

# MICROSTRUCTURE AND THERMAL CONDUCTIVITY OF POLYMER COMPOSITES WITH NANO AND MICRO FILLERS

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## Introduction

Thermally conductive polymer composites offer new possibilities for replacing metal parts in electric systems. The advantages of polymer composites as compared to metals include improved corrosion resistance, lighter weight, and the ability to adapt the conductivity properties to suit the application needs. Applications of conductive composites as heat sinks in electric systems require new composites with a thermal conductivity from approximately 1 to 30W/mK[1].

An approach of current interest to improve the thermal conductivity of polymers is the selective addition of nano fillers with high thermal conductivity. Carbon nanotubes (CNTs) have exceptionally high mechanical properties and thermal conductivity [2]. One promising application of CNTs is as fillers in polymer composites to provide enhanced thermal properties.

The effects of type, shape and size of fillers on thermal performance of composites has been addressed by many researchers. Recently, Sanada et al.[3] fabricated the composites with the random close-packed structure of spherical micro particles and nano fillers, and investigated the potential of nano fillers to enhance thermal conductivity of the composites. The objective of this paper is to study the effective thermal conductivity of the composites with nano and micro fillers focusing on the shape and size of micro fillers. Packing simulation and finite element analysis were performed to predict the composite thermal conductivity. In addition, experimental measurements of the thermal conductivity of the manufactured polymer composites were carried out by using a steady-state method.

## Experimental characterization

Polymer composites were prepared by mixing Epikote 828 epoxy resin with Epikure YH300 curing agent, Epikure BMI12 accelerator and fillers. All the above chemicals were manufactured by Japan Epoxy Resins Co., Ltd. Nano filler was multi-walled carbon

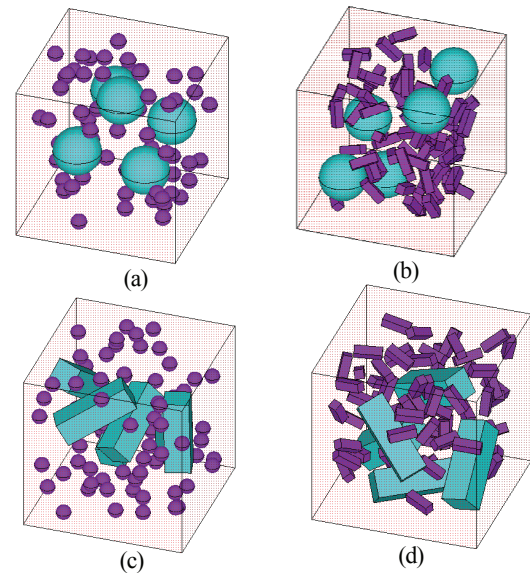


Fig.1 Random unit cell models: (a) two-particle; (b) large-particle/ small-fiber; (c) large-fiber/small-particle; (d) two-fiber

nanotube (MWNT) from Microphase Co., Ltd. The MWNT volume fraction in the matrix  $V_f^N$  is 3%. Three types of alumina particle were DAW05 (average diameter of  $4.5 \mu\text{m}$ ), DAW10 (average diameter of  $9.0 \mu\text{m}$ ) and DAW45 (average diameter of  $45.2 \mu\text{m}$ ) from Denki Kagaku Kogyo Co., Ltd. The alumina particle had a perfect spherical shape. The carbon fibers were from Kureha Corporation and three types (M207S, M201S and M2007S) were selected. The average diameter of M207S, M201S and M2007S is  $14.5 \mu\text{m}$ . The average lengths of M207S, M201S and M2007S are 400, 150 and  $90 \mu\text{m}$ , respectively. All the above fillers were used as received. The epoxy mixture was then degassed, poured into a mold and cured for 2h at  $100^\circ\text{C}$ , followed by 4h at  $150^\circ\text{C}$ . The samples were composite disks of diameter 50mm and thickness 15mm. Thermal conductivity of the composites  $\lambda_c^{\text{exp}}$  were measured using a HC-110 thermal conductivity tester (Eiko Instruments Co., Ltd.).

## Numerical approach

A Monte-Carlo algorithm in the MacroPac program from Intelligensys was used to generate a unit cell with randomly distributed micro fillers. Fig.1 shows random

unit cell models of two-particle, large-particle/small-fiber, large-fiber/small-particle and two-fiber. For two-filler system, the micro filler volume fraction  $V_f^M$  is given by

$$V_f^M = \frac{V_1 N_1 + V_2 N_2}{L^3} \quad (1)$$

where  $V_1$  is the volume of a large micro filler,  $V_2$  is the volume of a small micro filler,  $N_1$  is the number of large micro fillers,  $N_2$  is the number of small micro fillers and  $L$  is the unit cell dimension. The filler size ratio  $\alpha$  is defined as  $\alpha = \sqrt[3]{V_1/V_2}$ .

The steady state thermal problem for the random unit cell model was solved using ANSYS and the effective thermal conductivity  $\lambda_c^{FEM}$  of the composites was evaluated. The thermal conductivities of the particle (alumina), the fiber (carbon), and the matrix (epoxy resin) are 36[4], 100 and 0.208W/mK[3], respectively.

## Results and discussion

The effect of addition of MWNTs to the matrix on the thermal conductivity of the composites was studied. The predicted thermal conductivity of two-filler models with  $\alpha=3$  for various  $V_f^M$  are plotted in Fig.2 as a function of thermal conductivity of the matrix containing MWNT  $\lambda_m^*$ . In Fig. 2,  $\beta_1$  is the aspect ratio of large fibers and  $\beta_2$  is the aspect ratio of small fibers. Improving the thermal conductivity of the matrix is through the addition of MWNTs. Two-fiber caused the largest increase in composite thermal conductivity, followed by large-fiber/small-particle and large-particle/small-fiber, and then two-particle.

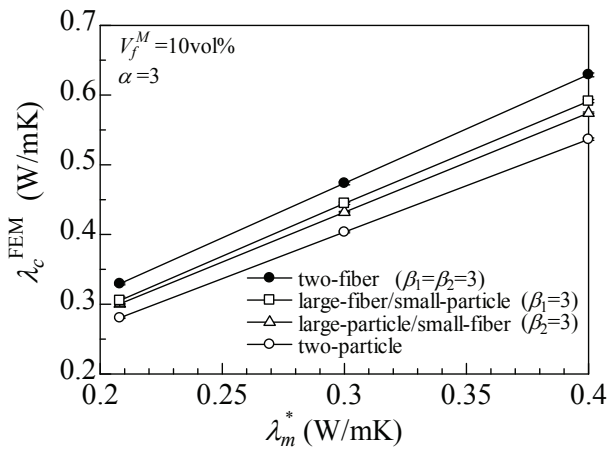


Fig.2 Variation of predicted thermal conductivity of the composites with thermal conductivity of matrix containing MWNT

Table 1 shows the measured thermal conductivity of M201S/M2007S (two-fiber system), M207S/DAW45

(large-fiber/small-particle system) and DAW10/DAW05 (two-particle system) composites with/without MWNTs. The results for DAW10/DAW05 composites are from reference 3. M201S/M2007S composites (two-fiber system) likely had a lower thermal conductivity, compared to the other composites, due to poor wetting between carbon fiber and epoxy (imperfect interfacial thermal contact). Moreover, the results demonstrate that the improvement in the thermal conductivity of the composite is attained through the addition of MWNTs.

Table 1 Measured thermal conductivity of the composites with/without MWNTs

System	$V_f^M$ (vol%)	$\lambda_c^{exp}$ (W/mK)	
		without MWNT	with MWNT ( $V_f^N=3\text{vol}\%$ )
M201S/M2007S (two-fiber, $\alpha=1.2$ , $\beta_1=10$ , $\beta_2=6$ )	30	0.53	—
	50	0.94	—
M207S/DAW45 (large-fiber/small-particle, $\alpha=1.1$ , $\beta_1=28$ )	30	0.80	—
	50	1.12	1.50
DAW10/DAW05 (two-particle, $\alpha=2$ )	30	0.60	—
	50	1.15	1.74

## Conclusions

The thermal conductivity of the polymer composites with nano and micro fillers were investigated numerically and experimentally. The results showed that the addition of MWNTs to the matrix significantly increased the thermal conductivity of the composites.

## References

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