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The Synergistic Effect of BN and MWCNT Hybrid Fillers for the Thermal Conductivity of Ultra-High-Molecular-Weight Polyethylene Composites

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***Abstract*—In this novel, (BN+MWCNT) /UHMWPE composites were prepared by powder blending method. The thermal conductivity of the polymer can be improved by adding boron nitride and multi walled carbon nanotubes .When the loading of BN was 50wt%, the thermal conductivity rate of ultra high mole-cular weight polyethylene reaches from 0.456W/ (M·K) to 0.9157W/ (M·K); when adding 1wt% MWCNT, the thermal conductivity coefficient of the composite further increased to 1.370W/ (M·K), more than 33% compared with single filler. It is showed that there was a significant synergistic effect between BN and MWCNT filler, which can form a good conductive pathway in the material. Scanning electron microscopy (SEM) analysis showed that the MWCNT effectively improved the interfacial bonding strength between BN fillers. Thermal gravimetric (TGA) analysis showed that the thermal stability of UHMWPE was significantly improved by adding MWCNT. Depth Digital Optical Microscopy (DDOM) confirmed that the composites formed segregated structure of "filler -polymer interface" and conductive pathways effective formation.**

Keywords-Ultra High Molecular Weight Polyethylene (UHMWPE); Boron Nitride(BN); Multi Walled Carbon Nanotubes; Thermal Conductivity

I. INTRODUCTION

With the development of science technology, national economy, integrated technology and assembly technology have been widely applied in social life, and the volume of electronic components and logic circuits has been reduced by thousands of times. And the operating frequency has increased sharply, causing its working environment to move to high temperature[1], which puts higher requirements on packaging materials, such as high thermal conductivity and thermal stability. General purpose high heat conduction materials and metal materials due to their corrosion resistance, electrical insulation and poor molding technology, can not meet the packaging requirements of electronic products, while thermal conductive polymer materials with its advantages of easy processing, low cost, good mechanical properties and electrical insulation have shown a good application prospect in the field of electronic packaging[2]. China will take high-performance polymer materials as one of the key strategic research directions of "made in China 2025", which will improve the innovation level of China's intelligent manufacturing key strategic materials industry technology.

However, the general polymer thermal conductivity is poor, improve the thermal conductivity of polymer is mainly mixed with high thermal conductivity filler. There are three kinds of filler particles for preparing thermal conductive polymers: metal, inorganic thermal conductive particles and carbon materials. In order to meet the requirements of heat dissipation and heat conduction for some occasions with higher requirements for electrical insulation, insulating heat conduction inorganic particles are mainly used for preparing electrical insulation heat conduction polymer composites[3]. In recent years, boron nitride (BN) has become a hot research topic, its structure is similar to the graphene, in addition to high electric breakdown strength, thermal conductivity, low moisture absorption rate, high temperature oxidation resistance, the dielectric constant and loss is the lowest, which is the most close to the polymer, is the preparation of low dielectric constant and loss, high thermal conductivity polymer ideal filler[4].

At present, BN composites have been widely used in research. Yang wenbin et al. [5] prepared BN/ PSF thermal insulation composites with thermal conductivity up to 2.08 W/mK by powder blending. Chen et al. [6] found that 3wt% t-zinc oxide whisker (t-ZnO)/ boron nitride (BN) filler could significantly improve PTFE. Yu et al. [7] found that a small amount of one-dimensional single-wall carbon nanotubes (SWCNTs) and two-dimensional graphene nanosheets (GNP) could significantly enhance the heat conductivity of epoxy resin matrix to the medium.

In conclusion, thermal conductivity of polymer matrix can be effectively improved by adding different types of mixed filler. In addition, the thermal conductivity of composites is largely dependent on the dispersion state of fillers in the polymer matrix. When the filler is distributed in the network state, the interface thermal resistance between the filler and the substrate can be effectively reduced, and the thermal conductivity of the composite material of the isolation structure can be improved. In this paper, an ultra-high molecular weight polyethylene nanocomposite with isolated structure (boron nitride + carbon nanotube) was prepared by powder mixing method, and compared with the thermal conductivity of pure UHMWPE and BN/UHMWPE composites, through the microscopic study of the formation mechanism of the heat conduction path in the heat flow direction of the nano-thermal composite material, the synergistic effect of the BN and MWCNT composite filler at different contents on the UHMWPE composite material is investigated. The interfacial thermal resistance between filler and matrix, filler and filler is reduced to improve the thermal conductivity.

II. EXPERIMENTAL RESEARCH METHOD

A. *Material*

Boron nitride (BN) is provided by Shanghai naiou nanotechnology co., LTD. Ultra-high molecular weight polyethylene (UHMWPE density 0.98 g/cm^3), carbon nanotubes (MWCNT) were provided by ZhongKe times nano co., LTD.

B. *Preparation of (BN+MWCNT)/UHMWPE*

The preparation of composite materials is mainly through mechanical blending and hot press forming processes (as shown in Fig.1). First, a certain amount of ultra-high molecular weight polyethylene, boron nitride and carbon nanotubes were weighed according to the formula ratio and added into the grinder, mixed and stirred evenly, and mixed for 10min at 32000 r/min to obtain the mixed powder of composite materials. Put the mixed powder into the grinding tool, and use the hot press to press the prepared powder into a sample (the temperature of the upper and lower plates of the hot press is set as 200 ℃, the pressure is 10MPa, and the pressure is maintained for 20 min). Then take out the cold press to room temperature (the pressure given by the cold press is 5 MPa and the pressure is maintained for 10 min). Finally, take out the sample (the sample is directly 15 mm, the thickness is 6 mm, and the shape is cylindrical, as shown in Fig.2).

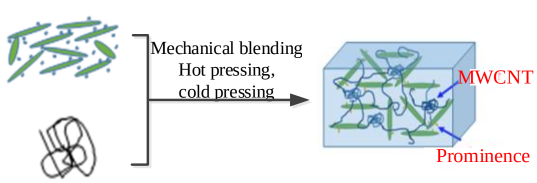

Figure 1. The process flow chart for preparation of composite materials (BN+MWCNT)/UHMWPE

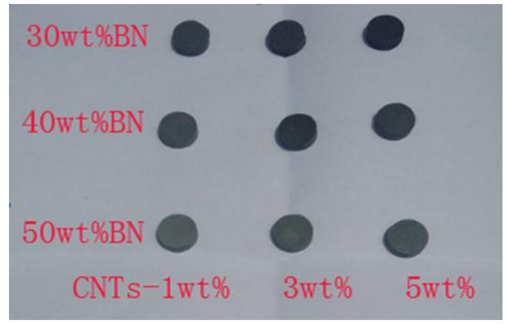

Figure 2. Composite material samples with different content of (BN+MWCNT)/UHMWPE

C. *Morphological characterization and performance testing*

1) *Scanning electron microscope(SEM)*

SEM analysis is to cut the sample into strips and soak them in liquid nitrogen for 5 min for quenching. After spraying gold on the section, JEOL jsm-5900lv scanning electron microscope was used for analysis and test.

2) *Digital optical microscope(DDOM)*

The two-dimensional digital depth profile of (BN+MWCNT)/UHMWPE film was observed by Japanese Keyence VH-Z100UR digital optical microscope.

3) *Thermal conductivity test*

The thermal conductivity of the composite material was measured by the thermal conductivity analyzer (Hotdisk thermal detector 1500), and the thermal conductivity of the material was measured by the transient plane heat source (TPS) method.

4) *Thermogravimetric analysis(TGA)*

III. RESULTS AND DISCUSSION

A. *Thermal conductivity of composite materials*

Fig.3 shows the thermal conductivity of UHMWPE composites filled with BN and MWCNT at different contents. A small amount of MWCNT (only MWCNT=1wt%) was added to significantly improve the thermal conductivity of the composites. With the increase of BN content, thermal conductivity increases continuously. When the content of BN reaches 50wt%, the thermal conductivity of BN/UHMWPE composite material reaches 0.9157W/mK. After a small amount of MWCNT filling, the thermal conductivity increased from 0.9157W/mK to 1.370W/mK, which exceeded the corresponding single packing system. This indicates that the synergistic effect between the composite fillers can effectively improve the thermal conductivity. BN is precisely because of its two-dimensional shape and larger ratio of width to thickness, which is easier to connect with one-dimensional MWCNT, form a thermal conductivity pathway, and improve the thermal conductivity of the composite material. At the same time, there are some protruding synapses on the surface of BN. With the help of the synapses, MWCNT is also dispersed more evenly on the BN nanocrystals and entangled with BN. In addition, MWCNT avoids agglomeration and forms a more compact heat conduction pathway under the synaptic stretching action.

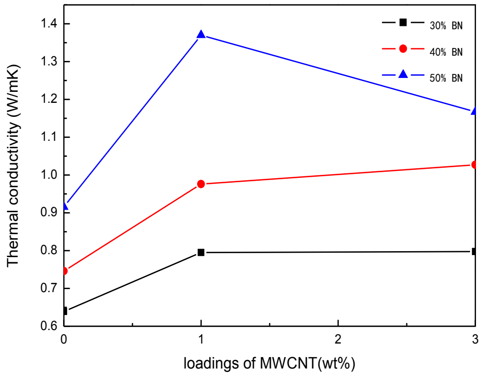

Figure 3. The coefficient of thermal conductivity of composite material (BN+MWCNT)/UHMWPE

B. *Microstructure and thermal conduction mechanism of composite materials*

Fig.4 is the SEM diagram of BN and MWCNT filled UHMWPE composite material, respectively (a)UHMWPE/BN 50wt% and (b)UHMWPE/(BN+MWCNT)(50wt%+1wt%). It is obvious from the electron microscope that BN and MWCNT are evenly dispersed on the surface of UHMWPE. At the same time, significant differences in interface between BN/UHMWPE and (BN + MWCNT) /UHMWPE composites can be observed. A small amount of MWNT will make the fracture surface of BN/UHMWPE composites change from flat to rough. Meanwhile, it also indicates that MWCNT effectively improves the interface adhesion between BN packing, which forms a causal relationship with the increase of thermal conductivity after the addition of MWCNT.

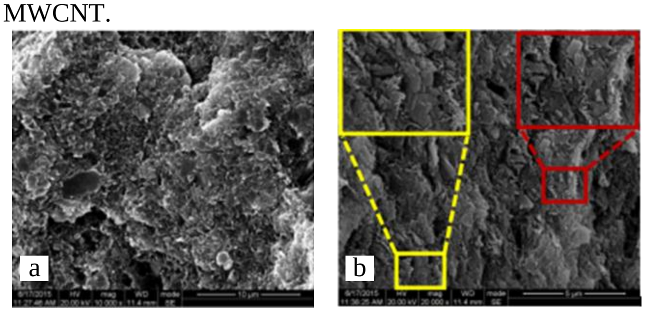

Figure 4. The SEM of composite material (BN+MWCNT)/UHMWPE

Fig.5 shows the depth digital optical microscopy of UHNWPE/(BN+MWCNT) composite material. It can be clearly seen from the figure that the isolation structure has an obvious filler-polymer interface, and more thermal conduction pathways are formed between the fillers, so that the heat flow is more unobstructed and the thermal conductivity is improved.

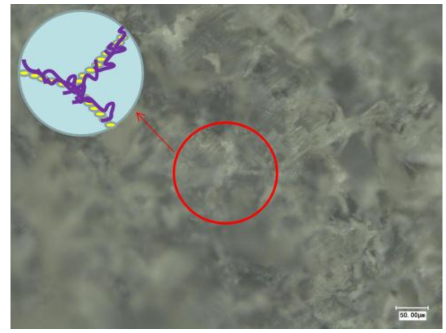

Figure 5. The depth digital optical microscopy of composite material (BN+MWCNT)/UHMWPE

Figure 6 shows the TGA curves of composites filled with 50wt%(BN+MWCNT) of different proportions. The initial decomposition temperature (Ti) and peak decomposition temperature (Tp) of 50wt% BN were 406.2℃ and 436.2℃, respectively, while the Ti and Tp values of filling 50wt%(1wt%BN+MWCNT) were significantly increased, which were 427.1℃ and 445.4℃, respectively. As the content of MWCNT continued to increase, the peak temperature showed a small decline trend (Ti and Tp of 5wt%BN+MWCNT were 409.5℃ and 435.6℃ respectively). The addition of MWCNT has two effects on the thermal stability of composites. On the one hand, the increase of thermal stability is due to the addition of filler, which limits the movement of the polymer chain segment and leads to the increase of decomposition temperature. At the same time, laminar filler can improve the gas barrier of the composite material. The gas generated in the thermal decomposition process of the composite material is difficult to volatilize out, so the thermal stability of the composite material is improved. On the other hand, MWCNT content continues to increase, resulting in a certain heat conduction pathway in the matrix. When the temperature rises, the heat generated in the matrix can dissipate quickly, which destroys the thermal stability of the material itself, thus leading to a decrease in the thermal degradation temperature. This is consistent with the improvement of thermal conductivity of composite materials. The dispersion state of MWCNT also affects the thermal stability of composites.

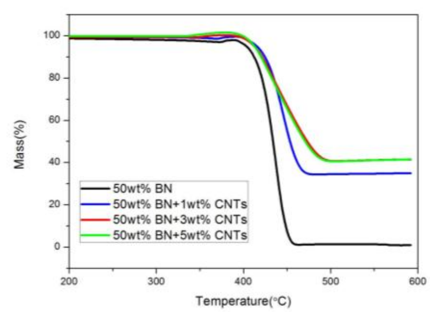

Figure 6. The TGA of composite material (BN+MWCNT)/UHMWPE

There are many factors that affect heat conduction of polymer, such as aggregation structure, orientation effect and temperature. In order to have good thermal conductivity, phonons must have a path to go, and a certain thermal conductivity network must be formed inside the composite material, so that the thermal conductivity in this direction will be higher than that in other directions. The thermal conduction mechanism of (BN+MWCNT)/UHMWPE composites is shown in Fig.7.

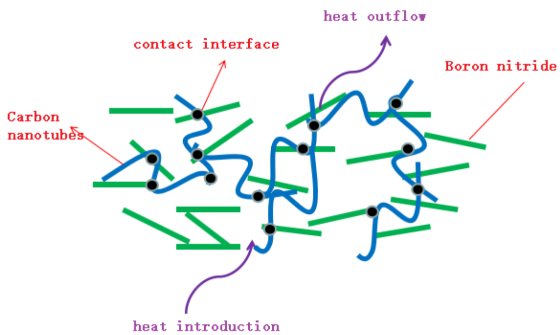

Figure 7. The transmission path of composite material (BN+MWCNT)/UHMWPE's heat

through the phonon propagation along the heat conduction path

The thermal conductivity of filled thermal conductive polymer depends on the synergistic effect between UHMWPE and thermal conductive filler. The heat conduction filler dispersed in UHMWPE has two shapes: sheet shape and tube shape. When the content and dosage of filler is low, the filler in the matrix mainly exists in an isolated form, without contact or influence between each other. At this time, the continuous phase is the polymer matrix, and the filler, as the dispersed phase, is covered by the polymer matrix, which is similar to the "sea-island two-phase system" in the polymer blend system. As the amount of filler increases, the particles begin to form contact with each other. At this time, the heat in the composite material spreads through the phonon along the path with the least thermal resistance, namely the thermal conduction path composed of thermal conduction particles. If the amount of filler is further increased, the partial heat conduction chain or heat conduction network will be connected and penetrated to form a heat conduction network through the whole polymer matrix material. At this point, the polymer and filler will form a continuous phase, so that the thermal conductivity of the composite material significantly improved.

When BN/MWCNT fillers are added into the matrix, when the content of BN is 50wt% and the content is high, a multi-chain thermal conductivity network can be formed inside the composite material, and the whole system will run through the thermal conductivity chain, which will improve the thermal conductivity of the filled composite material. Meanwhile, BN packing is easy to combine with multi-walled carbon nanotubes, and 2D dish-shaped BN has the characteristics of small volume and large aspect ratio, which can be easily collected into the blank of MWCNT. In the mixing process, with sharp protrusions, the carbon nanotubes will be evenly dispersed among BN sheets and form a stable connection between BN filler, thus improving the thermal conductivity.

IV. CONCLUSION

The composite material was prepared by powder blending and molding with ultra-high molecular weight polyethylene as the substrate and boron nitride and carbon nanotubes as the thermal conductivity filler. When the content of BN reaches 50wt%, the thermal conductivity of BN/UHMWPE composite material reaches 0.9157W/mK, and when a small amount of 1wt% MWCNT is added, the thermal conductivity increases from 0.9157W/mK to 1.370W/mK. It shows that the thermal conductivity of composites can be effectively improved through the synergistic effect between fillers of different dimensions, and the thermal conductivity of composites is more easily formed in the inner part of the material, which is also the main reason for the improvement of thermal conductivity of composites.

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REFERENCES

[1] Ma Chuanguo,Rong Minzhi.Research and application of thermal conductive polymer composites[J].Materials engineering, 2002 (7): 40～45.

[2] Chu Jiurong,Zhang Xiaohui,Xu Chuanrang.Research and application of thermally conductive polymer materials[J].Polymer materials science and Engineering,2000,16(4):17～20.

[3] Agari Y, Shimada M, Ueda A. Thermal diffusivity and conductivity of PPO/PS blends [J]. Polymer, 1997, 38(1): 2649-2655.

[4] Wang Jiajun.Preparation and research of polyimide / aluminum nitride composites[D].Zhejiang University,2001.

[5] Yang Wenbin,Zhang Kai,Yang Xuping.Preparation and properties of composite BN/ polysulfone thermal insulation composites[J].Journal of Southwest University of Science and Technology,2011, 26(1):24-27.

[6] Luan B C D, Huang J, Zhang J. Enhanced thermal conductivity and wear resistance of polytetrafluoroethylene composites through boron nitride and zinc oxide hybrid fillers[J]. Journal of Applied Polymer Science, 2015, 132(30).

[7] Yu A, Ramesh P, Sun X, et al. Enhanced Thermal Conductivity in a Hybrid Graphite Nanoplatelet – Carbon Nanotube Filler for Epoxy Composites †[J]. Advanced Materials, 2008, 20(24):4740–4744.