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Effect of graphene nanoplatelets on coefficient of thermal expansion of polyetherimide composite

Huang Wu^{a,b,*}, Lawrence T. Drzal^{a,b}

^a Composite Materials and Structures Center, Michigan State University, East Lansing, MI 48864, USA
^b Department of Chemical Engineering and Material Science, Michigan State University, East Lansing, MI 48864, USA

HIGHLIGHTS

- Coefficient of thermal expansion (CTE) of polymer composite is characterized.
- Reduction of linear thermal expansion depends on filler orientation.
- Filler orientation is characterized based on the location of the specimen.
- Filler orientation is changed by annealing, causing subsequent change in CTE.
- CTE and linear thermal expansion coefficient are modeled.

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ABSTRACT

Thermal expansion is one of the major concerns for polymer composites. In this research, graphene nanoplatelets (GNPs) were added to polyetherimide (PEld) thermoplastic polymer in order to reduce the coefficient of thermal expansion (CTE) of the injection molded composite. First, the coefficient of linear thermal expansion (LTE) was measured in three directions in the anisotropic coupon: 0° , 90° and the out of plane *Z* direction. It is found that the GNP particles are very effective in terms of reducing the LTE in 0° direction due to high degree of alignment. After annealing above glass transition temperature, significant increase of 0° LTE and decrease of Z° LTE were observed. The bulk CTE was calculated by adding up the LTEs in all three directions and is found to be independent of annealing. Second, several models were applied to predict both CTE and LTE. It is found that Schapery's lower limit model fits the experimental CTE very well. Chow's model was applied for LTEs in three directions. The behavior of GNP-5/PEld composites is explained by the combination of Chow's model and morphology obtained by scanning electron microscope (SEM).

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1. Introduction

Thermoplastic materials are receiving more and more attention as the matrix to host either continuous fibers or nano-scale particles. Compared to thermoset polymers, thermoplastics are generally tougher and more resistant to chemical decomposition [1]. Moreover, thermoplastics can be processed much faster than thermosets which require a dedicated curing cycle. The thermoplastics also have a much longer shelf life. However, a key factor that has been bothering the thermoset matrices is still not solved by switching to thermoplastics: the expansion and contraction of the polymer due to change in temperature. If used as a structural material, the dimensional change of the polymer due to cyclical (season or location) changes may cause serious deformation. In a composite consisting of continuous fibers, the difference in the coefficient of thermal expansion (CTE) between fiber and matrix would result in significant residual stress in the composite [2–5] which will generate defects [6,7] and affect matrix-dominated properties such as moisture absorption [8] and glass transition temperature [9].

Many efforts have been made to reduce the CTE of a thermoplastic system. Nano fillers such as carbon nanotubes (CNTs) [10– 13], nano clay [14–17], nano graphite [18] and mesoporous silica [19–22] are mostly used in such applications. Although the difference in CTE still exists for nano fillers as compared with their





^{*} Corresponding author. Department of Chemical Engineering and Material Science, Michigan State University, East Lansing, MI 48864, USA. Tel.: +1 517 899 9640. *E-mail address:* huang.wu.84@gmail.com (H. Wu).



Fig. 1. Schematics of TMA sample preparation.

matrices [23], the extremely small size of the filler minimizes the damage especially when the filler is homogeneously dispersed. Warrier et al. [10] incorporated CNTs into carbon fiber (CF)/epoxy composite and succeeded in increasing the glass transition temperature and reducing the CTE, indicating the effectiveness of using nano fillers to minimize the residual stress in a fiber reinforced composite.

Nano clay is the most popular filler for reducing CTE of the composites. Gu and Chang [16] reported a 65% reduction in CTE by adding 3 wt% organomontmorillonite in polyimide by a special processing route. This reduction is huge compared to other results [15,17] where a reduction around 10% is normally found for the same loading. However, the major contributor in Gu's work is the thermal imidization of the matrix instead of the clay. For CNT, the influence is smaller. A 2% reduction is found in 3 wt% CNT/epoxy system [11].

Suzuki et al. fabricated mesoporous silica and studied its effect on reducing the CTE of epoxy [21,22] and silicone rubber [20] systems. The large pore volume of the mesoporous silica material is very suitable for enhancing thermoset type material since the monomers of epoxy or silicone rubber can be effectively drawn into the porous structure by capillary force. However this type of filler may not work as well for a thermoplastic system due to its high viscosity.

A newly-developed nano filler, graphite nanoplatelet (GNP) has already shown great potential to reduce the CTE in the composites. With the same 2D geometry, the GNP can provide a reduction in the CTE of its composite comparable to nano clay, while adding other functionalities such as electronic conductivity to the composite. Kalaitzidou et al. [18] reported a 25% reduction for 3 vol% GNP filled polypropylene. Moreover, the GNP also possesses high thermal and electrical conductivity and can be produced by a scalable, costeffective process [24], making it the ideal filler for multifunctional composites.

PEId is an engineering thermoplastic that has unique flameresistance properties and would only generate a very small amount of low-density gas when forced to burn, making it well suited and widely used in airplane, aerospace, submarine and highfloor building applications. However, these applications may also involve dramatic ambient temperature change, which would benefit from a lower CTE. The need of using continuous CF in PEId matrix also would benefit from a reduction in the transverse CTE as a result of adding GNP to the GNP/PEId composite. In this research, the linear coefficient of thermal expansion (LTE) is investigated in injection molded GNP/PEId composites in three dimensions: 0° (flow direction), 90° and the out-of-plane Z direction. The CTE of the thermally annealed samples are also measured. In addition, models for both LTE and CTE are applied according to the morphology of the GNP/PEId composites.

2. Experimentation

2.1. Materials

Polyetherimide (PEI) was obtained from Sabic-IP (Ultem 1010). GNP-5 (3.9 micron average diameter, 40 m² g⁻¹ surface area) was provided by XG-Science Inc. (Lansing, MI).

2.2. Melt-extrusion

GNP and PEId were blended by melt-extrusion in a Leistritz twin-screw extruder (MIC27/6L-48D). The screws were operated in the co-rotation setup. Barrel temperatures were set to 310 °C, with a melt temperature at 340 °C. The die pressure was at around 5.5 MPa. Screw speed was set at 150 rpm.

2.3. Injection molding

The composite material was injection molded by a Milacron VSX 85-4.44 (Cincinnati, OH) injection molder with a Master Precision tensile coupon (Type I dog bone) die (Series No. 24157, Greenville, MI). The melt pressure during injection molding was 90 MPa. The resulting material was checked for morphology of the GNP filler after processing [25]. It was found that the size of the GNP particles reduced from 3.9 to about 2 μ m.



Fig. 2. LTE of injection molded GNP/PEId composites.

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