

Preparation and Performance of High Thermal Conductivity Polypropylene-based Composites

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Abstract: Polypropylene-based composites with high thermal conductivity were obtained by pressure injection molding using a hybrid form of PP as the matrix and FG as the thermally conductive filler with a particle size of 37 μm . Microscopic morphologies of the material were examined by SEM to determine the effect of FG content on the thermal conductivity and mechanical properties of the composites. The study found a clear correlation between the thermal conductivity of the composites and the FG content. The research confirmed a direct link between the thermal conductivity of the composites and FG content. At 70 wt%, the material demonstrated the greatest average, axial, and radial thermal conductivity of 7.52 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, 12.6 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, and 4.50 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively. However, any subsequent increase in fractional gradient (FG) content resulted in a decrease in the strength and modulus of the material. The highest tensile and flexural strength values of 34.9 and 63.7 MPa respectively, were achieved when the FG content was 60 wt%. At this particular FG content, the tensile and flexural modulus also reached 9.78 and 10.7 gigapascals (GPa), respectively. As the FG content increased, the strain on the composite material decreased. Note that the maximum tensile and flexural strains were measured at 50 wt% FG content, with values of 0.77% and 0.79%, respectively. The glass fiber sheets in the injection molded composites were uniform and predominantly vertically oriented.

Key word: Polypropylene-based, High thermal conductivity, Composite materials, Compression molding, Preparation and performance

1. Introduction

Polymer-based thermally conductive composite materials have received attention in the chemical and electronics industries over the past few years. Compared with metal materials, polymer-based thermal conductive composites have the advantages of corrosion resistance, lower cost, and easier processing. As a result, there is an urgent need for researchers to develop polymer-based thermally conductive composites that offer excellent performance [1-6]. In order to achieve thermal conductivity comparable to that of metallic materials, the construction of a complete thermal conduction path within the material is crucial for polymer-based thermal composites. Currently, commonly used techniques are the template method and the electromagnetic field-assisted induction method, among others. Nonetheless, these methods incur high costs and involve complicated preparation processes that do not meet the requirements for large-scale production. Therefore, the molding method with a simple operation, reduced material usage, and precise composition ratios is the best choice for composite material preparation. However, composites made using conventional molds often have high radial thermal conductivity and low axial thermal conductivity, which restricts their usage in chemical, pharmaceutical, construction, electronics, seawater desalination, and other industries [7-11]. The author used a simple molding technique to create a thermal conductivity pathway perpendicular to the surface of the composite sample. Using a mixture of polypropylene (PP) as a matrix and flake graphite (FG) as a thermal conductivity filler, a high thermal conductivity



composites were obtained utilizing the injection molding process. The research focused on exploring the impact of graphite content on the thermal conductivity and mechanical properties of the composite material. The orientation of graphite in the composites was analyzed with SEM to comprehend its impact on the properties of the composites.

2. Experimental methods

2.1. Materials

Polypropylene (PP230#, powder, Sinopec Maoming Petrochemical Company; PPK4038, granular, Formosa Chemical & Fibre Corporation); Flake graphite (FG, 99%, particle size 37 μ m, Hebei Haoteng Trading Co., Ltd); Titanate coupling agent (NDZ-201) were bought from Dinghai Plastic Chemical Co., Ltd; Absolute ethanol (analytical pure, Tianjin Yongda Chemical Reagent Co., Ltd); Antioxidant 1010, PML168 and Zinc stearate were obtained from Shanghai Aladdin Biochemical Technology Co., Ltd.

2.2. Equipments

Double-roll open mill (ZG-180, Dongguan Zhenggong Electromechanical Equipment Technology Co., Ltd); Electric drying oven (101-3AB, Tianjin Taisite Instrument Co., Ltd); Flat vulcanizer (ZG-200T, Dongguan Zhenggong Electromechanical Equipment Technology Co., Ltd); Universal prototype (WZY-240, Chengde Hengtong Test Instrument Co., Ltd); Thermal constant analysing (TPS-2500S was produced by Sweden Hot disk Co., Ltd); Electronic universal testing machine (104C, Shenzhen Wance Testing Machine Co., Ltd.); Scanning Electron Microscope (SEM, Inspect-S50, Thermo Fisher Scientific).

2.3. Preparation of polypropylene-based high thermal conductivity composites

Mixing: NDZ-201 was dissolved in anhydrous ethanol and then mixed well with FG. The mixture was dried in an oven at 140°C. A certain amount of PP, zinc stearate, antioxidant and treated FG were pre-mixed in a high-speed mill, mixing not less than 3 times, each time 1 min, placed in the opening machine mixing uniform, to obtain mixed masterbatch standby.

Pressure Injection Molding: The mixture was weighed and poured into a preheated mold, then the mold was pressed, the molten masterbatch was uniformly injected into the cavity by pressure injection, and the pressure was maintained for a certain period of time and allowed to cool to room temperature, and the molded composites obtained. The press injection conditions are as follows: upper template at 220 °C, lower template at 198 °C, and pressure at 12 MPa.

2.4. Testing methods

The thermal conductivities were tested according to GB / T 32064-2015. The radius of probe was 3.189mm, the test temperature was 25°C, and the sample size was 40×40×3mm。

The tensile properties were tested according to GB / T 1040.1-2006. Sample size and shape: 170mm×10mm×3±0.2mm, 1B.

The bending properties were tested according to GB / T 9341-2008. Sample size: 80mm×10mm×3±0.2mm. Testing speed: 2mm/min.

The microscopic morphology of the samples was observed by SEM.

3. Results and discussion

3.1. Thermal conductivity

To investigate the effect of FG content on the thermal conductivities of the composites, the FG particle size was $37\mu\text{m}$, the coupling agent was NDZ-201, and the dosage was 0.7wt% of the FG content. The composite materials with different FG content by pressing molding, and the thermal conductivity test results were shown in Fig. 1.

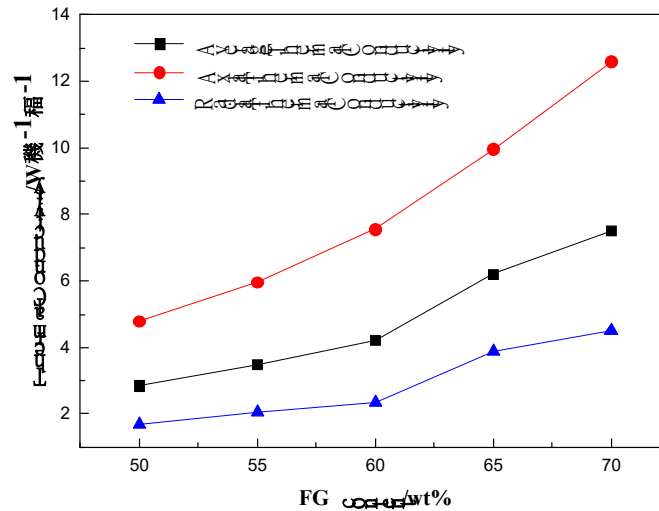


Fig.1 Effect of FG content on thermal conductivities of composites

The thermal conductivities of the composites increased with the increase of FG content. When the FG content was 50 wt%, the average, axial and radial thermal conductivities of the materials were $2.86 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, $4.80 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, $1.70 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively. When the FG content was increased to 70 wt%, the average, axial and radial thermal conductivities of the materials were $7.52 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, $12.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and $4.50 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, which were increased by 1.61, 1.58, and 1.65 times, respectively. The higher the FG content in composites, the more FG will overlap, and the greater the chance that the FG in the laminate will overlap. As the molten material entered the cavity and folds during compression molding, the flat FG was oriented vertically, resulting in the highest axial thermal conductivity.

3.2. Mechanical property

To investigate the effect of FG content on the mechanical properties of composite materials, the FG particle size was $37\mu\text{m}$, the coupling agent was NDZ-201, and the dosage was 0.7wt% of FG content. The composite materials with different FG content were prepared by pressing molding. The performance test results were shown in Fig. 2, Fig. 3 and Fig. 4.

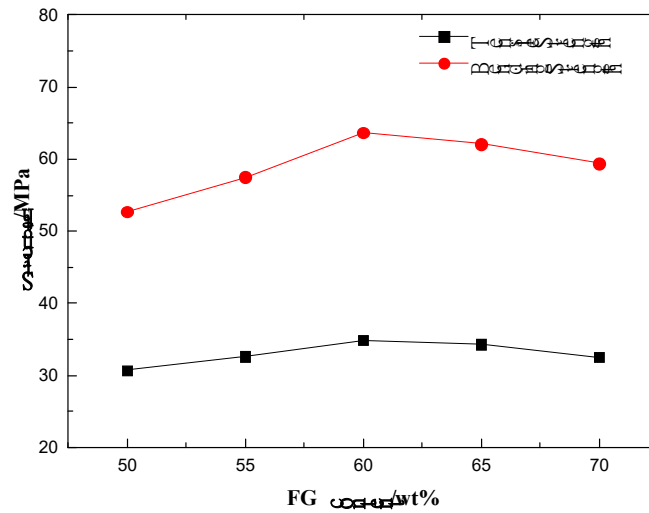


Fig.2 Effect of FG content on mechanical properties of composites

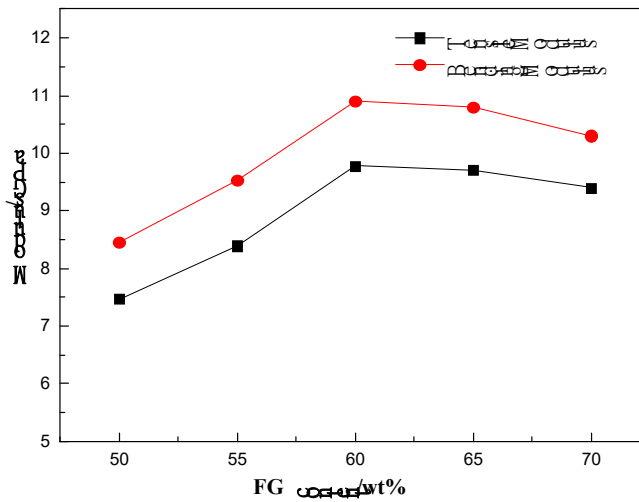


Fig. 3 Effect of FG content on modulus of composites

According to Fig. 2 and Fig.3, the strength and modulus of the composites first increased and then decreased with the increase of FG content. When the FG content was 50wt%, the tensile strength and bending strength of the composite were 30.7MPa and 52.7MPa, and the tensile modulus and bending modulus were 7.47GPa and 8.46 GPa, respectively. The strength and modulus of the composite increased with the increase of FG content. When the FG content was 60wt%, the strength and modulus of the composite reached the maximum, the tensile strength and bending strength reached 34.9MPa and 63.7MPa, the tensile modulus and bending modulus reached 9.78GPa and 10.7 GPa, respectively. When the weight percentage of FG was increased to 70%, both tensile and bending strengths exhibited a decrease, to 32.5 MPa and 59.4 MPa, respectively. Similarly, the tensile and bending modulus decreased to 9.60 GPa and 10.5 GPa, respectively. When the FG content was as high as 50 wt%, it acted as a rigid particle and provides reinforcement to the composites [12]. However, if FG content was too high, it became difficult to uniformly wrap FG with PP, leading to material defects. As a result, the composite was more likely to fracture when subjected to external forces, causing a decrease in material strength and modulus.

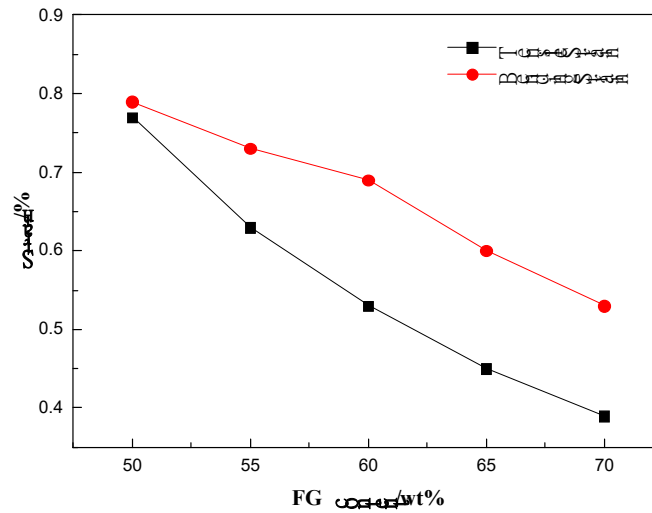
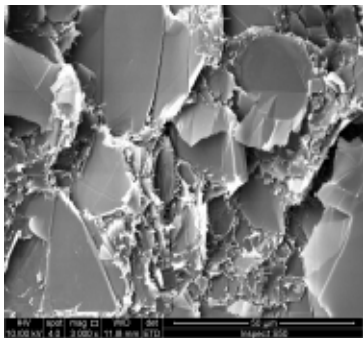


Fig. 4 Effect of FG content on strain of composites

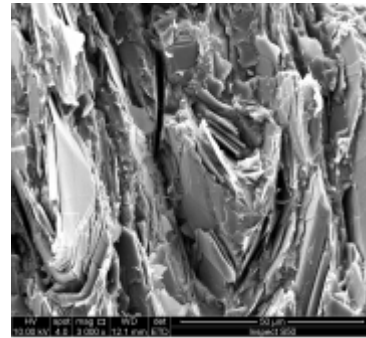
As can be seen in Fig. 4, the strain of the composite material decreased with the increase of FG content, when the FG content was 50 wt%, the tensile and bending strains were the largest, which were 0.77% and 0.79%, respectively; when the FG content was increased to 70 wt%, they decreased to 0.39% and 0.53%, respectively. These can be attributed to the increasing rigidity of the composites with higher FG content and the consequent reduction of toughness, thereby resulting in decreased strain of the material.

3.3. SEM analysis

To visualize the distribution and orientation of FG in compression injection molded composites, SEM observations of the cross-sections for composites with 70 wt% FG were taken and presented in Fig. 5.



(a) Section perpendicular to feed direction



(b) Section parallel to feed direction

Fig. 5. SEM observations of the cross-sections for composites with 70 wt% FG were taken and presented

From Fig. 5, it can be seen that FG were uniformly dispersed throughout the material, resulting in excellent mechanical properties and high thermal conductivity of the composite materials. Figure 5 (a) and (b) showed that the composites in the direction normal to the surface, the orientation of the FG in the direction of vertical orientation were governed by vertical orientation, consistent with the direction of heat flow, the composites in the vertical direction of the thermal conduction path were increasingly complete.

4. Conclusion

The study analyzed the properties of compression-molded PP-based composites with high thermal conductivity filled with FG. Increasing the FG content resulted in improved thermal conductivity and enhanced mechanical properties of the composites. The average, axial, and radial conductivities of the composites peaked at 70 wt% of FG. The maximum strength and modulus of the composites were achieved at 60 wt% of FG. Increasing the FG content to 50 wt% led to the maximum strain of the composites. SEM analysis exhibited predominantly vertical orientation and uniform distribution of FG in the compression-molded PP-based composites with high thermal conductivity.

Acknowledgments

This work was financially supported by the Fundamental Research Funds Pilot Project of the Hebei Academy of Sciences (Project No: 2023PF03-1) and the Two-institute Cooperation Project of Hebei Academy of Sciences (Project No: 22713) .

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