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Superior mechanical and thermal properties of poly(vinylidene fluoride) composites with dense carbon nanotubes

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Superior mechanical and thermal properties of poly(vinylidene fluoride) composites with dense carbon nanotubes

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Abstract. High thermal conductivity and mechanical properties are in great need for plenty of applications. In this study, a CNTs/PVDF composite was fabricated through hot press, which results in the dense CNTs in the polymer. The thermal conductivity of composite can reach to 0.6 W/(mK), increased by 328% compared with pure PVDF. Moreover, the Young's modulus and tensile strength of the composite are also largely increased by the strategy. This method can be a promising fabrication approach for the development of high-performance polymeric composites.

1. Introduction

With the rapid development of electronic devices, the electronic packaging becomes more and more important. The thermal interface material is one of the key parts to make sure the work of devices. However, at present, the balance of the mechanical and thermal properties of thermal interface material is still a challenge. Heat transfer is essential for the performance, reliability, and lifetime of electronic devices [1], which significantly depend on the thermal conductivity of the thermal interface material. Therefore, a plenty of works focus on the enhancement of thermal conductivity [2-5], especially the out-of-plane value. Polymer based composites filled by carbon materials are promising for fabricating the thermal interface material with superior performances, such as high strength and thermal conductivity. The effective method of previous investigation on polymeric composites is filling particles with high thermal conductive into polymer matrix. The common used fillers include alumina, boron nitride, graphene, and carbon nanotubes. However, an important issue that should be considered is the dispersion of nanofillers in the polymer matrix. That is because they tend to aggregate together, which could largely decrease the efficiency of fillers. On the other hand, because of the aggregate phenomenon, the composites may fracture easily under the external force.

Carbon nanotubes possess prominent thermal and mechanical properties at the same time, which make this materials very popular to prepare excellent functional composites. Carbon nanotubes have been regarded as one exciting nanomaterial, due to their notable physical properties, such as low density, large specific area, high strength, and superior thermal conductivity [6]. It is demonstrated to be potential as a thermal conductive material. However, polymeric composites containing high loading carbon nanotubes may exhibit many problems [7]. As we know, when the filler loading is not enough, the filler cannot construct the heat transfer pathway, which results the low effectivity of thermal transfer. Therefore, dense carbon nanotubes dispersion in polymer matrix is always desired. Hot press is accepted as an effective strategy to get the composite with uniform distribution and high filler



density. Because of that, we can strongly expect that hot press can also cause the dense carbon nanotubes in polymer composites.

Herein, we try to fabricate a carbon nanotubes/poly(vinylidene fluoride) composite with dense filler distribution by hot press. The thermal conductivity and mechanical properties are also measured, which shows remarkable enhancement through this strategy.

2. Experimental section

2.1. Materials

Poly(vinylidene fluoride) (PVDF) is supplied by Shanghai Dongfu Chemical Technology Co., Ltd. Multiwall carbon nanotubes (MWCNTs) are purchased from Nanjing XFNANO Materials Tech Co., Ltd. Ethyl acetate is provided by Xilong Scientific Co., Ltd., Shantou, China.

2.2. Fabrication of CNTs/PVDF composites

PVDF is mixed with ethyl acetate at the mass ratio of 1:10. Then CNTs are added into the mixture, followed by stirring and strong ultrasonication for about 1 hour. After that, the mixture is put into drying oven to remove the ethyl acetate. Subsequently, the obtained composite is put into hot press machine. The press is conducted under 0.2 MPa and 120 °C for 2 hours, then the dense composite can be achieved and be cut to the shape needed.

2.3. Characterization

Microstructure and morphology of the composites were obtained through scanning electron microscope (SEM, SU-8010, Hitachi). Tension measurement were performed on Instron 5980 with the loading rate of 10 mm/min. The thermal diffusivity coefficient was tested by a laser flash diffusivity instrument (DXF-500, TA Instruments). The density of materials was observed on an automatic density analyzer (PEAB, XS105DU, METTLER TOLEDO, Switzerland). The specific heat capacity was obtained on a differential scanning calorimetry instrument (DSC Q20, TA Instrument). Thermal conductivity (TC) was calculated from: $\lambda = \alpha \rho C_p$, where λ , α , ρ , and C_p refers to thermal conductivity, thermal diffusivity coefficient, materials density, and specific heat capacity of composites, respectively.

3. Results and discussion

3.1. Morphologic and structure characterization of the composite

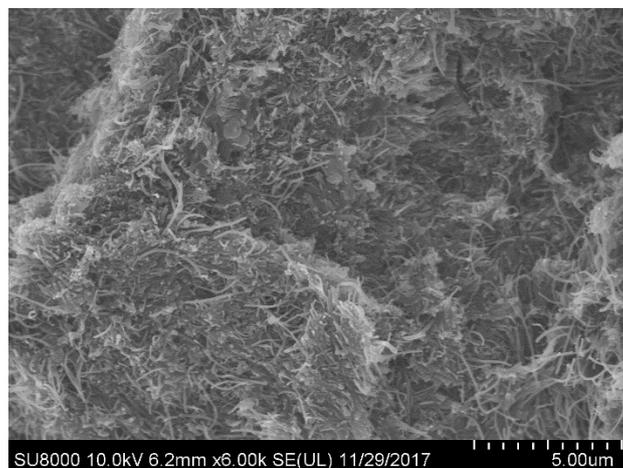


Figure 1. SEM image of CNTs/PVDF composite

Figure 1 shows the microstructure of CNTs/PVDF composite. It can be clearly seen that the numerous CNTs are well-dispersed in the PVDF matrix. The CNTs are dense and contact with each other, which is very useful for the construction of heat transfer pathway. Meanwhile, after hot press, the CNTs tend to disperse along the direction vertical to the compression direction. According to the knowledge, the thermal conductivity along the length-wise direction of CNTs is much higher than that of radial direction. Therefore, the orientation of CNTs in PVDF can significantly attribute to the heat transfer speed and the thermal conductivity enhancement of composite. In addition, this dense and oriented distribution of CNTs is also very helpful to the reinforcement of mechanical properties, such as Young's modulus and tensile strength.

3.2. Thermal conductivity of composite

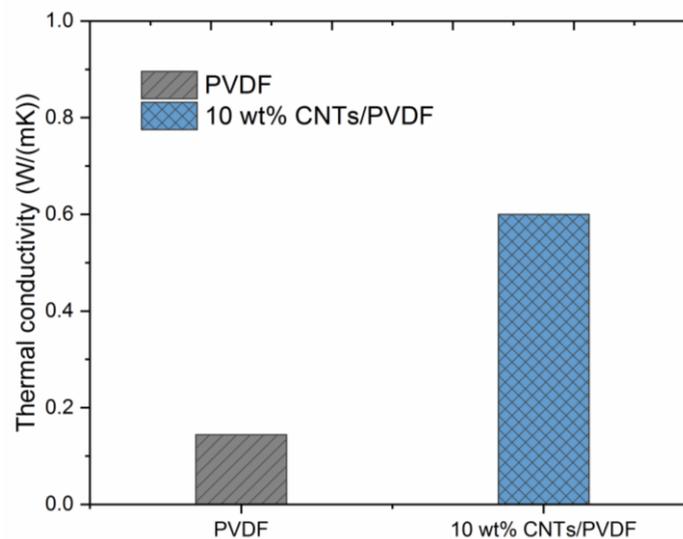


Figure 2. Thermal conductivity of pure PVDF and CNTs/PVDF composite.

Thermal conductivity values of pure PVDF and CNTs/PVDF composite are shown in Figure 2. As we can see, the thermal conductivity of CNTs/PVDF composite is much higher than that of pure PVDF. The value of composite is 0.6 W/(mK) which is enhanced by 328% compared with pure polymer. This result benefits from the hot press treatment and the dense CNTs network in the composite. The weight loading of the composite is 10 wt%, which is not very high. However, due to the hot press, the thermal conductivity can be increased effectively at such a low fraction. Thus, hot press is obviously useful for building the contacted heat transfer pathway. Compared with the earlier results in the literatures [8], the result in this work also has superiority in the enhancement efficiency of thermal conductivity at the same loading of 10 wt%.

3.3. Mechanical properties of composite

Figure 3 shows the tensile stress-strain curves of PVDF and CNTs/PVDF composite. It can be seen that CNTs/PVDF composite exhibits higher Young's modulus (1.01 MPa) and tensile strength (8.11 MPa) compared with pure PVDF which has the Young's modulus of 0.57 MPa and tensile strength of 4.42 MPa. Therefore, filling with CNTs and hot press are obviously useful for the reinforcement of PVDF composite.

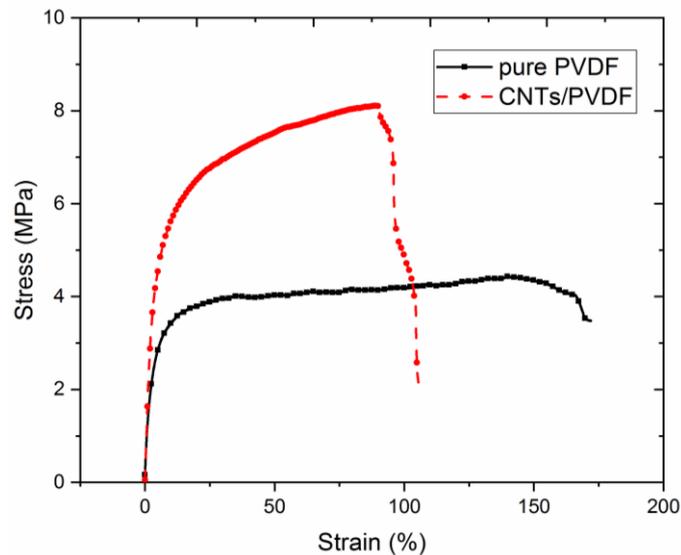


Figure 3. Tensile stress-strain curves of pure PVDF and CNTs/PVDF composite.

4. Conclusions

In this study, the hot press was utilized in the fabrication of CNTs/PVDF composite, which results in the dense CNTs dispersion. Benefiting from that, the thermal conductivity and tensile performance of the composite are largely enhanced. The thermal conductivity is tuned to 0.6 W/(mK), increased by 328% compared with pure PVDF.

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