Effect of carbon nanotubes on the electrical, thermal, mechanical properties and crystallization behavior of continuous carbon fiber reinforced polyether-ether-ketone composites

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Abstract
The continuous carbon fiber reinforced Polyether-ether-ketone (PEEK) prepreg tapes with the addition of carbon nanotubes (CNTs) were prepared by a wet powder impregnation process. Their electrical conductivity, thermal conductivity, the tensile properties, dynamic mechanical behavior, fracture morphology and crystallization melting behavior were investigated. The results show that, the electrical conductivity ($\sigma$), thermal conductivity ($\lambda$), tensile strength ($\sigma_t$) and interfacial adhesion of the prepreg tapes were obviously improved with the addition of CNTs. When CNT content was 1.0 wt%, the $\sigma$ of CNT/CCF/PEEK prepreg tapes in the $0^\circ$ direction reached a maximum value of 0.701 s cm$^{-1}$, which was increased 165% than that of CCF/PEEK prepreg tapes. The $\lambda$ reached 1.053 W m$^{-1}$ K$^{-1}$, which was improved by 12.14%. The tensile strength was 1489 MPa, which was increased by 16.4%. The results of SEM images show that the interface adhesion between the fiber and the matrix is good. The results of DMA indicate that, the capability of deformation resistance of the prepreg tapes were further improved with the addition of CNTs. When the temperature was 280 °C, the decline rate of $E'$ of 1.0 wt% CNT/CCF/PEEK prepreg tapes were 3.58%. The results of DSC indicate that, the $T_c$ of CNT/CCF/PEEK prepreg tapes moved to high temperature after adding CNTs, which indicate that CNTs played a role of heterogeneous nucleation in the PEEK matrix.

1. Introduction

In recent decades, with the improvement of properties and manufacturing process of fiber reinforced polymer composites, they have been widely used in many fields, e.g., aerospace, automotive and energy[1–3]. The bulk of such structures use thermoset composites despite the excellent mechanical and thermal properties exhibited by composites based on high-performance engineering thermoplastic polymers. Presently, it has attracted great interest in fiber reinforced thermoplastic composites based on polyaryletherketones (PAEKs) with excellent mechanical properties, high temperature resistance, corrosion resistance and so on, which applied in automotive, aerospace and the energy sector, both in alternative energy, e.g. wind turbine blades, and conventional energy generation, e.g., underground oil-drilling[4, 5]. Polyether ketone (PEEK) is the most widely applied in high pressure, high temperature in the PAEK family. However, the poor interfacial adhesion of fiber fiber reinforced thermoplastic composites restricts its application under some harsh conditions. To address this problem, it was developed a new technology for enhancing the interfacial bonding of fiber reinforced thermoplastic composites, for example, the modification of matrix[6, 7], the ozone or plasma treatment on fiber[8–10], surfaces the introduction nano particles[11, 12], increasing the fiber surface grafting[13] and other multiscale modification methods. Not all of the above methods are suitable for industrial production, some of which are only used in laboratory.

Incorporation of nanoparticles into composite materials can improve surface roughness and crack growth resistance. Carbon nanotubes (CNTs) have excellent mechanical, electrical and thermal properties, which make
them promising candidates to be used as nanofillers in polymer composites, opening up new perspectives for the development of the next generation of high-performance multifunctional materials. Su YN et al\textsuperscript{[14]} prepared CNTs/CF/PEEK multiscale composites with different rCNTs content by prepreg spraying method. And studied the mechanical performance found that the ILSS, flexural strength and flexural modulus of CNT/CF/PEEK composites were improved by 35.8%, 25.4% and 23.7% after introducing 0.5 wt% CNTs, respectively. Díez-Pascual AM et al\textsuperscript{[15]} studied mechanical and thermal properties of polyether ether ketone (PEEK)/single-walled carbon nanotube (SWCNTs) glass fiber laminates incorporating polysulfone as a compatibilizing agent. The results showed that significant enhancements thermal conductivity, significant enhancements in stiffness, strength and toughness by the incorporation of single-walled carbon nanotubes (SWCNTs) wrapped in the compatibilizer. Bekyarova et al\textsuperscript{[16]} investigated mechanical properties of the carbon nanotube/carbon fiber preforms in infiltrated with epoxy resin using the vacuum-assisted resin transfer molding method. The interlaminar shear strength was increased by 27% when the introduction of 0.25% of MWNTs in the CF/epoxy composites. Shen et al\textsuperscript{[17]} studies the effects of incorporating carbon nanotubes (CNT) into polyamide-6 (PA6) on mechanical, thermal properties and fire performance of woven glass reinforced CNT/PA6 nanocomposite laminates. Incorporation of up to 2.0 wt% CNT in CNT/PA6/GF laminates improved the flexural stress of the laminates up to 36%, the thermal conductivity by approximately 42%. These studies indicate that CNTs have great potential in the development of composite.

Obviously, there are less research on CNTs in CCF/PEEK composites at present. In order to meet the application demanding, the multifunctional properties and high performance of CCF/PEEK composites need to be investigated.

In the present work, the carbon nanotube/continuous carbon fibers reinforced polyether-ether-ketone (CNT/CCF/PEEK) prepreg tapes were prepared by wet powder impregnation process\textsuperscript{[18]}. Mainwhile, the effects of CNTs content on electrical conductivity, thermal conductivity, the tensile properties, dynamic mechanical behavior, crystallization and melting behavior of CNT/CF/PEEK prepreg tapes were studied. Their fracture morphologies are also observed by scanning electron microscope (SEM).

2. Experimental

2.1. Materials

PEEK powder (330UPF-650) was produced by Jilin Joinature Polymer Co., Ltd, China ($T_g = 143 \, ^\circ C$, $T_m = 343 \, ^\circ C$, $d_{50^{\circ}C} = 1.3 \, g \, cm^{-3}$, D50 = 25 $\mu m$). Carbon fibers (T700SC–12K–50C) were produced by Toray Industries, Inc., Japan. Its density is 1.80 g cm$^{-3}$, and consists of bundles of micro fibers (tows) with a single fiber diameter of 7 $\mu m$. CNTs (GT-210) with a diameter of approximately 10–20 nm with length of 5–50 $\mu m$ were supplied by Shandong Dazhan nanomaterials Co., Ltd, China.

2.2. Preparation of prepreg tapes

The preparation process of CNT/CCF/PEEK prepreg tapes was shown in figure 1. Firstly, PEEK powder was dispersed into alcohol by magnetic stirring for 4 h. Weighed a certain amount of CNTs (0.5 wt%, 1.0 wt%, 1.5 wt%) and dispersed it into alcohol by ultrasonic dispersion for 1 h and magnetic stirring for 4 h. The polyvinyl pyrrolidone of 2 wt% was added to as dispersant. The CNTs dispersions were well maintained. Secondly, the CNTs dispersion was dispersed in PEEK powder suspension and mechanically stirred for 4 h. The CNT/PEEK suspension were prepared by solution blending, and the CNT/PEEK dispersion was well maintained. Finally,
the CNT/PEEK dispersion was put into the dip-coating tank. By adjusting the tension, the CCF bundles were expanded in the dip-coating tank, which made PEEK and CNTs more easily attach to the surface of the CCF. Then, the CNT/CCF/PEEK prepreg tapes were prepared by calendering the fiber bundles through a self-designed preheated melting mould.

2.3. Measurements
2.3.1. Electrical conductivity
The electrical conductivity of CNT/CCF/PEEK prepreg tapes were analyzed with a FT-8100A four-probe powder conductivity measuring instrument manufactured by Ningbo Rlco instrument technology Co., Ltd, China. The prepreg tapes were cut into rectangular sheet with size of 2 mm × 2 mm and the surface were washed by absolute ethyl alcohol before testing.

2.3.2. Thermal conductivity
The thermal conductivity were measured at room temperature by a Model TC3200 instrument of measuring the thermal conductivity manufactured by Xi’an Xiatech Electronics Co., Ltd, China. The specimen dimensions were 20 mm × 50 mm. At least 5 readings for each specimen were tested to report an average value.

2.3.3. Tensile tests
The tensile property was performed by an INSTRON 5969 universal material testing machine. The tensile specimens of prepreg tapes were prepared based on ASTM D3039/D3039M-14, and the specimen dimensions were 250 mm × 15 mm × 0.1 mm and the stretching speed was 10 mm min⁻¹.

2.3.4. Microscopy analysis
The surface morphology of tensile-fractured specimens was observed using Quanta 250 scanning electron microscope manufactured by FEI Company of the United States. The tensile-fractured specimens were coated with Au/Pt overlayer.

2.3.5. Dynamic mechanical analysis
Dynamic Mechanical Analysis (DMA) of the the PEEK and CNT/CCF/PEEK prepreg tapes were conducted by a Q800-DMA instrument manufactured by TA Instruments, USA. The samples were performed in tensile mode at a frequency of 1.0 Hz and a heating rate was 5 °C min⁻¹ within the temperature range of 50 °C–320 °C under nitrogen atmosphere.

Retention rate of thermal modulus ($R_T$) was calculated from the equation below (1):

$$ R_T = \frac{E'_{T}}{E'_{T}} \times 100\% $$

$E'_{T}$: the storage modulus at different temperatures, $E'_{T}$: the storage modulus onset temperature (T: 50 °C).
2.3.6. Differential scanning calorimetry
Differential Scanning Calorimetry (DSC) tests were conducted by a Q2000-DSC manufactured by TA Instruments, USA. The samples were heated from room temperature to 400 °C at a heating rate of 20 °C/min under nitrogen atmosphere, and kept it for 2 min to remove the thermal history, ensuring that the sample preparation process had no influence on the study of crystallization behavior, then cooled it down to room temperature at a rate of 10 °C/min. Then cooled it to room temperature at a rate of 10 °C/min, and the crystallization temperature was obtained by cooling scanning, after then the sample was reheated to 400 °C at a rate of 10 °C/min to obtain the melting temperature.

3. Results and discussion
3.1. Electrical conductivity
The electrical properties of unidirectional continuous carbon fiber reinforced composites exhibit strong anisotropy [19]. As an excellent conductor, CNTs can greatly improve the electrical conductivity of the composites, and also improve the transverse conductivity [20, 21]. Figure 2 presents the relationship between the electrical conductivity (σ) of CNT/CCF/PEEK prepreg tapes in 0° and 90° direction and the addition content of CNTs. As shown, the σ of CNT/CCF/PEEK prepreg tapes increases obviously as compared to CCF/PEEK prepreg tapes. It is the formation of an interconnected network in PEEK matrix with the introduction of CNTs into PEEK matrix, which are conducive to electron transport leading to increasing the electrical conductivity of prepreg tapes. The σ of CNT/CCF/PEEK prepreg tapes increased first and then decreased slightly with the CNTs content increasing. When the CNTs content was 1.0 wt%, the σ of CNT/CCF/PEEK prepreg tapes in 0° and 90° direction were 0.701 s cm⁻¹ and 0.1934 s cm⁻¹, which were improved by 165% and 92% in comparison to the CCF/PEEK prepreg tapes, respectively. This is due to the fact that CNTs are dispersed in PEEK matrix and form a conductive path with carbon fibers, which promotes the electron transport and greatly improves the electrical conductivity of the materials. In 90° direction, the distance between two adjacent fibers were far away and the surface of fiber wrapped with substantial PEEK matrix, which might hinder the electron transport. The σ of CNT/CCF/PEEK prepreg tapes decreased slightly when the CNTs content was 1.5 wt%. This is due to the presence of CNTs aggregates in polymer matrix, which can not give full play to the excellent electrical properties of CNTs, and may hinder the diffusion of matrix. It lead to the formation of conductive path defects, which is not conducive to electron transport, thus reducing the overall conductivity. Therefore, the appropriate incorporation of CNTs can fully play its excellent conductivity, and form a good conductive network, thereby improving the electrical properties of the composite.

3.2. Thermal conductivity
Figure 3 shows thermal conductivity (λ) of CNT/CCF/PEEK prepreg tapes with different CNTs content. The λ of CNT/CCF/PEEK prepreg tapes increased first with increasing of CNTs content and then decreased. When the content of CNTs was 1.0 wt%, the λ was 1.053 W m⁻¹ K⁻¹, which was 12.14% higher than that of CCF/PEEK prepreg. The λ of CNT/CCF/PEEK prepreg tapes had decline trend when the addition content of CNTs was more than 1.0 wt%. It is affected by three factors such as the content of reinforcement, the PEEK matrix and interfacial adhesion. Owing to the wide distribution of the molecular weight and the uneven chain segment...
length of PEEK matrix, the formation easy of amorphous areas within the material, it is difficult to form a complete crystal structure and is not conducive to heat transfer[22]. When thermal conduction network structure was formed between CCF and PEEK, the heat transfer was restricted to a certain extent by irregular interlinking and winding between segments. CNTs exhibits ballistic transport properties and good thermal conductivity[23], so they are dispersed homogeneously in PEEK matrix, which is conducive to heat transfer. The network structure formed between CNTs, CCF and PEEK in the composites was gradually improved with increasing CNTs content within 0.5–1.0 wt%. which was propitious to heat transfer and improving the \( \lambda \) of prepreg tapes. When CNTs content was 1.5 wt%, CNTs was difficult to disperse in PEEK matrix and even aggregates appear, resulting in a slight decrease in the \( \lambda \) of CNT/CCF/PEEK composite.

3.3. Mechanical properties

3.3.1. Tensile property

The tensile strength of CNT/CCF/PEEK prepreg tapes with different CNTs content was shown in figure 4. The tensile strength of CCF/PEEK prepreg tapes was 1279 MPa. The addition of CNT further increases the tensile strength of the composites. The tensile strength of CNT/CCF/PEEK prepreg with CNT content of 0.5 wt% and 1.0 wt% is 1335 MPa and 1489 MPa respectively improved by 4.38% and 16.42% comparing with CCF/PEEK prepreg tapes, respectively. CNT occupy a small volume fraction in the composites, but they can change the macro mechanical properties, disperse in peek matrix to play a reinforcing role[24], and enhance the interface bonding between CCF and matrix. Therefore, when the composite is subjected to external load, the load transfer between matrix and reinforcement phase is enhanced, and the tensile strength is improved. On the other hand, the tensile strength of CNT/CCF/PEEK prepreg reaches the maximum value with the increase of CNT content and then decreases. When the content of CNTs was 1.5%, the tensile strength of CNT/CCF/PEEK prepreg was 1158 MPa, which was lower than that of CCF/PEEK prepreg. This may be due to the aggregation of CNTs on peek matrix. It is known that the aggregation will significantly affect the interfacial bonding strength of the composites[25]. Due to the aggregation of carbon nanotubes on peek matrix, the internal defects of the prepreg increase, which leads to the destruction of the prepreg and the decrease of the tensile strength.

3.3.2. Fracture morphology of tensile specimens

To further confirm the effect of the CNTs content on the tensile strength of CNT/CCF/PEEK prepreg tapes. The fracture morphology of fabricated tensile samples of prepreg tapes with different CNTs content is displayed in figure 5. As shown in figure 5(a), (b), there were some voids between fibers and PEEK matrix, and the surfave of fiber is smooth, which illustrates the interfacial adhesion between fibers and PEEK matrix was poor resulting in decreasing the tensile strength of CCF/PEEK prepreg tapes. As can be seen from figure 5(c), (d), when CNTs content was 0.5 wt%, a small amount of carbon fibers were pulled from the PEEK matrix on the fracture surface. The surface of carbon fiber was wrapped with a little matrix. It indicates that the introduction of CCF into the PEEK matrix is propitious to improve the interfacial adhesion between fibers and PEEK matrix. As the CNTs concentration increases, figure 5(e), (f) clearly shown that the surface of protruding carbon fiber was wrapped with substantial matrix. The fibers pulled out from matrix took a lot of energy during the tensile process, which increased the energy required for the fracture of prepreg tapes. This is one of the important reasons for the higher tensile strength of the prepreg tapes. As shown in figure 5(g), (h), the fractured surface of prepreg tapes was regular and no fiber was pulled out from the matrix, which indicates the interfacial adhesion...
between fibers and PEEK matrix is good. However, the excess addition of CNTs in matrix might agglomerate in the interfacial regions. When it is subjected to tensile stress, the defects are formed in the agglomeration regions and then fractured, resulting in the decrease of the tensile strength of prepreg tapes [26].

3.3.3. Dynamic mechanical analysis

The curves of storage modulus ($E'$) with temperature for pure PEEK and CNT/CCF/PEEK prepreg tapes are shown in figure 6. The $E'$ of pure PEEK at the initial temperature is 3.7 GPa. After adding carbon fiber, the $E'$ of CCF/PEEK prepreg tape increased significantly, reaching 76.6 GPa, which was an 18-fold increase compared with pure PEEK. The prepreg tapes were subjected to alternating stress, which was effectively transferred to CCF
through the matrix, indicating that the stress of the prepreg tapes was mainly borne by fiber. The $E'$ of CCF/PEEK prepreg tapes increased first and then decreased with the addition of CNTs. The $E'$ of 0.5 wt%, 1.0 wt% CNT/CCF/PEEK prepreg tapes are 92.45 GPa, 104.3 GPa increases by 21.08%, 36.16% as compared to that of CCF/PEEK prepreg tapes, respectively. CNTs are uniformly incorporated into PEEK matrix to improve the interfacial adhesion fiber-matrix. The $E'$ of 1.5 wt% CNT/CCF/PEEK prepreg tapes is 97.4 GPa decreases by 7% than that of 1.0 wt% CNT/CCF/PEEK prepreg tapes, probably due to agglomeration of CNTs at PEEK matrix,
which hinders the stress transfer when the prepreg tape is subjected to alternating stress. These results are consistent with the trend of tensile properties.

Retention rate of thermal modulus ($R_T$) versus temperature are presented in figure 7. $R_T$ refers to the ratio of $E'_T$ ($E'$ at different temperatures) to $E'_R$ ($E'$ at the initial temperature), a measure of variation of storage modulus with temperature. It can be seen that, $R_T$ of pure PEEK declines sharply and $R_T$ of prepreg tapes decreases slowly, indicating that the deformation resistance of prepreg tapes is improved. When the temperature is 250 °C, the $R_T$ of pure PEEK is only 10.09%, which exhibits the deformation resistance of prepreg tapes is poor. After addition of the CCF, the $R_{250}$ significantly increased, reached 86.56%. More importantly, the $R_{250}$ of CCF/PEEK composites containing 0.5 wt%, 1.0 wt% and 1.5 wt% CNT reaches 90.19%, 96.42% and 94.64% respectively, which means that the addition of CNT further improves the deformation resistance of CCF/PEEK composites. Table 1 lists the $R_T$ of CF/PEEK composites compared with other modified composites, it can be seen that the $E'$ and $R_T$ of the composites in this study are higher, especially at 250 °C. There is still a higher modulus retention rate, indicating that the prepared composites have stronger resistance to deformation, and have greater long-term use potential at high temperature (250 °C). The high thermal modulus retention is closely related to the content and surface modification of nano carbon materials, carbon fiber volume fraction and molding process, so as to give full play to the best performance of CF/PEEK composites. This comparison also shows the potential of nano carbon materials in improving the thermal properties of CF/PEEK composites.

Figure 8 shows the evolution of loss modulus ($\tan \delta$) as a function of temperature of pure PEEK and CNT/CCF/PEEK composites. Compared with the pure PEEK, the $\tan \delta$ of CCF/PEEK and CNT/CCF/PEEK prepreg tapes is significantly increased. This is because the movement of the PEEK segment is blocked when the prepreg tape is under alternating stress. The movement of the chain segment cannot keep up with the change of stress, and more energy is lost in the form of heat energy. The $\tan \delta$ of CNT/PEEK prepreg tapes is lower than that of CCF/PEEK prepreg tapes. The addition of CNTs improves the interfacial adhesion between PEEK matrix and CF and the stress transformation ability. Meanwhile, the energy dissipated in the form of heat energy decreases being subject to external forces.

The evolution of $\tan \delta$ versus temperature is shown in figure 9. It exhibits the strongest peak corresponding to the glass temperatures ($T_g$), these data are listed in table 2. The inclusion of fiber hindered the movement of the polymer chain segment, which led to the $T_g$ of CCF/PEEK prepreg tapes moving towards high temperature comparing with pure PEEK. Meanwhile, compared with CCF/PEEK prepreg, the $T_g$ of CNT/CCF/PEEK prepreg gradually moves to high temperature with the addition of CNT. The $T_g$ of prepreg tapes moves towards high temperature probably arose from several factors. Firstly, adding fillers reduces the free volume and restricts the mobility of PEEK chains. Secondly, the presence of the CNTs, which is dimensionally similar to the PEEK

Table 1. Retention rate of thermal modulus of CF/PEEK composites reported in published works.

| Composites | Modifications | Molding Process | $E'_R$ (GPa) | $R_{250}$  | $R_{250}$ References |
|------------|---------------|------------------|--------------|------------|-----------------------|
| CCF/PEEK   | CNT/PVP       | Wet powder impregnation process | 76–104 (max) | 97.69%     | 96.42% This study     |
| CF/PEEK    | GO/PEI        | Hot-press processing | 39–47 (max)  | 68.07%     | 55.29% [27]           |
| CF/PEEK    | Activated-CF  | Hot-press processing | 7–22 (max)   | 39.29%     | 37.22% [6]            |
| SCF/PEEK   | GNP           | Injection molding | 6–8 (max)    | 44.08%     | 31.77% [28]           |
segment units, more efficiently hindered the chain motion [15]. The $T_g$ of 1.5 wt% CNT/CCF/PEEK prepreg tapes moved to low temperature than 1.0 wt% CNT/CCF/PEEK prepreg tapes. The result indicates that PEEK matrix has some CNTs aggregates, which alters the flow behavior of the matrix during impregnation, leading to increasing the free volume of the PEEK molecules and the mobility of polymer molecular chain segments [26]. The $\tan\delta$ peak value of pure PEEK was 0.133. The $\tan\delta$ peak value of CCF/PEEK prepreg tapes decreased as carbon fibers added. It indicates that the addition of CCF hinders the movement of PEEK segments. The decrease of $\tan\delta$ peak value is more obvious after adding different content of CNTs, and because of they hinder the movement of the polymer chain segments, which may can explain their lower $\tan\delta$ peak value, indicating that the mechanical loss of overcoming friction between molecular chains decreases after adding CNTs.

3.3.4. Crystallization and Melting Behavior

Crystallization behavior plays an important role in the properties of semicrystalline polymers. Actually, there are two factors controlling the crystallization of polymeric composite systems. One is that fillers may have heterogeneous nucleation, which results in higher $T_c$. The other is that they hinder the diffusion and migration of polymer chains, which inhibits crystallization [29–31]. Figure 10 displays the non-isothermal crystallization curves of CNT/CCF/PEEK prepreg tapes with different CNTs content. Corresponding thermal parameters are summarized in table 3. It can be observed that the $T_c$ of prepreg tapes moves to lower temperature comparing with pure PEEK. It can be attributed to the distance between two adjacent fibers was smaller when the fiber

![Graphical representation of crystallization behavior](image-url)
content was 60 wt%, which hindered the movement of the polymer chain segment and constrains the spherulites growth. The $T_m$ of CNT/CCF/PEEK prepreg tapes moved to higher temperature with the introduction of CNTs into PEEK matrix. It occurred increased first and then decreased with the increase of the content of CNTs. When CNTs content was 0.5 wt%, CNTs played a role of heterogeneous nucleation in the PEEK matrix leading to $T_m$ moving towards higher temperature. When the CNTs content was further increased to 1.0 wt%, although the nucleation surface became larger under the heterogeneous nucleation of CNTs, the formation of network structure between CNTs, CF and PEEK impeded the crystal nucleus growth resulting in $T_m$ moving towards lower temperature. When CNTs content was 1.5 wt%, some aggregates of CNTs dispersed in PEEK matrix hindering the movement of the polymer chain segment leading to $T_m$ moving further towards lower temperature. A similar crystallization inhibition behavior of PEEK has been reported in glass fiber/PEEK composites, which is due to the stronger interaction between nanotubes and polymers [15]. In Table 3, it can find that the $X_0$ of CNT/CCF/PEEK prepreg tapes is higher than CCF/PEEK prepreg tapes, which proves that the addition of CNTs promoted the crystallization of PEEK. The melting curves of prepreg tapes are shown in figure 10(b). It can be seen that the $T_m$ changes little with the inclusion of CF or CNTs, which indicates that the addition of CNT and CF has little affect on the melting behavior of the polymer [15, 32, 33].

4. Conclusions

In this work, the continuous carbon fibers reinforced Polyether-ether-ketone prepreg tapes with the addition of carbon nanotubes were prepared by a wet powder impregnation process, which exhibited excellent electrical conductivity, thermal conductivity and mechanical properties. The proper content of CNTs was homogeneously dispersed in PEEK matrix, and the network structure formed between CNTs, CCF and PEEK in the composites, which improved the properties of the composites. It was also found that CNTs agglomeration appeared with the increase of CNTs content. The tensile strength increased from 1279 MPa to 1489 MPa for CNT/CCF/PEEK prepreg tapes with CNTs content of 1 wt%. DMA results demonstrated significant enhancements in storage modulus and glass transition temperature of CNT/CCF/PEEK prepreg tapes as compared to that of CCF/PEEK prepreg tapes. The results of DSC indicated that, CNTs played a role of heterogeneous nucleation in the PEEK matrix, but had less influence on the melting behavior of PEEK. Overall, the CNTs were efficient in reinforcing thermoplastic/carbon fiber composites, and can be used to prepare high-performance multifunctional composites, which should have great potential for advanced engineering applications.

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Conflicts of interest

No author has a financial/commercial Conflict of Interest.

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References

[1] Barile C, Casavola C and De Cillis F 2019 Mechanical comparison of new composite materials for aerospace applications Comps Part B-Eng 162 122–8
[2] Brooks R, Ramanan S M S and Arun S 2017 Composites in automotive applications: design In:Reference Module in Materials Science and Materials Engineering, (Amsterdam: Elsevier)
[3] Brøndsted P, Lilholt H and Lystrup A 2005 Composite materials for wind power turbine blades Annu. Rev. Mater. Res. 35 505–38
[4] Barbosa L C M et al 2018 Fractographic study of welded joints of carbon fiber/PPS composites tested in lap shear Eng. Fail. Anal. 93 172–82
[5] Veazey D, Hsu T and Gomez E D 2017 Next generation high-performance carbon fiber thermoplastic composites based on polyaryletherketones J. Appl. Polym. Sci. 134 44441–57
[6] Hassan E A M et al 2018 Highly boosting the interlaminar shear strength of CF/PEEK composites via introduction of PEKK onto activated CF Comps Part A-Appp S 112 155–60
[7] Sorrentino L et al 2015 Mechanical performance optimization through interface strength gradation in PP/glass fibre reinforced composites Comps Part B-Eng 76 201–8
[8] Li J 2010 Interfacial studies on the ozone and air-oxidation-modified carbon fiber reinforced PEEK composites Surf. Interface Anal. 41 310–5
[9] Luo H et al 2014 Air DBD plasma treatment on three-dimensional braided carbon fiber-reinforced PEEK composites for enhancement of in vitro, bioactivity Surf Coat Tech 242 1–7
[10] Jang J and Kim H 2010 Improvement of carbon fiber/PEEK hybrid fabric composites using plasma treatment Polym. Compos. 18 125–32
[11] Su Y N et al 2017 Preparation and properties of graphene/carbon fiber/poly (ether ether ketone) composites New Carbon Mater. 32 152–9
[12] Ming Y et al 2017 The interlaminar shear strength and tribological properties of PA 6 composites filled with graphene oxide-treated carbon fiber Surf. Interface Anal. 49 755–8
[13] Shiba K et al 2014 Effective surface functionalization of carbon fibers for fiber/polymer composites with tailor-made interfaces Chemplaschem 79 197–210
[14] Su Y N et al 2018 Preparation and properties of carbon nanotubes/carbon fiber/poly (ether ether ketone) multiscale composites Compos Part A-App 710889–98
[15] Díez-Pascual A M et al 2011 Influence of carbon nanotubes on the thermal, electrical and mechanical properties of poly (ether ether ketone)/glass fiber laminates Carbon 49 2817–33
[16] Bekyarova E et al 2007 Multiscale carbon nanotube-carbon fiber reinforcement for advanced epoxy composites Langmuir 23 3970–4
[17] Shen Z Q et al 2009 The effects of carbon nanotubes on mechanical and thermal properties of woven glass fibre reinforced polyamide-6 nanocomposites Compos. Sci. Technol. 69 239–44
[18] Zhu K L et al 2019 Crystallization and mechanical properties of continuous carbon fiber reinforced polyether-ether-ketone composites Fibers Polym. 20 859–66
[19] Li Y and Ma M 2018 Study on the Electrical Properties of CNT/Carbon fiber reinforced composite Mater. Sci. Forum 813 315–22
[20] Deng F and Zheng Q S 2008 An analytical model of effective electrical conductivity of carbon nanotube composites Appl. Phys. Lett. 92 071902
[21] Yan K Y et al 2007 The interface effect of the effective electrical conductivity of carbon nanotube composites Nanotechnology 18 255705
[22] White C T and Todorov T N 1998 Carbon nanotubes as long ballistic conductors Nature 393 240–2
[23] Panzer M A et al 2008 Thermal properties of metal-coated vertically aligned single-wall nanotube arrays J. Heat Transfer 130 1306–13
[24] Kanagaraj S et al 2007 Mechanical properties of high density polyethylene/carbon nanotube composites Compos. Sci. Technol. 67 3071–7
[25] Papageorgiou D G, Kinloch I A and Robert J Y 2015 Graphene/elastomer nanocomposites Carbon 95 460–84
[26] Díez-Pascual A M et al 2009 Development and characterization of PEEK/carbon nanotube composites Carbon 47 3079–90
[27] Chen J, Wang K and Zhao Y 2018 Enhanced interfacial interactions of carbon fiber reinforced PEEK composites by regulating PEI and graphene oxide complex sizing at the interface Compos. Sci. Technol. 154 175–86
[28] Papageorgiou D G et al 2019 Hybrid poly(ether ether ketone) composites reinforced with a combination of carbon fibres and graphene nanoplatelets Compos. Sci. Technol. 175 60–8
[29] Fan Z, Hsiao K T and Advani S G 2004 Experimental investigation of dispersion during flow of multi-walled carbon nanotube/polymer suspension in fibrous porous media Carbon 42 871–9
[30] Velisaris C N and Seferis J C 1986 Crystallization kinetics of polyetheretherketone (peek) matrices Polym. Eng. Sci. 26 1574–81
[31] Lee Y and Porter R S 2010 Crystallization of poly(etheretherketone) (PEEK) in carbon fiber composites Polymer Eng Sci 26 633–9
[32] Lyu H et al 2021 Enhanced interfacial and mechanical properties of carbon fiber/PEEK composites by hydroxylated PEEK and carbon nanotubes Comps Part A-App 8 1106364
[33] Karśli N G and Aytac A 2013 Tensile and thermomechanical properties of short carbon fiber reinforced polyamide 6 composites Comps Part B-Eng 51 270–5