

Establishment of interlaminar structure and crack propagation in carbon fiber reinforced epoxy composites by interleaving CNTs/PEK-C film

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Abstract

The synergistic combination of carbon nanotubes (CNTs) and ductile thermoplastic resin has shown large potential in the improvement of fracture resistance for the epoxy matrix composites using the interleaving toughening method in recent years. The hybrid structure of CNTs and thermoplastic resin in interlayers affects directly the interlaminar structure and the resultant crack propagation path of the interleaved composites. In this work, the CNTs and thermoplastic polyetherketone-cardo (PEK-C) were used to prepare the interlayer with different hybrid structures to interleave the carbon fiber reinforced epoxy composites and the influence of hybrid structure on the interlaminar structure and the fracture toughness was investigated. The results showed that PEK-C/CNT/PEK-C sandwich interlayer produced the best toughening effect in mode I interlaminar fracture toughness (G_{IC}) and the G_{IC} was 446.76 J/m², increased by 138.11% compared to blank composites, which benefited from the multilayered structure in the interlaminar region formed during curing process and the resultant tortuous crack propagation.

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Keywords

Fracture toughness, Crack propagation, Hybrid structure, Carbon nanotubes, Epoxy matrix composites

Abstract

The synergistic combination of carbon nanotubes (CNTs) and ductile thermoplastic resin has shown large potential in the improvement of fracture resistance for the epoxy matrix composites using the interleaving toughening method in recent years. The hybrid structure of CNTs and thermoplastic resin in interlayers affects directly the interlaminar structure and the resultant crack propagation path of the interleaved composites. In this work, the CNTs and thermoplastic polyetherketone-cardo (PEK-C) were used to prepare the interlayer with different hybrid structures to interleave the carbon fiber reinforced epoxy composites and the influence of hybrid structure on the interlaminar structure and the fracture toughness was investigated. The results showed that PEK-C/CNT/PEK-C sandwich interlayer produced the best toughening effect in mode I interlaminar fracture toughness (G_{IC}) and the G_{IC} was 446.76 J/m², increased by 138.11% compared to

blank composites, which benefited from the multilayered structure in the interlaminar region formed during curing process and the resultant tortuous crack propagation.

Introduction

Carbon fiber reinforced plastics (CFRPs) are widely used as structural materials in the fields of aerospace, aviation, automobile, etc. due to superior specific strength and stiffness, excellent fatigue and corrosion resistance. Epoxy has the advantages of good adhesion strength, thermal resistance and high mechanical properties and is largely used as the matrix in CFRPs. However, the intrinsic brittleness of epoxy due to the crosslinking network after curing reaction results in the poor fracture toughness and low crack resistance, which has always been the weakness of epoxy matrix composites and restricts the application [1-3].

Interleaving toughening refers to inserting a discrete intermediate layer between adjacent layers of composite laminates [4-5]. The discrete layer can promote the formation of mechanical bridges between the crack interface and the tortuous crack path, and a crack tip plastic zone, which can greatly improve the interlaminar fracture toughness of composites. The interleaving toughening method has little impact on other properties and the preparation process of composite laminates. Thermoplastic resin is widely utilized to prepare the interlayer because of good ductility and compatibility with epoxy and exhibits excellent toughening effect [6-8].

In recent years, the nanomaterials, such as zero-dimensional inorganic nanoparticles [9,10], one-dimensional carbon nanotubes (CNTs) [11,12], two-dimensional nanosheet [13] and multidimensional nanomaterials [14], have been used to interleave composites due to the nanoscale features and the outstanding mechanical properties, revealing large potential in improving fracture toughness. The CNTs attract widespread attention of researchers due to the large length to diameter ratio and the relatively low cost. On the one hand, the CNTs are individually used in the interlaminar region of composite laminates and exert the toughening effect through the debonding, pulling-out and breakage. Y. Y. Yu et al. deposited directly the CNTs film on the surface of carbon fiber fabric using floating catalytic chemical vapor deposition (FCCVD) method to prepare the interleaved composites. The mode II interlaminar fracture toughness (G_{IIC}) was improved by 94% and the bridging effect and the pulling-out of CNTs from resin contributed to the toughness improvement [15]. In the study of K. Almuhammadi et al., the spray gun was used to spray CNTs/ethanol solution on the surface of composite prepreg to prepare the interleaved composites and the study results showed that the mode I interlaminar fracture toughness (G_{IC}) was only increased by 17%. The researchers considered that the toughening efficiency of CNTs was restricted because the cracks were deflected from the interlaminar CNTs-rich zone to the interface zone between interlaminar region and fiber/epoxy ply [16]. On the other hand, the CNTs are used in combination with the ductile materials, such as the rubber and the thermoplastic resin. X. G. Xu et al. used multi-wall carbon nanotubes (MWCNTs)/polyetherketone-cardo (PEK-C) hybrid films as interlayers for carbon fiber/bismaleimide (BMI) composite laminates. The compression after impact (CAI) strength was significantly improved due to the effect of BMI/PEK-C dualphase structure and the pulling-out and breakage of CNTs [17]. O. Kaynan et al. studied the mixed I + II fracture toughness (G_C) of the interleaved composite by MWCNTs /Polyvinylbutyral (PVB) nanofiber film. The G_C was greatly increased by about 2 times, benefiting from the synergistic effect of stiff CNTs and adhesive PVB nanofibers in prompting plasticization and deflecting the induced crack propagation [18]. V. Eskizeybek et al. used CNTs/polyacrylonitrile (PAN) hybrid nanofiber film to interleave composite and the G_{IC} was increased by 77%. The introduction of CNTs reduced the diameter of nanofibers, resulting in the increase of the surface area and the adhesion enhancement between the layers. What's more, the bridging, pulling-out and breakage of CNTs also contributed to the improvement of fracture toughness [19]. In conclusion, the network, composed of CNTs, ductile materials and matrix, plays an important role in the toughening effect of interlayer. The hybrid structure of CNTs and ductile material affects certainly the interlaminar structure and the resultant crack propagation path of the interleaved composites. In the study of N. Zheng et al., the CNTs were attached to both sides of polysulfone (PSF) nanofiber by the vacuum filtration method to prepare CNT-PSF-CNT sandwich interlayer. The G_{IC} and G_{IIC} were further improved by 53.1% and 13.3% respectively compared with pure PSF nanofiber film [20]. Y. Gao et al. used the EMAA-CNT-EMAA

sandwich structure to interleave composite, but the G_{IC} exhibited a downward trend compared with pure ethylene methacrylic acid (EMAA) film [21], which was contrast to the study of N. Zheng. However, the different change trend might be caused by different material system and process method. No conclusion on the influence of hybrid structure has been reached. Therefore, this study aims to investigate the influence of CNTs/ductile materials hybrid structure on the toughening efficiency and establish the targeted interlaminar structure and the crack propagation path by interleaving method.

In this study, the CNTs and the ductile thermoplastic resin PEK-C were used to prepare the interlayers with different hybrid structures to interleave carbon fiber reinforced epoxy composite laminates. The interlaminar fracture toughness of composites was evaluated through mode I interlaminar fracture toughness tests. The interlaminar structure, composed of CNTs, PEK-C resin and epoxy matrix, was characterized by scanning electron microscope (SEM). The crack propagation path was depicted and the toughening mechanism was developed.

Materials and experiments

2.1. Materials

The unidirectional carbon fiber reinforced epoxy (T700/H[®] 6240) composite prepreg with a fiber volume fraction of about 60% and PEK-C powder were provided by Tianjin Hanshuo Advanced Materials Co., Ltd. The high-purity multi-walled carbon nanotubes (TNMC3) was provided by Chengdu Organic Chemistry Co., Ltd. The parameters were shown in Tab.1. The tetrahydrofuran (THF) and the ethanol were supplied by Tianjin Komiou Chemical Reagent Co., Ltd. The THF was used to dissolve PEK-C powder and disperse the CNTs in the preparing process of PEK-C/CNTs hybrid films and the ethanol was used to disperse the CNTs in the preparing process of CNTs films without the effect of organic solvent on the prepreg and PEK-C film. The polytetrafluoroethylene (PTFE) film with the thickness of 10 μm used for preparing precracks was brought from Hong Fluoro Insulation Materials Co., Ltd.

Tab.1 Parameters for PEK-C and TNMC3

Materials	Characteristics	Characteristics
PEK-C	Chemical formula	
	Density (g/cm^3)	1.309
	Glass transition temperature (T_g) ($^{\circ}$)	213
	Tensile strength of PEK-C film (MPa)	131
	Elongation at break of PEK-C film	112%
TNMC3	Outer diameter (nm)	10-20
	Purity (%)	>98
	Length (μm)	10-30
	Specific surface area (m^2/g)	>150
	Tap density (g/cm^3)	0.22
	Content of carboxyl (-COOH) (wt %)	2

2.2. Preparation

2.2.1. Interlayer preparation

Five types of interlayers were prepared in this study and the schematic diagram was shown in Fig.1. They were PEK-C film (P), CNTs film (C), PEK-C/CNTs hybrid film (PC), PEK-C/CNT/PEK-C sandwich film (PCP) and CNT/PEK-C/CNT sandwich film (CPC), respectively.

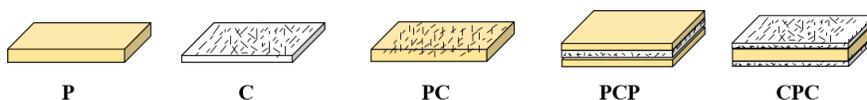


Fig.1 The schematic diagram of five types of interlayers

PEK-C film and PEK-C/CNTs film

The PEK-C film and PEK-C/ CNTs film were prepared by spin coating method using the coater machine (KW-4A, SETCAS Electronics, China).

A certain amount of PEK-C powder was dissolved in THF to form PEK-C/THF solution by fully stirring. And then the CNTs were uniformly dispersed in THF to form CNTs/THF dispersion by the ultrasonic homogenizer (JY92-IIDN, SCIENTZ, China). Finally, the PEK-C/THF solution and CNTs/THF dispersion were uniformly mixed by the ultrasonic homogenizer to form PEK-C/CNTs/THF solution with the PEK/THF weight ratio of 12% and CNTs/PEK-C weight ratio of 3% [22].

The mixture was deposited onto the glass substrate of coater machine and the parameters were the low-speed of 500 rpm for 30 s and then high-speed of 1500 rpm for 30 s. The films were removed from the substrate with hot vapor after the volatilization of THF [22]. The preparation process of PEK-C/CNTs films was depicted in Fig.2. The PEK-C films were prepared following the same procedure without CNTs.

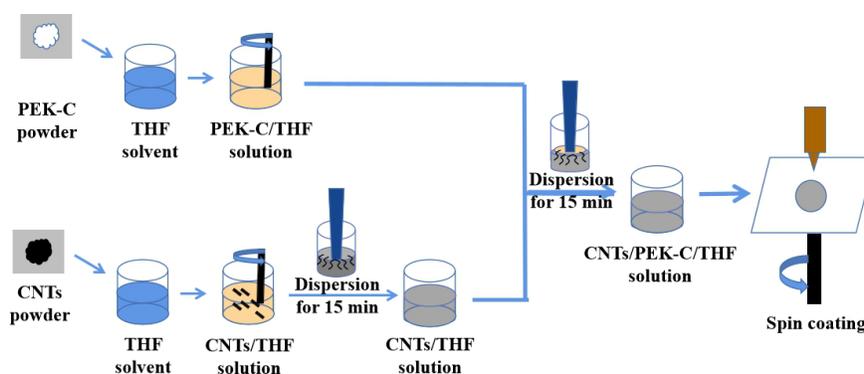


Fig.2 Preparation procedure of PEK-C/CNTs hybrid film

CNTs film, PCP film and CPC film

The certain amount of CNTs and the ethanol were mixed and uniformly dispersed by the ultrasonic homogenizer. The amount of CNTs was taken based on the amount of CNTs in PC hybrid film. The CNTs dispersion was directly sprayed onto the surface of T700/H[®] 6240 prepreg or PEK-C film to form CNTs film and the ethanol was evaporated at ambient temperature for 2h.

2.2.2 Laminate preparation

The hot press molding process was applied in the paper to prepare composite laminates by autoclave (YT-14-01, Dalian Yingtian, China). Cut the T700/H[®] 6240 prepreg into pieces of 250 mm×250 mm and prepare the lay-up [0]₁₆ by hand. The PTFE film and the interlayer were placed into the mid-plane of composites during lay-up process. Put the lay-up into the autoclave with the apparatus (shown in Fig.3). The curing pressure, time and temperature were respectively set up to 0.6 MPa, 2h and 135.

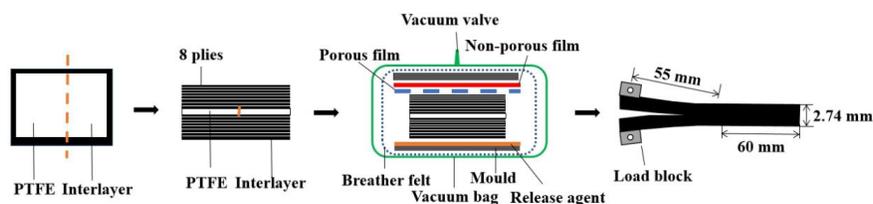


Fig.3 Preparation process of composite laminates for G_{IC} tests

2.3. Experiments

2.3.1. Mode I interlaminar fracture toughness test

Mode I interlaminar fracture toughness (G_{IC}) of composites was conducted on a universal electronic testing machine (Instron 5982, Instron, USA) based on the ASTM D5528 standard [23]. The double cantilever beam (DCB) tests were adopted in this paper. The specimen was shown in Fig.3. The initial crack length was 55 mm. Mark every 1 mm or 5 mm on the side of the test specimens from 55 mm to 105 mm. The test was conducted following the three stages: firstly, load until the crack length reached 60 mm; secondly, upload until the displacement returned to the initial position; finally, reload until the crack expanded to the marked 105 mm. Five specimens were tested for each group of experiments. The Modified Beam Theory (MBT) was used to calculate G_{IC} and the G_{IC} calculation formula is as follows:

$$G_{IC} = \frac{3P\delta}{2b(a+\Delta)} \quad (\text{Eq.1})$$

Where P is the load, δ is the load point displacement, b is the specimen width, a is the crack length, and Δ is the corrective factor of crack length and determined by generating a least squares plot of the cube root of compliance based on the standard. In the following text, the notation G_{IC} represented the fracture toughness at the initial stage of crack growth and G_{IR} represented the averaged fracture toughness in the first 20 mm of crack propagation, characterizing the crack propagation resistance [22, 24].

2.3.2. Morphology characterization

The fracture surface after G_{IC} tests was investigated using a scanning electronic microscope (SEM, S-3400N, Hitachi, Japan). Gold layer was sputter-coated on the surface before tests. To figure out the phase morphology, the fracture surfaces were chemically etched with THF in a beaker for 96 h at 25 °C, then washed in an ultrasonic bath for 1 h and finally air-dried at room temperature.

Results and discussion

3.1 Mode I interlaminar fracture toughness (G_{IC})

The mode I interlaminar fracture toughness test results of blank and all the interleaved composite laminates were shown in Fig.4. Fig.4(a) displayed the R curves, the change of G_{IC} versus the crack length. It could be seen that the fracture toughness was improved via introducing interlayers. The G_{IC} and G_{IR} values were depicted in Fig.4(b). The G_{IC} and G_{IR} of blank composites were 187.63 J/m² and 198.43 J/m², respectively. The toughening efficiency of PEK-C and CNT films was quite different. The G_{IC} and G_{IR} were increased by 101.03% and 78.79% via interleaving PEK-C film, whereas only 10.79% and 25.71% via CNT film. It was worth noting that the G_{IC} and G_{IR} were decreased after introducing 3% CNTs into PC hybrid film in comparison to the composite interleaved by neat PEK-C film. The composite interleaved by CPC and PCP sandwich film exhibited better toughening effect. The composite interleaved by PCP film exhibited the highest G_{IC} and G_{IR} values, which were respectively 446.76 J/m² and 451.04 J/m², increased by 138.11% and 127.30% compared to blank composites.

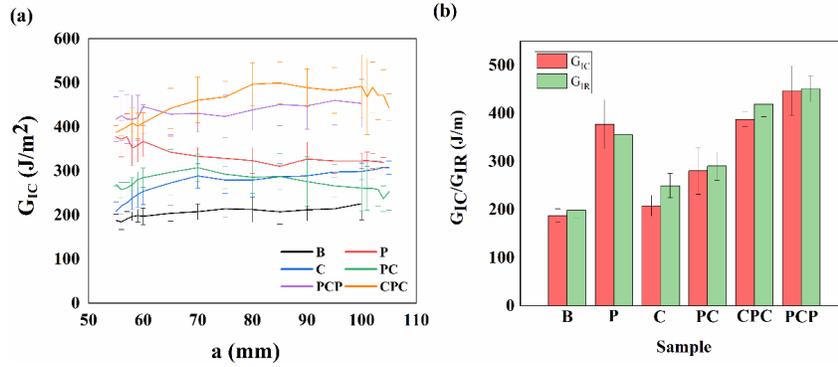


Fig.4 Test results of mode I interlaminar fracture toughness of blank and all the interleaved composite laminates: (a) R curves; (b) G_{IC} and G_{IR} values

3.2 SEM morphology

Fig.5 displayed the fracture morphology of blank composites, which showed the peeling-off of the fibers and smooth surface of neat epoxy matrix. The strips caused by matrix fracture could be seen in the enlarged image (Fig.5(b)), which demonstrated the brittle fracture features and poor fracture resistance.

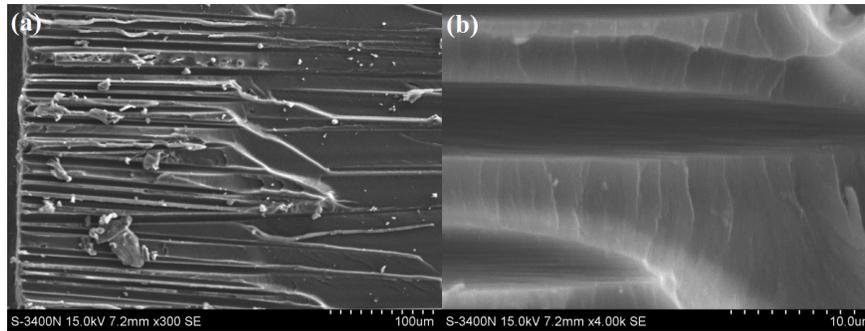


Fig.5 Fracture surface of blank composites after G_{IC} test

Fig.6 represented the fracture surfaces of the interleaved composites by CNTs (Fig.6(a1)-(d1)), PEK-C (Fig.6(a2)-(d2)) and CNTs/PEK-C film (Fig.6(a3)-(d3)), respectively. The fibers were exposed on the fracture surface of the CNTs interleaved composites. The fiber surface and the surrounding resin surface were rougher in comparison to blank composites (Fig.6(a1)). Two types of morphology characteristic could be distinguished through the magnified image. The first one was the resin fragmentation (Fig.6(b1)), which might be because the CNTs were scattered in the resin and the resin was torn along some CNTs during the crack propagation. The other one was the ductile fracture of resin (Fig.6(c1)), which was consistent with most of the research results that the introduction of CNTs could toughen resin [25, 26]. The two fracture patterns contributed to the dissipation of fracture energy and the improvement of fracture toughness. From the Fig.6(d1), the scattered CNTs could be seen, indicating the pulling-out and breakage.

Fig.6(a2)-(d2) represented the fracture surface of the interleaved composites by PEK-C film. The fracture surface was mostly covered with resin. It could be seen that the layered structure, consisting of “scale-like” PEK-C monophase structure, nodular and sea-island dualphase structures, was formed in the interlaminar region (Fig.6(b2)), which was explained by the concentration changes of PEK-C due to the interdiffusion of resin in curing process. The significant improvement of fracture toughness was benefited from the layered structure, as reported in our previous study [22]. Fig.6(c2) and (d2) showed the enlarged “scale-like” PEK-

C monophase structure and the dualphase resin surrounding fibers. It was worth noting that the fibers exhibited smooth surface.

Fig.6(a3)-(d3) displayed the fracture surface of the interleaved composites by PEK-C/CNTs hybrid film. The surface was rougher compared to the PEK-C interleaved composites. Fig.6(b3) showed similar layered structure with Fig.6(b2). The “scale-like” structure was enlarged as shown in Fig.6(c3). It could be found that the CNTs were unevenly embedded in the “scale-like” structure and they were clustered together at the center of “scale” and surrounded by “burrs”. The “scale-like” structure turned into smaller pieces and the surface was rougher due to the introduction of CNTs, which was totally different from that of the PEK-C interleaved composites (Fig.6(c2)). Fig.6(d3) showed the fiber surface and the surrounding resin morphology. The fibers were covered with dualphase structure, which was also different from the PEK-C interleaved composites due to the adding of CNTs.

The neat PEK-C film could effectively improve the fracture toughness through the layered structure formed between epoxy and PEK-C resin. The PEK-C/CNTs hybrid film with introducing 3% CNTs weakened the toughening effect in G_{IC} with respect to the PEK-C film. The layered structure was also formed in the interlaminar region of the PEK-C/CNTs film interleaved composite, whereas the introduction of 3% CNTs resulted in the different morphology characteristics. From the point of view of fracture morphology, the toughness gap could not be explained. It was speculated that the different crack propagation path played a more important role in the improvement of fracture toughness, which would be rediscussed in the discussion section.

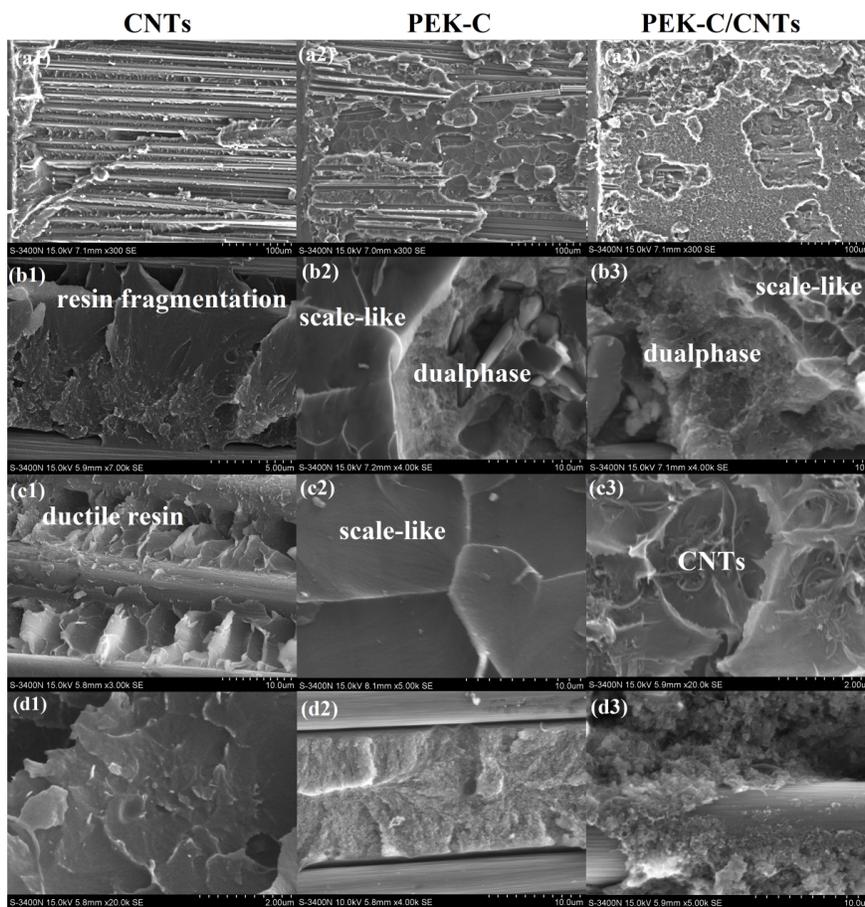


Fig.6 Fracture surface of the composites interleaved by CNTs, PEK-C and CNTs/PEK-C films after G_{IC}

test: (a1)-(d1) CNTs films, (a2)-(d2) PEK-C films and (a3)-(d3) CNTs/PEK-C hybrid films

Fig.7(a) showed the fracture surface of the interleaved composites by PCP sandwich interlayer, illustrating more complex layered morphology due to the sandwich structure of interlayer, which could be clearly seen from the magnified view Fig.7(b) and Fig.7(c). From Fig.7(b), the fibers in the upper ply were peeled off and the grooves were left. The “scale-like” and “needle-like” morphology could be found and lay below the upper ply. From Fig.7(c), the fibers in the lower ply were exposed and above the lower ply exhibited also the “needle-like” and “scale-like” morphology. Therefore, it could be reasonably concluded that the layered morphology, composed of upper ply, dualphase structure between PEK-C and epoxy, CNTs and lower ply, was formed in the interlaminar region of the PCP interleaved composites. The “needle-like” structure had not been found in the previous samples and was only formed in the PCP and CPC interleaved composites. Fig.7(d) displayed the enlarged image of “needle-like” structure, which was most probably caused by the drawing of CNTs from epoxy resin because the fracture morphology of resin as shown in Fig.7(d) did not exhibit any evidence of dualphase structure and ductile deformation.

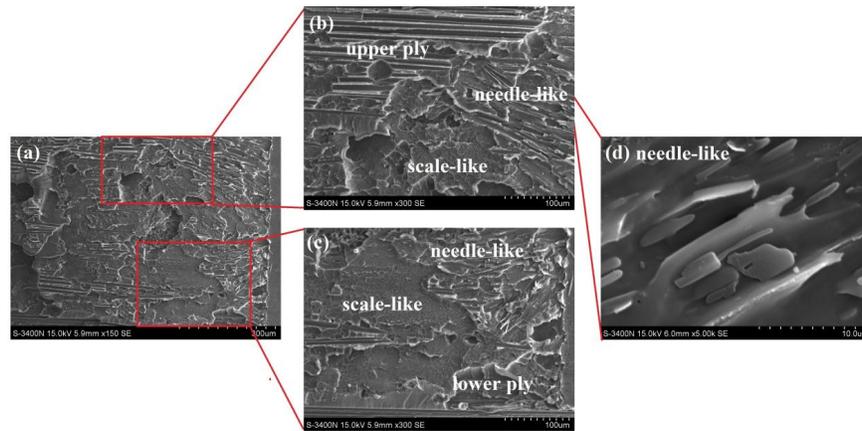


Fig.7 Fracture surface of the composites interleaved by PCP sandwich film

after G_{IC} test

Fig.8 displayed the surface morphology after chemical etching of fracture surface in Fig.7, which retained mainly the morphology before etching (Fig.8(a)). The “scale-like” structure was removed and the smooth epoxy surface was exposed, confirming the “scale-like” PEK-C monophase structure . The “needle-like” structure was not changed (Fig.8(d)), indicating the epoxy fracture.

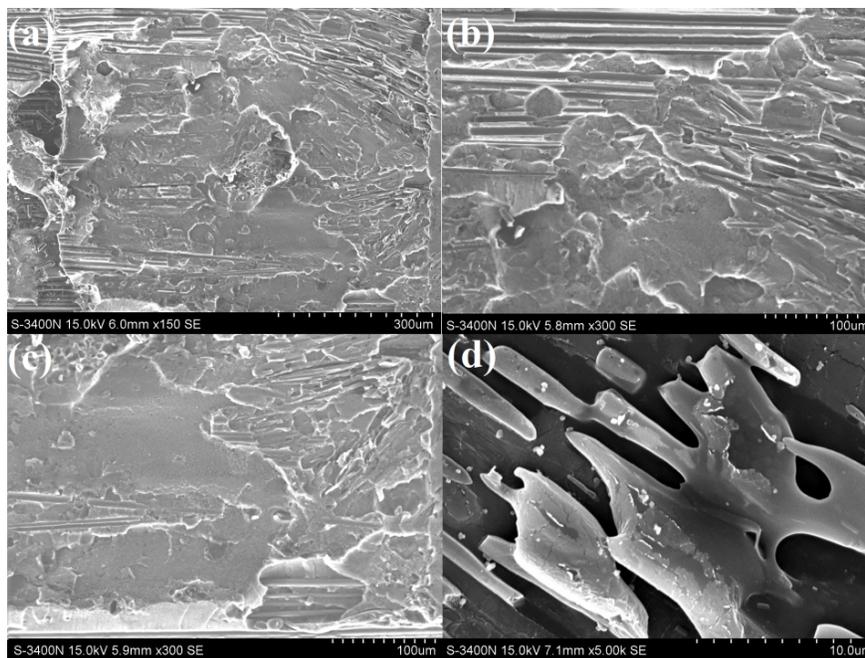


Fig.8 Fracture surface after chemical etching in Fig.7

Fig.9 displayed the fracture surfaces of the interleaved composites by PCP (Fig.9(a1)-(d1)) and CPC (Fig.9(a2)-(d2)) sandwich interlayers, respectively. Similarly, the CPC interleaved composites formed the layered structure (Fig.9(a2)). From Fig.9(b2), the “scale-like” and “needle-like” structures existed between upper and lower ply and the “needle-like” structure lay next to fibers and between lower ply and “scale-like” structure.

The enlarged “scale-like” structures in the PCP and CPC interleaved composites were shown in Fig.9(c1) and (c2), which were totally different from that in the PC interleaved composites (Fig.6(c3)). From Fig.9(c1), the CNTs was relatively uniform. Therefore, it could be speculated that the film formation in the spinning coating preparation process of CNTs/PEK-C hybrid film led to the concentration of CNTs in PEK-C resin. The CNTs influenced the nucleation and growth of PEK-C film and the structure of PEK-C film developed from the CNTs [27].

Fig.9(d1) and (d2) showed the resin surrounding the fibers in PCP and CPC interleaved composites, respectively. The PEK-C/epoxy dualphase structure was formed surrounding the fibers in PCP interleaved composites. What’s more, the resin fragmentation caused by CNTs could be found, which was similar to that of the CNTs interleaved composites (Fig.6(b1)). From Fig.9(d2), the resin surrounding fibers was reinforced by CNTs. The resin fragmentation and toughening of resin cause by CNTs were found.

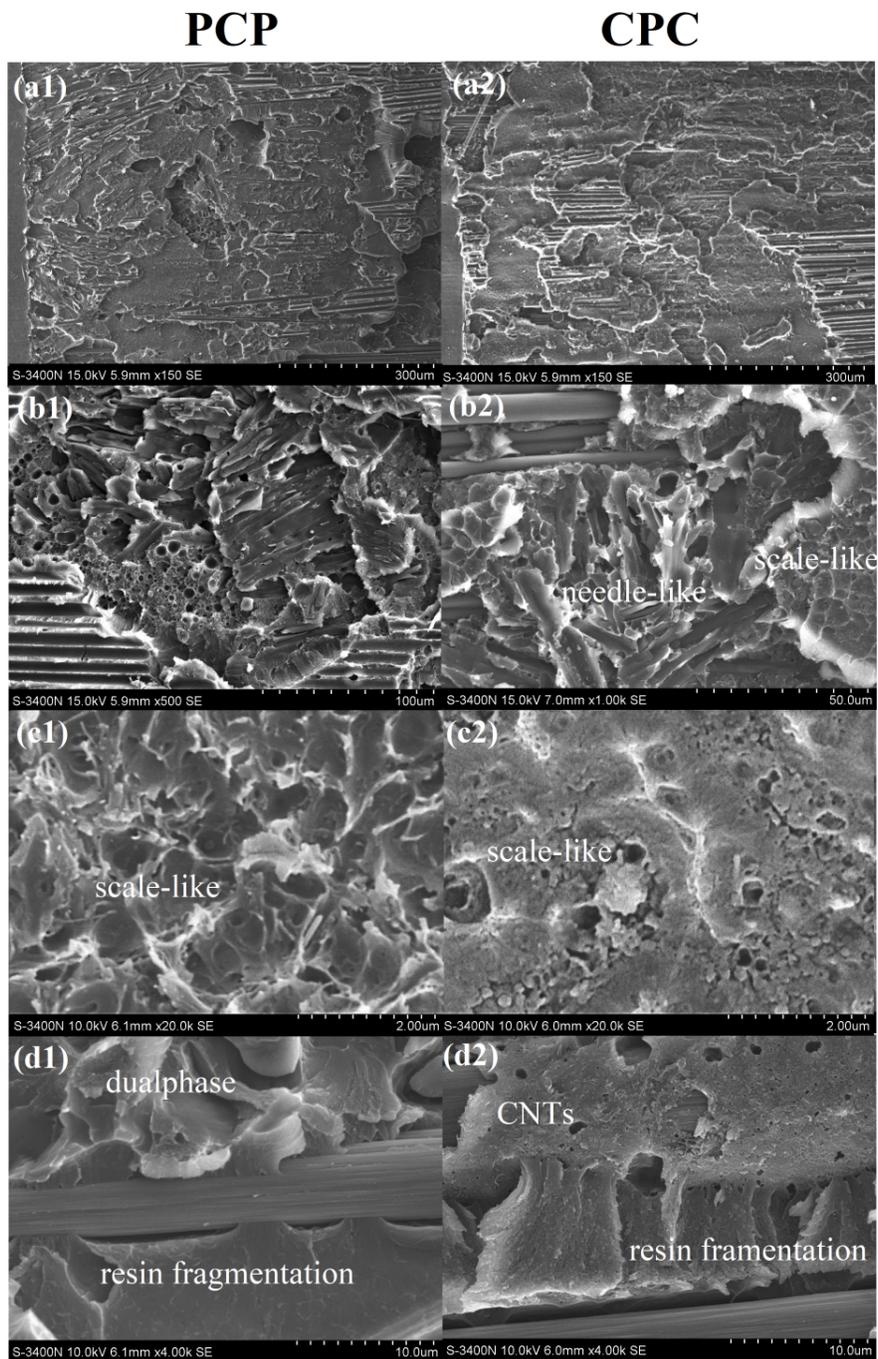


Fig.9 Fracture surface of the composites interleaved by PCP and CPC sandwich films after G_{IC} test: (a1)-(d1) PCP films, (a2)-(d2) CPC films

The morphology characterization was conducted on the etched “scale-like” structures of the interleaved composites by PC hybrid film, PCP and CPC sandwich films to identify the difference, which was showed in Fig.10. Before and after etching chemically, the enlarged “scale-like” structures were completely different. The interconnected grooves were found on the surface of the PCP film interleaved composite, exhibiting the PEK-C/EP co-continuous feature[28]. The dense epoxy particles were exposed on the surface of the

CPC interleaved composites, suggesting that a nodular phase-inverted morphology occurred [29]. For the PC interleaved composite, the smooth epoxy surface and the concentration of CNTs could be seen, which confirmed the “scale-like” monophasic PEK-C structure [22].

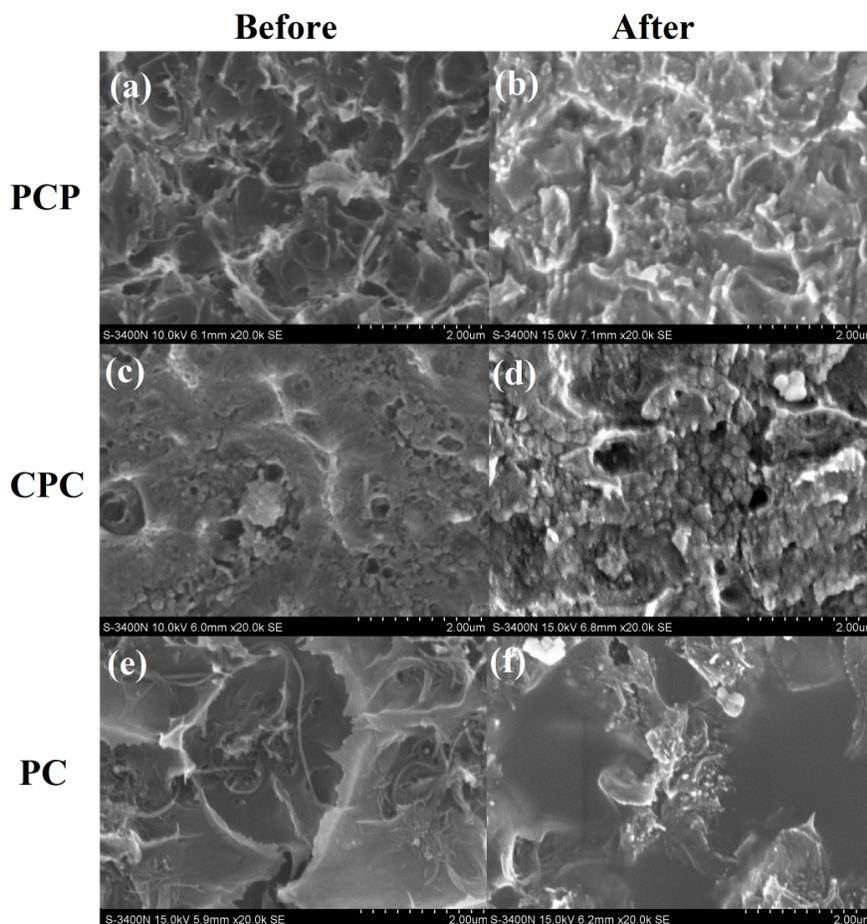


Fig.10 Surface morphology of the etched “scale-like” structures of the interleaved composites by PC hybrid film, PCP and CPC sandwich films: (a)-(b) PCP film, (c)-(d) CPC film and (e)-(f) PC film

3.3 Discussion

The different hybrid structure of interlayer produced the various microstructure in the interlaminar region, leading to the change of crack propagation and fracture toughness under the tension state. Fig.11 summarized the schematic of the influence of interlayer hybrid structure on the interlaminar layered structure formed during curing process and the crack propagation path in G_{IC} tests based on the previous topography study. The cracks extended along the interface between epoxy matrix and carbon fibers or in the epoxy matrix for the black composites, leaving the smooth fragile fracture surface due to the intrinsic brittleness of epoxy resin (Fig.11(a)). From the Fig.11(b), the epoxy matrix in the interlaminar region was well reinforced by CNTs, resulting in that the cracks propagated only through the interface and many fibers and small CNTs were exposed on the fracture surface. The CNTs played little role in toughening, which might be the main reason for the small improvement in fracture toughness of the CNT interleaved composites.

The layered structure and the tortuous crack propagation could be found from the Fig.11(c)-(f). The characteristic layered structure, consisting of homogeneous PEK-C resin, nodular dualphase and sea-island

dualphase, was formed in the interlaminar region of the PEK-C interleaved composites. The interdiffusion of resin, curing reaction of epoxy and PEK-C/EP phase separation occurred simultaneously under high temperature until the gel point, leading to the formation of different phase structure depending on the concentration of PEK-C [22]. The crack propagated along the homogeneous PEK-C layer and the interface between the interlaminar region and the fiber/epoxy ply, traversing the nodular dualphase structures, which resulted in the exposed fibers, grooves and “scale-like” PEK-C monophase on the fracture surface (Fig.11(c)).

The PEK-C/CNTs hybrid film produced the similar layered structure to the neat PEK-C film. However, more “scale-like” monophase was exhibited on the fracture surface with respect to the PEK-C interleaved composite. The crack tended to advance along the path with low resistance, so the “scale-like” PEK-C monophase with CNTs might exhibit lower toughening efficiency due to the centralized CNTs.

The composites interleaved by PCP and CPC sandwich interlayers formed more complex multilayered structure in the interlaminar region due to the sandwich structure of interlayers. For the PCP interleaved composite, the dualphase structure surrounding the fibers, “scale-like” dualphase with CNTs and the “needle-like” epoxy with CNTs was formed and for the CPC interleaved composites, the CNTs reinforced epoxy surrounding the fibers, the “needle-like” epoxy with CNTs and “scale-like” dualphase with CNTs were formed, which was due to the interdiffusion and the phase separation of resin during curing process. The complex layered structure led to the most tortuous crack path and the best fracture toughness.

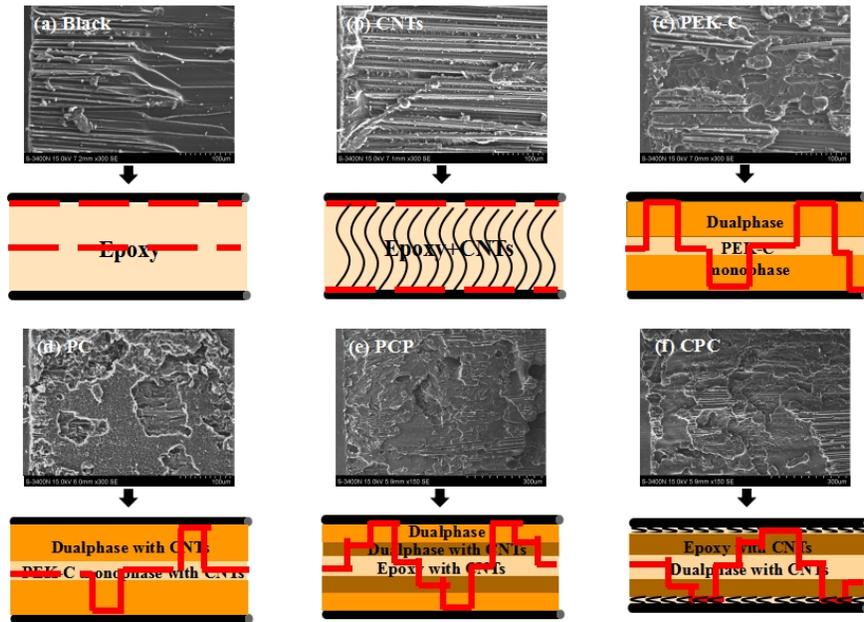


Fig.11 Schematic of the interlaminar layered structure and the crack propagation of blank and all the interleaved composites

Conclusion

The different layered structure in the interlaminar region was established in the carbon fiber reinforced epoxy composite by inserting the CNTs/PEK-C interlayer with different hybrid structures, resulting in the different toughening efficiency.

The DCB tests showed that the mode I interlaminar fracture toughness (G_{IC}) for the laminates interleaved by neat PEK-C film increased by 101.03% due to the phase separation, whereas G_{IC} of the CNTs interleaved laminates was only 10.79% higher than that of black composites due to the fact that the crack propagated

along the interface between the interlaminar region and ply and the CNTs played little role. The synergistic combination of CNTs and PEK-C improved also the fracture toughness. PEK-C/CNT/PEK-C sandwich films produced the best toughening effect and the G_{IC} and G_{IR} were 446.76 J/m² and 451.04 J/m², increased by 138.11% and 127.30%, respectively compared to blank composites, benefiting from the multilayered structure in the interlaminar region and the resultant crooked crack propagation.

Data availability

The raw data required to reproduce these findings are available to download from [DOI: 10.17632/mfb366wv6x/1].

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Authorship contribution statement

Jiawei YAO: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Writing—original draft, Writing—review & editing.

Hongfu LI: Methodology, Project administration, Supervision, Validation, Writing—review & editing.

Ying WU: Conceptualization, Funding acquisition, Supervision, Writing—review & editing.

Kangmin NIU: Funding acquisition, Supervision, Writing—review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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