eventually the mechanical properties.

Epoxy resins, a dominant thermosetting polymer, are widely used because of their superior thermal and mechanical properties in adhesives, coatings, and structural materials. The benefits of epoxy over other thermosets have been well documented in the literature.9 Thus, among the various thermosets available, epoxy resins play a dominant role as a matrix in the development of high-performance materials because of their excellent thermal stability, mechanical properties, and processability.1,2 Their highly cross-linked structure after curing makes them inherently brittle and prone to cracks, which becomes challenging upon their utility in structural applications. Therefore, many scientific studies have been performed with an aim to improve the interface between epoxy and CF mats.

To this end, there exist studies that discuss different strategies to improve the properties of the epoxy resin by addition of various fibers and also by blending with other thermoplastics.10 Recently, with the use of nanofillers such as carbon nanotubes (CNTs), graphene oxide (GO) is also being explored as modifiers for epoxy resins as it offers exceptional properties.11 GO with abundant functional groups on the basal plane can be dispersed well in polar polymers.12−15 The main advantage of GO over other carbon derivatives such as CNTs is the availability of abundant functional groups, which can be harnessed to improve its dispersion in epoxy resin.16 GO has a high tendency to agglomerate because of van der Waals’ forces and strong π−π stacking.17,18 Therefore, the recent research is focused on addressing this challenge by functionalizing the GO sheets.11,19,20

Poly(ether ether ketone) (PEEK) is a thermoplastic and has attracted many researchers because of the exceptional...
properties that it offers, which can be utilized for various applications such as high-performance automotive and space applications.\textsuperscript{21–23} Hence, in an attempt to tailor the interfacial adhesion between epoxy and CF, PEEK was grafted onto GO sheets and used as interconnects. In the present work, the mechanical properties of epoxy-based laminates were evaluated in the presence of GO and HPEEK-g-GO utilizing the benefits of HPEEK, which is miscible with the epoxy prepolymer. The grafting of HPEEK onto GO improved not only the dispersion of GO in epoxy but also the interfacial adhesion between epoxy and CF in the laminates. The tensile, flame retardant, and electromagnetic interference (EMI) shielding properties of the laminates were systematically investigated.

\section*{CHARACTERIZATION}

The synthesis procedure of GO and HPEEK-g-GO is discussed in our previous work.\textsuperscript{24} Both GO and modified GO were characterized by transmission electron microscopy (TEM) using an FEI Tecna T20 S-TWIN, Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction analysis (XRD), and X-ray photoelectron spectroscopy (XPS). Limiting oxygen index (LOI) tests were performed according to ASTM D2863. The sample size used for the LOI was 70 mm \(\times\) 6.5 mm \(\times\) 3 mm (length \(\times\) width \(\times\) thickness). The apparatus used was procured from Concept Equipment Ltd., U.K. The specimen was mounted on a holder and ignited with a burner so that only the upper face was exposed to the flame. All the measurements were repeated 3 times, and the average value is reported. The morphology of fractured tensile specimens was observed under a scanning electron microscope (SEM; ULTRA 55, FESEM (Carl Zeiss)). Modulated differential scanning calorimetry (DSC-Q2000 from TA Instruments) was performed to gain insight into the thermal transitions in the composites. Briefly, the samples were heated at 2 °C/min with an amplitude of 1 °C and a period of 60 s. About 70 nm sections of epoxy composites were trimmed using a Leica Ultramicrotome at room temperature for TEM analysis. The tensile test of epoxy/CF laminates containing GO or HPEEK-g-GO was carried out as per ASTM D 3039. The specimen size used for tensile testing was 250 mm \(\times\) 12.5 mm \(\times\) 2 mm. An interlaminar shear strength (ILSS) test was carried out as per ASTM D 2344. The specimen size was 20 mm \(\times\) 10 mm \(\times\) 2 mm. Dynamic mechanical thermal analysis (DMTA) was performed using three-point bending on a TA Q800 in the temperature range from 40 to 200 °C under a constant vibration frequency of 1 Hz with an amplitude of 15 μm. EMI shielding of laminates was studied using an Anritsu MS4642A vector network analyzer (VNA). KEYCOM waveguide is used to measure the parameters of the thin layered samples in the 12–18 GHz frequency region.

\section*{RESULTS AND DISCUSSION}

\subsection*{DSC Analysis of Epoxy/Carbon Fiber Laminates.} \(T_g\) of the epoxy/CF laminates was studied by MDSC (see Figure 1). It is observed that for the neat epoxy/CF laminate, the \(T_g\) is ca. 171 °C, which reduced to 167 °C in the presence of 0.5 wt % GO. Interestingly, the \(T_g\) increased to 176 °C in the presence of HPEEK-g-GO at a similar loading. The decrease in \(T_g\) in the case of GO suggests a reduced crosslinking density in epoxy. Similar observations were reported by Putz et al.\textsuperscript{25–27} Alternatively, the literature also reports that upon functionalizing, the GO sheets can also increase the \(T_g\) of the epoxy composites.\textsuperscript{28,29} In our case, the increase in \(T_g\) in the presence of modified GO is plausibly due to the improved dispersion of GO due to the functionalization with PEEK, which, in turn, reduces the macromolecular mobility of the epoxy chains and increases the \(T_g\).

\subsection*{Flammability and Thermal Properties.} The flame-retardant properties of the epoxy resins were examined by measuring the limiting oxygen index (LOI) as shown in Figure 2. It is well-known that epoxy resins are highly flammable, with an LOI value of 21.\textsuperscript{30} With the addition of CFs, the LOI value increased to 30. It was interesting to note that with the addition of GO, the LOI values further enhanced marginally. It showed about 31.5 and 32 for 0.5 wt % GO and 0.5 wt % HPEEK-g-GO filled epoxy/CF laminates, respectively. This ultimately indicates that the addition of small amount of HPEEK-g-GO\textsuperscript{31} improves the flammability properties as compared to neat epoxy samples. Figure 3 shows the degradation curves for various epoxy/CF laminates. It was observed that all epoxy CF laminates containing GO and HPEEK-g-GO were thermally stable up to 300 °C.

\subsection*{Mechanical Properties.} The major challenge with epoxy-based composites containing nanoparticles is the formation of voids and microvoids because of several factors such as moisture absorbed during storing and volatiles released by chemical reactions, which ultimately results in deterioration of the mechanical properties, especially the tensile properties.\textsuperscript{32–34} To avoid the above mentioned issues, the layup under vacuum was dried in an oven before infusing with the resin. The resin mixture was also heated up to reduce the viscosity, which resulted in the easy infiltration and release of volatiles to a certain level. The curing was carried out keeping
the vacuum suction continuous on the laminate. This almost eliminates the entrapment of volatiles. The specimens from the cured laminate were cut using a diamond cutter as per ASTM standard. Glass/epoxy composite tabs were bonded to the specimens. The tabs were bonded to protect the surface from damage during clamping in a universal testing machine and to obtain a monotonic stress strain profile without sudden stress spikes. The specimens were dried before testing at 120 °C for 30 min. The specimens were tested on a Zwick Z150 UTM at a test speed of 1 mm/min. The ultimate tensile strength (UTS) was measured by the maximum stress the specimens withstood before failure. The strains were measured using an extensometer during the test. The stress strain curves for epoxy/CF laminates are shown in Figure 4. These strain data were used to calculate the elastic modulus ($E$) of the specimens.

$$E = \frac{UTS}{\varepsilon}$$

where $E$ is the elastic modulus and $\varepsilon$ is the strain.

UTS was enhanced by about 6 and 8% with the addition of 0.5 wt % GO and HPEEK-g-GO in epoxy/CF laminates, respectively. The values are listed in Table 1. The improvement in tensile strength and modulus suggests that there is formation of a strong interfacial bond between CF and epoxy, which was also observed from the SEM micrographs. In other words, inclusion of GO and HPEEK-g-GO in composites acted as “interconnects”, thereby improving the stress transfer at the interface. It is well understood that HPEEK-g-GO is well dispersed in epoxy as HPEEK improves the interface between GO and epoxy. In the laminates, as inferred from the SEM micrographs, HPEEK-g-GO resulted in better interfacial adhesion of epoxy with CF. On the contrary, bare CF can be observed in the epoxy/CF laminates. This supports the fact that HPEEK-g-GO improves the interfacial adhesion between CF and epoxy and eventually enhanced the ILSS properties also. By suitably modifying the surface properties of CF, different functional groups can be harnessed, which, when they react with the epoxy chains, can improve greatly the ILSS. However, weak $\pi$-$\pi$ interaction between GO and CF possibly may result in “GO interconnects” at the CF surface. This may also result in improved interfacial adhesion between epoxy modified with HPEEK-g-GO and CF, which will be discussed in detail in the subsequent sections (Table 2).

Table 1. Mechanical and Flammability Properties of Epoxy Laminates

| laminates          | ultimate tensile strength (UTS) (MPa) | tensile modulus (GPa) | ILSS (MPa) | LOI values |
|--------------------|---------------------------------------|-----------------------|------------|------------|
| epoxy/CF           | 733 ± 68                              | 64 ± 2                | 45 ± 1.8   | 30         |
| epoxy/CF/GO        | 778 ± 34                              | 70 ± 4                | 47 ± 1.7   | 31.5       |
| epoxy/CF/HPEEK-g-GO| 798 ± 45                              | 71 ± 3                | 50 ± 3.4   | 32         |

Table 2. List of Samples and Their Codes Used in This Work

| sample                               | sample code   |
|--------------------------------------|---------------|
| epoxy + CF                           | epoxy/CF      |
| epoxy + CF + 0.5 wt % GO             | epoxy/CF/GO   |
| epoxy + CF + 0.5 wt % HPEEK-g-GO     | epoxy/CF/HPEEK-g-GO |

The improved tensile properties in the laminates in the presence of HPEEK-g-GO is due to the load-carrying capacity of GO in the CF/epoxy laminates. Higher energy is required for failure of the laminates containing GO and HPEEK-g-GO epoxy matrix compared to only neat epoxy matrix. The load that is applied is then transferred from the epoxy matrix to the CF through GO interconnects. When a crack propagates under loading during failure, the nanoparticles are pulled off from the epoxy matrix, and the energy is dissipated. This energy is due to debonding and fracture of CF and deformation of the epoxy matrix under loading. The failure mechanism depends on the types of fiber, matrix, and interfaces. The dispersion of GO and HPEEK-g-GO bridges mechanical interlocking between the CF and epoxy resin as shown in the cartoon below based on...
the observed SEM micrographs. This ultimately improves the ultimate mechanical properties.

The SEM images of fractured epoxy laminates are shown in Figure 5a−g. It is clear from the SEM micrographs that the CF mats are well sandwiched between the epoxy layers (Figure 5a). From Figure 5d−g, it is clear that the interfacial adhesion between epoxy and CF is enhanced because of HPEEK-g-GO when compared to GO. This is manifested in the layer of the epoxy on the CF surface in the case of HPEEK-g-GO, which also facilitated the improvement of the mechanical properties.

Figure 5. Fractured SEM images of (a−c) epoxy carbon fiber laminates, (d, e) epoxy/CF laminates containing GO, and (f, g) epoxy/CF laminates containing HPEEK-g-GO.
A cartoon shown in Figure 6 illustrates the possible interaction of GO and HPEEK-g-GO with epoxy/CF laminates on the basis of the observations from SEM analysis. The epoxy/CF and epoxy/GO/CF laminates failed predominantly by pulling out of the CF, whereas in the case of epoxy/HPEEK-g-GO/CF laminates, a thick layer of epoxy is observed to be well coated on the surface of the CF. Taken together, the debonding between the fiber and matrix is the dominant mechanism of failure, and the interface is the weakest part, even though a small amount of GO sheets was introduced, whereas, in the case of HPEEK-g-GO, because of the weak $\pi-\pi$ interaction between GO and CF and also because of the improved dispersion of GO aided by HPEEK in epoxy, the interfacial adhesion between epoxy and CF was greatly improved resulting in enhanced mechanical properties.

Delamination is one of the common and major failure modes in layered structures. The interlaminar stresses are the source of delamination and crack growth in composites. It is imperative to understand the behavior of nanoparticles at the interface of the layers. The interfacial adhesion should not get lower. The lower ILSS leads to easy delamination upon loading. It was envisaged that the addition of nanoparticles will improve the ILSS and, hence, was measured in the samples where the adhesion between layers resulted in shear under specific loading conditions. There are various methods to determine the ILSS, of which the better approach is by the short beam shear test (ASTM D 2344) due to its simplicity. The shear stress is independent of the support length because of the small sample size. The test fixture consisted of a loading cylindrical roller of 6.0 mm diameter and a side support of 3.4 mm diameter. Specimens were tested at a cross-head speed of 1 mm/min. The ILSS was determined using the following equation:

$$\text{ILSS} = 0.75 \times \frac{P_{\text{max}}}{b \times h}$$
where ILSS is the interlaminar shear strength, $P_{\text{max}}$ is the failure load, $b$ is the width of the sample, and $h$ is the thickness of the sample.

The ILSS results are tabulated in Table 1. It was observed that with the addition of nanoparticles, the ILSS of 0.5 wt % GO and 0.5 wt % HPEEK-g-GO was enhanced from 45 MPa to 47 MPa and 50 MPa, respectively. Improvement in the ILSS is attributed to enhanced dispersion of functionalized GO in epoxy. These GO sheets with abundant oxygen-containing functional groups impart polarity, hence, resulting in an interlocking effect between the fiber and matrix and eventually enhancing the ILSS by bridging the load between epoxy and CF.\textsuperscript{13}

**DMTA of Epoxy/CF Laminates.** DMTA was used to evaluate the mechanical properties of epoxy/CF laminates. The storage modulus, loss modulus, and tan $\delta$ curves are shown in Figure 7a–c. It is observed that for the neat epoxy CF laminate, the storage modulus was around 20.1 GPa, which was enhanced to 23.3 GPa with the addition of 0.5 wt % GO in the epoxy matrix. Further, the storage modulus has improved to 25.4 GPa with the addition of 0.5 wt % of HPEEK-g-GO in the epoxy matrix. The same trend was observed in the epoxy composites with the addition of 0.5 wt % GO and HPEEK-g-GO without addition of CF in our previous study. The enhancement in storage modulus suggests effective load transfer from epoxy to the filler, which is attributed to the reinforcing action by GO sheets and HPEEK-g-GO. The latter enhances the interfacial bonding between GO sheets and epoxy resin through HPEEK grafting.\textsuperscript{13,40} Figure 7c shows the rubbery to glassy transition in epoxy CF laminates using the tan $\delta$ curves. The $T_g$ is observed to be around 185 °C for neat epoxy, which decreases slightly to 182 °C with the addition of 0.5 wt % GO, whereas it increases to 192 °C with the addition of 0.5 wt % HPEEK-g-GO. These results exhibit the same trend as shown by MDSC in the previous section.

**Electromagnetic Interference (EMI) Shielding.** The EMI shielding effectiveness of various epoxy laminates was measured in the frequency range of 12 to 18 GHz and is shown in Figure 8. The total shielding effectiveness ($S_{TE}$) can be estimated using scattering parameters obtained by VNA and is expressed as\textsuperscript{41}

$$S_{TE}(\text{dB}) = 10 \log \left( \frac{1}{S_{12}^2} \right) = 10 \log \left( \frac{1}{S_{21}^2} \right)$$

where $S_{12}$ and $S_{21}$ are the reverse and forward transmission coefficients, respectively. There are two mechanisms, namely, reflection and absorption, responsible for EMI shielding. Reflection is enhanced by connectivity and conductivity of the shielding material. It is envisaged that CF can act as a waveguide and attenuate the incoming EM radiation.\textsuperscript{42} The laminate containing only CF displayed around $\sim$50 dB shielding effectiveness. The continuous CFs, which are present in the epoxy matrix, play an important role, which transports electrical charge over large distances without electrical losses.\textsuperscript{43} Interestingly, it was observed that the modified laminates with 0.5 wt % GO and 0.5 wt % HPEEK-g-GO also showed similar shielding effectiveness as compared to the epoxy neat CF laminate. This suggests that although insulators like HPEEK covers the GO sheets, the charge transfer through the CF mat is largely unaltered, although accumulation of charge at the interface may result in lossy interfaces.\textsuperscript{44,45} The mechanism behind the total shielding effectiveness for epoxy laminates is shown in Figure 9. This result suggests that multifunctional properties can be achieved in epoxy/CF laminates using HPEEK-g-GO.

**CONCLUSIONS**

In this study, a unique strategy was adopted wherein HPEEK was grafted onto GO sheets through esterification. As observed from TEM analysis, HPEEK-g-GO disperses well in the composites even after curing. Besides good dispersion, the observed 8% improvement in tensile strength, 10% increase in Young’s modulus and 26% enhancement in storage modulus of the epoxy/CF laminates are attributed to the good interfacial bonding between HPEEK-g-GO and epoxy. This was further supported by the fact that a layer of epoxy modified with HPEEK-g-GO was observed on the CF surface as compared to bare CF in epoxy/CF laminates. The addition of HPEEK-g-GO also led to enhanced glass transition temperature in the laminates. In addition, the flame-retardant properties were also enhanced in the presence of GO and HPEEK-g-GO, and the EMI shielding properties were retained, although an insulating layer of HPEEK is present on the GO surface. Taken together, the present work demonstrates that by grafting HPEEK onto GO sheets, the interfacial adhesion and the dispersion of GO sheets in epoxy can be improved, which eventually results in enhanced mechanical and thermal properties in the laminates even with small fractions of HPEEK-g-GO.

**EXPERIMENTAL**

**Materials.** Epoxy resin (EPOLAM 2063) and an anhydride-based hardener were obtained from Axson Technologies (France). The bidirectional CF mat was supplied by Hindoostan Technical Fabrics. Poly(ether ether ketone) was provided by Vestakeep, Germany. Graphite flakes and the reagents used in this study were procured from Sigma. The solvents were obtained from commercial sources and were used without any pretreatment.

**Fabrication of Carbon/Epox composite laminate.** The carbon/epoxy composite laminate was prepared using vacuum enhanced resin infusion technology (VERITY), a CSIR-NAL proprietary process.\textsuperscript{16} In this process, the reinforcement is held...
in a tool cavity and is infused with resin under a differential pressure to completely wet the fiber bundle. A toughened glass plate was used as a tool for fabrication of the laminate. The glass plate was used to capture the behavior of resin flow during infusion. Eight layers of carbon fabric (300 mm × 300 mm) were cut. The edges of the layer were trimmed, aligning warp and weft fibers. The glass plate was cleaned for any dust, and a release film was placed on the plate. A release fabric (peel ply) was placed over the release film. The carbon layers were placed above each other, aligning the warp fiber as reference. The layup sequence followed was 0/908. A release fabric was then placed on top, and then a porous release film was placed on top of the release fabric. The release fabric was used to get a rough texture on the surface, which would facilitate the tab bonding on the specimens for testing. Finally, a highly permeable resin distribution medium was placed on top, and the edges were sealed to avoid resin race tracking. The setup was vacuum-bagged keeping the resin feed line on one side and the vacuum on the opposite side. Full vacuum was ensured by sealing the vacuum bag completely. Simultaneously, the GO or HPEEK-g-GO was dispersed in ethanol, followed by sonication for 30 min. The EPOLAM 2063 epoxy resin was added to the mixture, and bath sonication was done for 30 min. To evaporate the solvent from the mixture, it was kept in a vacuum oven for 24 h. The resultant mixture was mechanically stirred at 400 rpm, followed by the addition of the hardener (in 100:107 weight ratio), followed by 15 min of mechanical mixing at 500 rpm. This resin mixture was then heated to 45 °C and degassed for 20 min. As prepared, the VERITY setup was heated in the oven for 30 min at 50 °C. Resin heating and glass plate heating was carried out to keep the resin viscosity low during infusion, ensuring the complete wetting of the fiber tows. The viscosity of the resin mixture at 45 °C is 250 cP. The resin mixture was then infused at 45 °C under a differential pressure, and the infused laminate was cured at 80 °C for 7 h. The weight ratio (60/40) of the CF and epoxy resin mixture was maintained during the laminate fabrication. Post-curing was carried after demolding of the laminate under free-standing conditions, that is, without vacuum and pressure at 180 °C for 4 h. The specimens were cut using a specially made composite specimen-cutting machine.
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