Efficient Thermal Transport Highway Construction Within Epoxy Matrix via Hybrid Carbon Fibers and Alumina Particles

Hao Wang,†‡§ Linhong Li,†‡§ Yapeng Chen,‡§ Meng Li,‡ Hui Fu,‡ Xiao Hou,‡× Xinfeng Wu,‡×∥ Yaping Chen,‡§ Meng Li,‡ Linhong Li,†‡§ Nan Jiang,‡§∥ and Jinhong Yu‡×§

†Nano Science and Technology Institute, University of Science and Technology of China, Suzhou 215123, China
‡Key Laboratory of Marine Materials and Related Technologies, Zhejiang Key Laboratory of Marine Materials and Protective Technologies, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China
§Polymer Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
∥College of Ocean Science and Engineering, Shanghai Maritime University, Shanghai 201306, China
×Supporting Information

ABSTRACT: Polymer composites with excellent thermal conductivity and superior mechanical strength are in high demand in the electrical engineering systems. However, achieving superior thermal conductivity and mechanical properties simultaneously at high loading of fillers will still be a challenging issue. In this work, a facile method was proposed to prepare the epoxy composite with carbon fibers (CFs) and alumina (Al₂O₃). This CF and Al₂O₃ hybrid structure can effectively reduce the interfacial thermal resistance between the matrix and the CFs. The thermal conductivity of epoxy composite with 6.4 wt % CFs and 74 wt % Al₂O₃ hybrid filler reaches 3.84 W/(m K), which is increasing by 2096% compared with that of pure epoxy. Meanwhile, the epoxy composite still retains outstanding thermal stability and mechanical performance at high filler loading. A cost-effective avenue to prepare highly thermally conductive and superior mechanical properties of polymer-based composites may enable some prospective application in advanced thermal management.

INTRODUCTION

Many significant electronic applications require effective heat dissipation, especially for electronic devices such as integrated circuits and light-emitting devices (LEDs), as overheating always causes rapid aging or even failure of the core chip.1−4 Polymer-based thermal management material is widely accepted as a high-performance material in many fields for its easy processability, excellent chemical properties, and low cost, but its low thermal conductivity is still a major drawback and limits its applications.5,6 Many methods have been developed to improve its performance. Traditionally, it is reported that a variety of thermally conductive fillers including metal nanoparticles (e.g., Ag),7 metal oxides (e.g., Al₂O₃, MgO),8,9 nitrides (e.g., AlN, BN),10,11 and most recently graphene12 and carbon nanotubes (CNTs)13 can be usually included in the polymer matrix. Among them, alumina (Al₂O₃) has received considerable attention due to its remarkable physical properties, such as high thermal conductivity (30 W/(m K)), abrasion resistance, corrosion resistance, thermal stability, and electrical insulation, as well as its low cost.14 Thus, epoxy resin modified by Al₂O₃ filler-containing composites is considered as a promising method for improving thermal conductivity.15 However, researchers are focused on adding above 80 wt % loading of Al₂O₃ filler to obtain high thermally conductive composites by the traditional blend method,16,17 yet leading to significant degradation of mechanical strength and processability.18 Therefore, the researchers are urged to find facile and novel approaches to prepare composites with both high thermal conductivity and excellent mechanical properties.

In recent years, two kinds of methods have been adopted to obtain superior thermal conductivity as well as excellent mechanical strength. In the first method, the functionalization of filler surface by surfactants and coupling agents19−25 for reducing the interfacial thermal resistance between the thermally conductive filler and the polymer matrix.24,25 Nevertheless, the effect of thermal conductivity enhancement (TCE) reported above is limited compared without functionalization.26,27 The second is based on constructing a continuous filler architecture that can provide thermal transport channels. This network yield high thermal conductivity at a relatively low filler loading. For example, a compact packing structure of filler particles can be formed by combining different particle sizes of spherical Al₂O₃ at different mass ratios,28 which slightly increases the thermal conductivity. The porous Al₂O₃ ceramic skeletons are constructed by a new
processing technique including the gel casting, sintering, and vacuum infiltration methods. This structure can dramatically improve the heat transport of the composites. But it should be noted that the entire fabrication process is very complicated and not suitable for industrial mass production.

Herein, an epoxy resin-based composite with thermal transport expressways is prepared by incorporating carbon fiber (CF) foams and Al₂O₃ particles. The CF foams are used as the premade template to connect the neighboring spherical Al₂O₃ particles, which provides the coherent thermal transport channels for CFs. A hybrid network structure composed of

Figure 1. Preparation process of epoxy/CF/Al₂O₃ composites and the tailored works of the logotype of Ningbo Institute of Industrial Technology of Chinese Academy of Sciences (CNITECH) with samples.

Figure 2. SEM images of (a) Al₂O₃, (b) carbon fiber, (c) epoxy resin. The cross-sectional SEM images of epoxy/CF composites with different ratio with (d) 0 wt %, (e) 12 wt %, (f) 29 wt %, (g) 44 wt %, (h) 58 wt %, (i) 74 wt % Al₂O₃ and its corresponding, (j) EDS elemental mappings of C, O, and Al.
Al₂O₃ and CFs provides prolonged pathways similar to heat dissipation highways in epoxy composites and shows a much higher thermal conductivity (3.84 W/(m K)) at the loading of 74 wt % Al₂O₃ than that of pure epoxy (0.18 W/(m K)), which may find a potential application for thermal management of electronic devices. Furthermore, our composites present more outstanding thermal stability and excellent mechanical strength compared to pure epoxy.

## RESULTS AND DISCUSSION

The preparation process of the epoxy/CF/Al₂O₃ (ECA) composites is shown in Figure 1. First, the CF foams were mixed with Al₂O₃ aqueous solution and dried to obtain CF foam and Al₂O₃ particles (CA) hybrid fillers. The amount of Al₂O₃ in the CA mixture block could be controlled by adjusting the concentration of Al₂O₃ in the solution (as shown in Table S1). Then, the epoxy precursor was infiltrated into the CA mixture block and degassed under vacuum. Finally, the resulted mixture was precured and postcured to obtain the ECA composites with different Al₂O₃ ratios.

As can be seen in Figure 2, the morphologies and chemical compositions of Al₂O₃, carbon fiber, epoxy resin, and the cross-sectional of our epoxy/CF composites with different ratios of Al₂O₃ were characterized by using scanning electron microscopy (SEM), ZETA, and Raman spectroscopy. The average diameter of Al₂O₃ particles is about 5 μm as shown in the inset of Figure 2a. Raman spectrum that exhibits two Raman shifts at 1354 cm⁻¹ (D-band) and 1583 cm⁻¹ (G-band) in the inset of Figure 2b demonstrates that the main component of the foam is carbon fiber (spectral range is scanning from 1000 to 3200 cm⁻¹). The D-band intensity (I_D) and the G-band intensity (I_G) of carbon fiber correspond to the defect-inducing vibration mode and in-plane vibrations of graphite lattice, respectively. The SEM images shown in Figure 2c–i reveal that the Al₂O₃ particles and CFs have good contact with epoxy resin and this microstructure can increase the strength of ECA composites greatly. As the Al₂O₃ content increasing, more Al₂O₃ balls appeared on the fracture surface. Figure 2j shows the SEM image of the composite with 74 wt % Al₂O₃ along with the corresponding energy-dispersive spectrometry (EDS) elemental mappings of carbon (C), oxygen (O) and aluminum (Al). It further illustrates that the Al₂O₃ balls are gradually and closely distributed in the ECA composites.

The thermal conductivity of the polymer composites is often determined by factors such as the interaction between thermal filler and matrix, the loading of filler, the structure and properties of filler and matrix, and so on. The thermal conductivity of the ECA composites was evaluated by using a laser flash method. Figure 3a shows the difference in thermal diffusivity (TD) and thermal conductivity (TC) of epoxy/CF/Al₂O₃ composites. The TC of pure epoxy resin is only 0.18 W/(m K) at 25 °C. After adding CFs, the TC was significantly improved up to 1.48 W/(m K). In particular, when Al₂O₃ was added in epoxy/CF composites, the TC of the ECA composites was improved even higher and increased monotonically with the incorporation of the Al₂O₃ fillers. As the increase in the loading levels of Al₂O₃ from 12 to 74 wt %, the TC increased almost linearly from 1.97 to 3.84 W/(m K). Similarly, the curve of the TD of composites also shows the same change trend. Herein, the thermal conductivity enhancement (TCE) is defined as (λ - λ_m)/λ_m, where λ and λ_m are the thermal conductivity of composites with addition to fillers and the thermal conductivity of pure epoxy, respectively. Figure 3b compares the TC and TCE of the epoxy/CF composite, the ECA composite with 74 wt % Al₂O₃ filler loading (ECA-74)
and the pure epoxy at 25 °C. These values were more than 7 and 20 times of the pure resin, respectively. Furthermore, it is remarkable that the TC of composites prepared with two kinds of fillers, carbon fiber, and Al₂O₃, presented more improvement than that prepared with the individual filler, carbon fiber. The results may be attributed to three reasons. First, even a very small loading of carbon-based thermal fillers can dramatically increase the TC of composites because of their high intrinsic TC. Thermal transport through epoxy/CF composites is dominated by thermal resistance at the interface. The resistance results principally from interfacial defects and from phonon scattering caused by phonon spectra mismatch in two phases (fiber and matrix). Second, at lower Al₂O₃ filler loading levels (<20 wt %), a slight increase of the TC was observed. Only at high Al₂O₃ loading levels, a more significant improvement can be observed. It is reported that the high filler content with above 60−70 vol % can lead to continuous thermal conduction paths of fillers in the polymer composite. This implies that effectively thermal conductive pathways begin to form at high Al₂O₃ loading levels due to filler-to-filler connections, as observed in Figure 2e−i. Simultaneously, the CFs dispersed in epoxy, effectively connect with Al₂O₃ particles and form a dense thermal conductive network throughout the material. These connections reduce the chance of contact between CFs and the surrounding matrix, resulting in a decrease in their interfacial thermal resistance. As a result, an enhanced hybrid network composed of Al₂O₃ and CFs provides prolonged pathways similar to heat dissipation highways and shows a much higher thermal conductivity at the loading of 74 wt % Al₂O₃ than that of pure epoxy and epoxy/CF composites. The increase in TC with the temperature, indicate that the temperature improves the interfacial bonding between the fillers and the polymer matrix. Figure 3d shows the variation in TC of pure epoxy. The EC composite and ECA-74 composite upon multiple heating and cooling cycles alternating in the range of 25 and 100 °C. The trend of TC of pure epoxy, epoxy composites with temperature is consistent with Figure 3c. Both EC composite and ECA composite maintained their stable and original thermal conductivities during the first 10 cycles, demonstrating stable heat dissipation capacity after multiple treatments.

To intuitively display the heat dissipation performance of epoxy composites, an infrared camera was used to record the surface temperature distribution and variation, which can characterize their heat dissipation performances. The samples were investigated by placing on a ceramic heating plate to study their thermal behavior. We used the electrified heating plate as the heat source and fixed the samples on the plate with conductive tape. From the infrared images in Figure 4a, the surface temperature of each sample was measured and the surface color became brighter as the temperature increasing, which means the surface temperature of the sample increases. In particular, the surface heating rate of the ECA-74 composite was faster than that of pure epoxy and EC composite. Quantitatively, the surface temperature of pure epoxy, EC composite, and ECA-74 composite were measured as 90, 94, and 102 °C, respectively, when the temperature rose to 115 °C within 240 s. These results demonstrate that the ECA-74 composite exhibits a better heating dissipation performance. Similarly, according to the data measured by the infrared camera, the curves of surface temperature with time during the heating and cooling process were made, as shown in Figure 4b. It is obvious that the pure epoxy, EC composite, and ECA-74 composite reached their highest temperature at 93, 97, and 104 °C, respectively. Besides, ECA-74 presented much faster heating and cooling rates compared with other samples. In summary, the ECA-74 composite shows better heating and
cooling behaviors than the pure epoxy and the EC composite. As is evident in Figure 4c, an experiment was conducted to demonstrate the high thermal conductivity of the ECA-74 composite. Both the pure epoxy sample and ECA-74 sample in the same size (10 × 10 × 1.5 mm³) were put on the heater and the same size of CuSO₄ saturated solution was dropped in the sample center by the same pipette gun. As the temperature of heater increasing, the solution placed on the ECA-74 sample quickly changed from blue droplet to white powder, while that placed on the epoxy sample only became slightly smaller. It shows that the surface temperature of ECA-74 increases sharply compared with pure epoxy resin and ECA-74 exhibits high thermal conductivity along with the heat dissipation performance.

In addition to thermal conductivity, thermal stability is one of the most important properties of investigation for the epoxy composites. Figure 5a,b shows the thermogravimetric analysis (TGA) and derivative thermogravimetry (DTG) profiles of pure epoxy resin and its composites as a function of temperature under air atmosphere at a heating rate of 10 °C/min, respectively. For pure epoxy, there was a double-step process for thermally degrading in the air as shown in the black curve in Figure 5a. It is believed that the main loss with a maximum rate in the first stage is due to the decomposition of the epoxy network structure at 370 °C while leaving tiny amounts of char residue that is further degraded by thermal oxidation at around 420–540 °C in the second stage. The thermal decomposition curves of epoxy-based composites were one step more than that of pure epoxy. It is attributed to the oxidative decomposition reaction of carbon fibers at around 540–900 °C. Note that all the samples exhibited similar thermal behavior except for the additional step of carbon fiber decomposition reaction, indicating that the presence of fillers do not significantly change the degradation mechanism of the epoxy matrix. With the increase in Al₂O₃ filler content, the second step decomposition became less obvious, which is attributed to the reduction of epoxy resin content. The temperature for 10% weight loss (T_{10%}) and the maximum decomposition (T_{max}) are selected as the characteristic thermal parameters and the results are listed in Table S2. Quantitatively, the T_{10%} of pure epoxy resin was measured as 331.4 °C, while the T_{10%} of EC composites with 0, 12, 29, 44, 58, and 74 wt % Al₂O₃ filler were 318.7, 335.4, 331.4, 334.0, 341.7, and 358.9 °C, respectively. It is obvious that all the ECA composites exhibited a slight increase in T_{10%} compared to pure epoxy. Considering the third stage is the carbon fibers decomposition stage in the air, the carbon residue in the third step degradation reaction stage (540–1000 °C) is defined as the char yield. As can be seen that the char yields of all the ECA composites were improved when compared with that of pure epoxy. Furthermore, it is observed from Figure 5b that the maximum decomposition temperature (T_{max}) of the composites was also mildly increased after adding thermal filler, compared with that of epoxy resin in DTG curves. It is considered that the compact chars of the polymer matrix and hybrid thermal fillers will be formed during the process of thermal decomposition, which is beneficial for the enhancement of thermal stability for the composites. Meanwhile, the structure of CFs and Al₂O₃ increases the crosslinking between the binding sites of polymer matrix and fillers, thus limiting the mobility of the polymer segments at the interface of the epoxy and the thermal motion of the polymer chains. Moreover, the stress–strain curves of pure epoxy and its composites were measured to show their mechanical properties, as shown in Figure S1. The thermomechanical properties of ECA composites are shown in Figure 5c,d. The storage modulus, i.e., the amount of energy stored in the polymer, is associated with the stiffness along with the dampening capacity of the material. All the composite showed a superior storage modulus (E′) than pure epoxy in the glass region, as shown in Figure 5c. Particularly, the storage modulus of the ECA-44 composite exhibited a significant increase (8580 MPa at 40 °C) in the glassy region, which was 246.5% larger than that of pure epoxy (2476 MPa), suggesting that the thermal fillers are uniformly dispersed and have powerful interfacial adhesion with polymer matrix. The storage modulus fell with the
increasing temperature, which indicates that the energy dissipation occurs in the transition from glass-like to rubber-like behavior. It is seen that the storage modulus of the composite materials appeared to be much larger than that of the pure epoxy within the rubber state. The storage modulus of the composite, for instance, was about 215% higher than pure epoxy at 100 °C when Al2O3 filler content was 74 wt %. In addition, the loss factor tan δ is defined as the ratio of loss modulus to storage modulus, and it is very susceptible to structural transition for the solid material. The peak temperature of tan δ is selected as the glass transition temperature (Tg). It is observed from Figure Sd that the Tg values of the epoxy composites reaches to higher temperatures compared with pure epoxy. These results can be attributed to the existing hybrid thermal fillers in the composite material, which will increase the obstruction of the segmental motion for the epoxy chains owing to the effects of interfacial entanglement and interaction.

CONCLUSIONS

In summary, an effective heat transport highway structure was constructed by the CF-Al2O3 hybrid three-dimensional foams in epoxy matrix, which can provide coherent bridges composed of spherical Al2O3 particles and connect dispersed CFs in an epoxy resin matrix. The epoxy composite has high conductivity (3.84 W/(m K)) with 6.4 wt % CFs and 74 wt % Al2O3 hybrid filler, which is increasing by 2096% compared to that of pure epoxy, showing great potential for thermal management in electronic applications. In addition, the composite still retains outstanding thermal stability and mechanical performance at high filler loading. Our work reports a new and cost-effective method for preparing highly thermally conductive and robust heat-conducting polymer-based composites by designing effective thermal transport channels.

EXPERIMENTAL SECTION

Materials. Al2O3 powders with an average diameter of 5 µm were produced by Yaan Bestry Performance Materials Co., Ltd. (Sichuan, China). The Al2O3 particles were irregular in shape and close to a spherical shape. Carbon fiber foams were supplied by Vulcan New Material Technology Co., Ltd. (Hangzhou, China). A cycloaliphatic epoxy resin monomer (UVR 6105, 95%) was purchased from DOW Chemicals. Neodymium(III) acetylacetonate trihydrate (Nd(III) acac) (Shanghai D&B Biological Science and Technology Co., Ltd.) and methylhexahydrophthalic anhydride (MHHPA, Zhejiang Neodymium(III) acetylacetonate trihydrate (Nd(III) acac) Ltd., Malvern, U.K.) were adapted to estimate the micrometric sizes of Al2O3 particles. The heat capacity and the thermal diffusivity of the ECA composites were respectively measured through the differential scanning calorimetry (DSC 214) at 10 °C/min heating rate and the LFA 467 Hyper flash instrument (NETZSCH, Germany) at room temperature. The thermal conductivity (λ, W/(m K)) of composites was calculated by density (ρ, g/cm³), heat capacity (Cp, J/(g K)), and multiplying thermal diffusivity (α, mm²/s)

\[ \lambda = \rho \times C_p \times \alpha \]  

The density (ρ) of samples was determined by using the Archimedes water (alcohol) displacement method. An infrared camera (Fluke, Ti400) was used to collect IR images and record the thermal conductive behavior. The thermal stabilities of pure epoxy resin and the composites were evaluated through the thermogravimetric analyzer (TG 209 F3 instrument, NETZSCH, Germany). Thermogravimetric analysis (TGA) was conducted under air in the range of 50–1000 °C at a heating rate of 10 °C/min. A DMA Q800 dynamic mechanical analyzer (TA Instruments) was used to carry out dynamic mechanical analysis (DMA) to study the thermomechanical properties of composites. During the measurement, the samples were tested by carrying out in the single cantilever mode in a single-frequency pattern at 1.00 Hz from 25 to 270 °C with a 5 °C/min heating rate.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b03465.

AUTHOR INFORMATION

Corresponding Authors
*E-mail: xfwu@shmtu.edu.cn (X.W.).
*E-mail: jiangnan@nimte.ac.cn (N.J.).
*E-mail: yujinhong@nimte.ac.cn (N.Y.).

ORCID
Xiao Hou: 0000-0002-6465-6813
The authors are grateful for the financial support from the National Natural Science Foundation of China (51573201), the Project Funded by China Postdoctoral Science Foundation (2017M611757), NSFC-Zhejiang Joint Fund for the Integration of Industrialization and Informatization (U1709205), Public Welfare Project of Zhejiang Province (2016C31026), the Scientific Instrument Developing Project of the Chinese Academy of Sciences (YZ201640), the Strategic Priority Research Program of the Chinese Academy of Sciences (XDA22000000), and the Science and Technology Major Project of Ningbo (2016S1002 and 2016B10038). We also thank the Chinese Academy of Sciences for the Hundred Talents Program, the Chinese Central Government for the Thousand Young Talents Program, and 3315 Program of Ningbo.

ACKNOWLEDGMENTS

The authors declare no competing financial interest.

REFERENCES

(1) Tao, P.; Shang, W.; Song, C.; Shen, Q.; Zhang, F.; Luo, Z.; Yi, N.; Zhang, D.; Deng, T. Bioinspired engineering of thermal materials. Adv. Mater. 2015, 27, 428–463.
(2) Zeng, X.; Sun, J.; Yao, Y.; Sun, R.; Xu, J. B.; Wong, C. P. A Combination of Boron Nitride Nanotubes and Cellulose Nanofibers for the Preparation of a Nanocomposite with High Thermal Conductivity. ACS Nano 2017, 11, 5167–5178.
(3) Chen, J.; Huang, X.; Zhu, Y.; Jiang, P. Cellulose Nanofiber Supported 3D Interconnected BN Nanosheets for Epoxy Nano-composites with Ultrahigh Thermal Management Capability. Adv. Funct. Mater. 2017, 27, No. 1604754.
(4) Moore, A. L.; Shi, L. Emerging challenges and materials for thermal management of electronics. Mater. Today 2014, 17, 163–174.
(5) Singh, V.; Bougher, T. L.; Weathers, A.; Cai, Y.; Bi, K.; Pettes, M. T.; McMenamin, S. A.; Li, W.; Resler, D. P.; Gattuso, T. R.; Altman, D. H.; Sandhage, K. H.; Shi, L.; Henry, A.; Cola, B. A. High thermal conductivity of chain-oriented amorphous polyethylene. Nat. Nanotechnol. 2014, 9, 384–390.
(6) Liang, Q.; Yao, X.; Wang, W.; Liu, Y.; Wong, C. P. A three-dimensionally aligned functionalized multilayer graphene architecture: an approach for graphene-based thermal interfacial materials. ACS Nano 2011, 5, 2392–2401.
(7) Li, X.; Wong, C. P. Recent advances of conductive adhesives as a lead-free alternative in electronic packaging: Materials, processing, reliability and applications. Mater. Sci. Eng., R 2006, 51, 1–35.
(8) Yao, Y. M.; Zeng, X. L.; Guo, K.; Sun, R.; Xu, J. B. The effect of interfacial state on the thermal conductivity of functionalized Al2O3 filled glass fibers reinforced polymer composites. Composites, Part A 2015, 69, 49–55.
(9) Yu, J. H.; Huang, X. Y.; Wang, L. C.; Peng, P.; Wu, C.; Wu, X. F.; Jiang, P. K. Preparation of hyperbranched aromatic polyamide grafted nanoparticles for thermal properties reinforcement of epoxy composites. Polym. Chem. 2011, 2, 1380–1388.
(10) Hou, J.; Li, G. H.; Yang, N.; Qin, L. L.; Grami, M. E.; Zhang, Q. X.; Wang, N. Y.; Qu, X. W. Preparation and characterization of surface modified boron nitride epoxy composites with enhanced thermal conductivity. RSC Adv. 2014, 4, 44282–44290.
(11) He, Z. H.; Dai, W.; Yu, J. H.; Pan, L. L.; Xiao, X. E.; Lu, S. R.; Jiang, N. Enhanced thermal and mechanical properties of polyimide composites by mixing thermotropic liquid crystalline epoxy grafted aluminum nitride. J. Polym. Res. 2014, 21, 595.
(30) Ko, T. H. Raman spectrum of modified PAN-based carbon fibers during graphitization. *J. Appl. Polym. Sci.* 1996, 59, 577−580.

(31) Zickler, G. A.; Smarsly, B.; Gierlinger, N.; Peterlik, H.; Paris, O. A reconsideration of the relationship between the crystallite size Lα of carbons determined by X-ray diffraction and Raman spectroscopy. *Carbon* 2006, 44, 3239−3246.

(32) Meyer, N.; Marx, G.; Brezinka, K. W. Raman-Spectroscopy of Carbon-Fibers. *Fresenius J. Anal. Chem.* 1994, 349, 167−168.

(33) Lévéque, D.; Auvray, M. H. Study of carbon-fibre strain in model composites by Raman spectroscopy. *Compos. Sci. Technol.* 1996, 56, 749−754.

(34) Sun, X.; Ramesh, P.; Itkis, M. E.; Bekyarova, E.; Haddon, R. C. Dependence of the thermal conductivity of two-dimensional graphite nanoplatelet-based composites on the nanoparticle size distribution. *J. Phys.: Condens. Matter* 2010, 22, No. 334216.

(35) Hasselman, D. P. H.; Johnson, L. F. Effective Thermal Conductivity of Composites with Interfacial Thermal Barrier Resistance. *J. Compos. Mater.* 1987, 21, 508−515.

(36) Kumar, A.; Ayyagari, N.; Fisher, T. S. Effects of Graphene Nanopetal Outgrowths on Internal Thermal Interface Resistance in Composites. *ACS Appl. Mater. Interfaces* 2016, 8, 6678−6684.

(37) Wang, W. S.; Chen, H. S.; Ku, S. H.; Ko, S. H.; Jang, J. M.; Lee, T. W.; Lim, S. H.; Park, J. P.; Kim, J. D. Formation and analysis of SiC coating layer on carbon short fiber. *Surf. Interface Anal.* 2010, 42, 1231−1234.

(38) Zhang, C.; Tjiu, W. W.; Liu, T.; Lui, W. Y.; Phang, I. Y.; Zhang, W. D. Dramatically enhanced mechanical performance of nylon-6 magnetic composites with nanostructured hybrid one-dimensional carbon nanotube-two-dimensional graphene nanoplatelet heterostructures. *Adv. Funct. Mater.* 2010, 20, 3039−3044.

(39) Zhao, Y.; Barrera, E. V. Asymmetric Diamino Functionalization of Nanotubes Assisted by BOC Protection and Their Epoxy Nanocomposites. *Adv. Funct. Mater.* 2010, 20, 3039−3044.

(40) Lu, S. R.; Li, S. R.; Yu, J. H.; Yuan, Z. K.; Qi, B. Epoxy nanocomposites filled with thermotropic liquid crystalline epoxy grafted graphene oxide. *RSC Adv.* 2013, 3, 8915−8923.

(41) Serena Saw, W. P.; Mariatti, M. Properties of synthetic diamond and graphene nanoplatelet-filled epoxy thin film composites for electronic applications. *J. Mater. Sci.: Mater. Electron.* 2012, 23, 817−824.

(42) Zhao, Y.; Barrera, E. V. Asymmetric Diamino Functionalization of Nanotubes Assisted by BOC Protection and Their Epoxy Nanocomposites. *Adv. Funct. Mater.* 2010, 20, 3039−3044.

(43) Li, C.; Lin, C.-T.; Jiang, N. Epoxy composites filled with one-dimensional SiC nanowires−two-dimensional graphene nanoplatelets hybrid nanofillers. *RSC Adv.* 2014, 4, 59409−59417.