Micro-scale Aluminium Filled Polymer Composites with Improved Mechanical and Thermal Properties via Hot-pressing

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Abstract: Polymer based thermal interface materials are widely utilized in aeronautical and aerospace equipment, and can significantly contribute to the cooling of devices, which depends on their high thermal conductivities. In this work, a poly(vinylidene fluoride) (PVDF) based composite with micro-scale aluminium particles is fabricated. The aluminium particles were successfully embedded into the PVDF matrix, and the composite possesses dense structure caused by the hot-pressing process. Benefiting from that, the value of thermal conductivity improves to 1.82 W/(mK), which is 767% higher than that on pristine PVDF. Meanwhile, compared with pristine polymer, the mechanical performances, including tensile strength and modulus, are also significantly improved by 99% and 517%, respectively. The phenomenon can be attributed to the dense inner structure and effective heat transfer pathways of aluminium particles. This simple and effective strategy provides an available method to fabricate the thermoplastic polymeric composite with superior thermal and mechanical properties, used for the high-performance aeronautical and aerospace equipment.

Keywords: Polymer composite, aluminium, Hot-pressing, Mechanical properties, Thermal analysis

1. Introduction

Because of their well performance, such as light mass, flexibility, and easy prepare features, polymeric composites are usually applied in electronic devices, weapon systems, aeronautical and aerospace industry and many other fields[1–3]. Nevertheless, the polymeric materials always have low strength and thermal conductivities, which has limited their applications as the structure requiring superior mechanical and thermal properties, for example thermal interface materials (TIMs)[4, 5]. TIMs are widely applied in the communication, electronic and cooling devices of aeronautical and aerospace equipment, and play a significant role in this field. More exactly, TIMs can significantly contribute to the reliability, lifetime, and high speed of aeronautical and aerospace devices, which largely depend on the thermal dissipation. Polymer-based composites can play a significant role in thermal dissipation [6–8], and are the typical materials as TIMs. It is widely known that the thermal dissipation capability of TIMs mainly depends on their thermal conductivity. Generally, the thermal conductivity of the composite is enhanced by filling fillers with high thermal conductive performance, thereby forming an
effective heat transfer pathway in the matrix. To achieve the high thermal conductivity, some kinds of inorganic fillers are embedded into polymer matrices, including metals [9], carbon nanotubes (CNTs) [10], alumina [4], graphene[11], hexagonal boron nitride [12], etc. However, for the nanoscale fillers, because of the strong van der Waals forces, the fillers present agglomeration easily in the polymer matrix.

Aluminium, as a typical metal material, occupies a high thermal conductivity of ~ 237 W/(mK), and has attracted many attentions for serving as thermal conductive material in thermal management field. It has been demonstrated that aluminium provides notable thermal dissipation effectively. Due to its high thermal conductivity, it can be obviously inferred that aluminium can obviously improve the thermal conductivity of polymer composites. Micro-scale aluminium particles, which are easily obtained, has been widely utilized in manufacturing industry [4]. Beside the excellent thermal conductive property, micro-scale aluminium is much more easily to be dispersed in polymer matrix than the fillers with nanoscale size. However, the separated filler particles are hard to construct the heat transfer pathways when they have a large distance. Moreover, because of the large distance between aluminium particles and polymer matrix, caused by the interface incompatibility and weak adhesion, the composite usually has a high interface thermal resistance[4]. Thus, the weight loading of filler should be effectively enhanced, and the fabricated composites should be denser to induce the interface thermal resistance.

Poly(vinylidene fluoride) (PVDF), a kind of fluoropolymer, has been extensively used for the thermoplastic matrix of polymer composites. Owing to its thermoplastic property which, it is very easy to mould this material into a film via the considerably simple methods, for example hot-pressing. Resulting from that, PVDF has been an available choice for fabricating TIMs [10]. Nonetheless, the thermal conductivity of PVDF is extremely low (~0.21 W/(mK)) that it is not suitable for acting as TIM directly. Therefore, an effective strategy should be developed to embed the thermal conductive materials into PVDF appropriately, which can improve the thermal conductivity of composite significantly.

Herein, an aluminium/PVDF composite was carefully fabricated by embedding micro-scale aluminium particles into PVDF matrix via hot-pressing approach. Meanwhile, the thermal properties and mechanical performances were investigated carefully, and both of thermal and mechanical performance of the composites were obviously enhanced, which can be attributed to the dense and effective pathway of heat and stress.

2. Materials and experiments

A. Materials

Aluminium particles with the average micro-scale size of ~48μm were supplied by Aladdin Co., Shanghai, China. Ethyl acetate (≥99.5%) was obtained from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. Poly(vinylidene fluoride) (PVDF) are provided by Shanghai Dongfu Chemical Technology Co., Ltd.

Figure 1. SEM image of aluminium particles.
B. Fabrication of aluminium/PVDF composite

PVDF polymer powder was first dissolved into ethyl acetate with a moderate speed stirring of 200 r/min. The micro-scale aluminium particles were added into the polymer solution with speed stirring for about 10 min. Subsequently, the mixture was under vacuum at 80 °C for 4h to dislodge the ethyl acetate. After that, the obtained composite was pressed under 120 °C with stress of 0.2MPa for 3 hours on hot press machine, and then obtained the final composite.

C. Characterization

The morphology of materials are obtained using scanning electron microscope (SEM, SU-8010, Hitachi). Laser flash diffusivity instrument (DXF-500, TA Instruments) was used to measure thermal diffusivity coefficient. The density was obtained from an automatic density analyzer (PEAB, XS105DU, METTLER TOLEDO, Switzerland). The specific heat capacity is measured via differential scanning calorimetry instrument (DSC Q20, TA Instrument). Tensile tests are conducted on Instron 5980 at tensile speed of 10 mm/min, and the sample possessed the dogbone profile with the tensile length of 12.5mm.

3. Results and analysis

A. Morphologic and Structure of the composite

![SEM images](image)

**Figure 2.** SEM images of (a) pure PVDF; (b) aluminium/PVDF composite; (c) magnification of aluminium/PVDF composite, respectively.

Figure 2 shows the morphologic and structural characterization of the materials used and prepared in this work. Aluminium particles have the micro-scale size, and their size values are not fully uniform, which is beneficial to form denser structural between the big and small particles (Figure 1). As shown
in Figure 2a and b, there are clearly different morphologies between pristine PVDF and aluminium/PVDF composite. It is obvious that the aluminium particles are homogeneously dispersed in PVDF matrix without obvious agglomeration. Figure 2c displays the magnification of aluminium/PVDF composite. We can see that the aluminium particles can be driven to be very close to each other, through this hot-pressing process, which can result in effective heat transfer pathways. Due to the higher flowability of PVDF comparing with aluminium particles at high temperature, the aluminium gradually flows towards each other, and they become harder to move than PVDF matrix, which results in the dense aluminium.

B. Thermal conductivity of the composite

TC of pure PVDF and aluminium/PVDF composite were tested, and the values are presented in Figure 3. It can be clearly found that TC of aluminium/PVDF is much higher than that of pure PVDF. Compared with low thermal conductivity value (0.21 W/(mK)) of PVDF matrix, TC of aluminium/PVDF composite can increase to 1.82 W/(mK), which is remarkably improved by 767%.

This result can be attributed to the introduction of micro-scale aluminium particles in PVDF and the hot-pressing procedure, which forms the relatively effective path for heat transfer in PVDF matrix. Thus, thermal can conductive along the path from one side to the other side of the composite more effectively, which leads to the improved thermal conductivity.

![Figure 3. TC of pure PVDF and aluminium/PVDF composite.](image)

C. Mechanical tests

Figure 4 presents the tensile curves of pure PVDF and the composite through tensile measurement at the speed of 10 mm/min. Obviously, the aluminium/PVDF composite exhibits extremely high modulus and tensile strength, which are helpful for the stability of TIMs. More exactly, the Young’s and strength are 3.7 MPa and 7.98 MPa, respectively, showing 517% and 99% enhancement, respectively, compared with pure PVDF. More than this, the elongation at break of the aluminium/PVDF is lower than that of pure PVDF, due to the high weight loading of aluminium particles.
4. Conclusion
In summary, the aluminium/PVDF is prepared using micro-scale aluminium particles by hot-pressing approach. TC of the aluminium/PVDF can be enhanced up to 1.82 W/(mK), and is improved by 767%. The elastic modulus and strength are improved by 99% and 517%, respectively. These can be attributed to the dense structure and effective heat transfer pathways of aluminium particles resulting from high weight loading and the hot-pressing process. This strategy develops an effective approach to fabricate excellent TIMs for aeronautical and aerospace devices.

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