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Analytical study on thermal conductivity enhancement of hybrid-filler polymer composites under high thermal contact resistance

Ich Long Ngo [a,b,*], Chan Byon [c,*], Byeong Jun Lee [a,*]

[a] School of Mechanical Engineering, Yeungnam University, Gyeongsan 38541, Republic of Korea
[b] School of Transportation Engineering, Hanoi University of Science and Technology, No. 01, DaiCoViet, Hanoi, Viet Nam
[c] School of Mechanical, Aerospace and Nuclear Engineering, Ulsan National Institute of Science and Technology, Ulsan 44919, Republic of Korea

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ABSTRACT

This paper presents an investigation on the thermal conductivity (TC) enhancement for polymer composites reinforced with randomly distributed hybrid filler using both numerical and theoretical approaches. Effects of thermal contact resistances (TCR) at the interfaces between fillers and the matrix material are considered comprehensively. Consequently, the deviation of ETC under the particle distribution effect generally increases as the filler volume fraction (VF) increases. Modified Hashin-Shtrikman model proposed in previous study can be used to predict accurately the TC of composites only if two fillers have the same TCR and TCR is less than prescribed value. The ETC depends significantly on the TCR ratio between two fillers, obviously at high sum of TCRs. Particularly, the TC can be enhanced significantly and effectively even at very high TCR by increasing VF ratio while keeping other appropriate conditions. Furthermore, the optimal TC ratio between two fillers at which the ETC is maximized, was shown to be independent on the TCR only if two fillers have the same TCR. These results provide a very good guideline for synthesizing hybrid-filler polymer composites and enhancing its TC under the effects of TCR.

1. Introduction

As the demands in synthesizing advanced materials, enhancing thermal conductivity (TC, hereafter) of a polymer composite is becoming increasingly important. This is due to its potential applications, for example flexible polymers in electronic packaging and encapsulations, satellite devices. One of the effective methods for enhancing the TC is the use of fillers. Three major groups of fillers have been known: Carbon-based fillers, metallic fillers, and ceramic fillers. In each filler group, single kind of filler (mono-filler) and the combination of two or more than two fillers with different properties have been used in literature.

Mono-fillers were first considered for enhancing the TC of composites since this comes from a simple idea of mixing an additional materials to the existing one. Many studies demonstrated that the TC of polymer composites depends significantly on properties and features of such fillers. Mu et al. [1] reported that the TCs of silicone rubber filled with ZnO increased with the volume content of filler. Polymer composites based on vapor-grown carbon fiber were estimated to obtain a very high TC of 1260 $W\,m^{-1}\,K^{-1}$ [2]. Tibbets et al. [3] indicated that tenfold improvement in TC was demonstrated in epoxy composites with vapor-grown carbon nanofiber. Furthermore, Tekce et al. [4] demonstrated the TC of copper-filled polyamide composites depends on the TC of the fillers, their shape and size, volume fraction (VF), and spatial arrangement in the polymer matrix. Additionally, Yu et al. [5] reported that the TC of composites filled with AlN reinforcement was higher for larger particle size. The TC obtained at about 20% VF of AlN was five times higher than that of pure polystyrene.

Recently, core-shell nanoparticles has also received an interest in the TC enhancement for pure polymers. Nanoparticles (cores) are first synthesized, then they are covered by one-layer or multi-layers materials (shells) with enhanced properties (higher TC), finally these core-shell nanoparticles are added into polymer matrix. As a result, the TC of polymer composites can be improved due to the higher TC of shells compared to using core-nanoparticle with lower TC only. Indeed, Zhou et al. [6] indicated that the TC was remarkably improved by adding core-shell Ag/SiO_2 nanoparticles into polyimide matrix. Kim et al. [7] reported the significant enhancement of the TC using FeCr metal core-aluminum oxide shell particles with a highly mesoporous shell layer compared to

* Corresponding authors at: School of Mechanical Engineering, Yeungnam University, Gyeongsan 38541, Republic of Korea (I.L. Ngo, B. J. Lee); School of Mechanical, Aerospace and Nuclear Engineering, Ulsan National Institute of Science and Technology, Ulsan 44919, Republic of Korea (C. Byon).

E-mail addresses: ngoichlong@ynu.ac.kr (I.L. Ngo), cbyon@unist.ac.kr (C. Byon), bjlee@ynu.ac.kr (B.J. Lee).

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Nomenclature

D	diameter of particle [m]
k	thermal conductivity [W m^{-1} K^{-1}]
L	length of unit cell [m]
N	number of particle [–]
q''	heat flux [W m^{-2}]
Q	overall heat flux [W]
R_c	thermal contact resistance [m^2 K W^{-1}]
t	thickness of the interface [m^{-1}]
T	temperature [K]
V	volume [m^3]
x, y, z	coordinate in x, y and z direction [m]
Greek symbols	
κ	thermal conductivity ratio [–]
ϕ	volume fraction [–]
λ	coefficient in Eq. (10) [–]
Subscripts, superscripts	
1, 2	index of particle 1 and 2 [–]
cell	unit cell [–]
d	division of two quantities [–]
eff	effective [–]
in	inlet [–]
int	interface [–]
m	matrix [–]
max	maximum [–]
opt	optimum [–]
out	outlet [–]
p	particle [–]
s	sum of two quantities [–]
*	non-dimensional form

the use of uni-modal particles. Recently, Ngo et al. [8,9] have performed an extensive study and a numerical analysis on the TC of core-shell nanoparticle polymer composites. Optimal conditions for enhancing and achieving the maximum TC were also proposed.

Heterogeneous fillers or hybrid fillers have been regarded as another effective method for enhancing TC of composites, as reported in literature. Two kinds of filler with the difference in the TC, size, or shape are filled together into polymer matrix. It was demonstrated that there is a positive synergic effect on the TC of composite when heterogeneous fillers are used [10]. The combination of fillers can effectively cause thermally conductive pathway/chain/network, thus much heats are transferred between fillers and surrounding, hence the TC can be improved significantly. Lee et al. [11] found that the TC of composites filled with spherical particles and fibrous fillers are higher than that of matrix material at low and intermediate filler content. Xu et al. [12] also indicated that using a hybrid filler at a volume ratio of 1:6 gives the TC higher than using each single filler. The highest TC of polyvinylidene fluoride (PVDF) composites was up to 11.5 W m^{-1} K^{-1} at total filler VF of 0.6. Chen et al. [13] reported the combination of fillers at a certain condition can result in higher ETC than using single filler for the same total filler loading. The optimal ratio of hybrid filler was shown to be valid for epoxy composites filled with single-walled carbon nanotubes and graphite nanoplatelets. The same conclusion was also provided by Mu et al. [1] for silicone rubber filled with hybrid ZnO particles.

Additionally, the TC of polymer composites can be improved by adding nanofillers into the polymer-matrix containing the micro fillers, as reported in Sanada's study [14]. The epoxy composites filled with aluminum nitride/graphene nano-hybrid filler were reported to be able to get the highest TC enhancement compared to the pure epoxy [15]. Recently, the influences of many effects on the composite TC including anisotropic distribution, particle size of hybrid fillers has been also studied [16]. Although previous studies provided the results relevant to the behaviors and characteristics in the TC of composites, no research considered comprehensively the TC enhancement of hybrid filler polymer composite under the effects of thermal contact resistance (TCR, hereafter), particularly at high values in combination with the aforementioned effects.

This paper focuses on the effects of TCR on the effective thermal conductivity (ETC, hereafter) of polymer composites with randomly distributed hybrid fillers while other effects such as particle distribution and particle size are also considered. The TC was predicted by COMSOL Multiphysics linked with user-defined code in MATLAB. The guidelines for optimizing or maximizing the TC are provided, and valuable conclusions are explored in the present study. In addition, the thermal behaviors of ETC due to the synergic effects of hybrid fillers are also considered and discussed under the effects of TCR.

2. Numerical methodology

Numerical model and boundary conditions (BC, hereafter) are shown in Fig. 1a. A unit cell can be used as a control volume since the number of particles dispersed throughout the matrix (continuous phase) is very large. In Fig. 1a, hybrid filler particles, particle-1 (green, larger) and particle-2 (red, smaller) are assumed to be isolated each other and randomly distributed inside the unit cell. Geometrical model was first built by user-defined code in MATLAB. First particles were then distributed randomly in such a manner that all of them do not contact each other. This ensures the assumption mentioned above. Second particles were distributed subsequently. At this point, second particles required do not contact with either itself or first ones distributed previously. Geometry was tested to make sure the requirements such as number of particles and VFs. This procedure is repeated until those requirements are satisfied, then the geometry was finally imported into COMSOL using Livelink for MATLAB. In this regard, the particle distribution is changed according to the different hybrid fillers considered. This is to include the effects of anisotropic particle distribution resembling to that in experimental process. The TCs are assumed to be constant for all spherical heterogeneous-particles (k_{p1}, k_{p2}) and matrix materials (k_m).

The thermal flow field is considered to obey the Fourier's law with no heat source. Therefore, Laplace equations can be used to describe the behaviors of heat transfer in a composite structure, which are given by

$$\frac{\partial}{\partial x}\left(\frac{\partial T_i}{\partial x}\right)+\frac{\partial}{\partial y}\left(\frac{\partial T_i}{\partial y}\right)+\frac{\partial}{\partial z}\left(\frac{\partial T_i}{\partial z}\right)=0 \quad i=1,2,m \tag{1}$$

The subscripts "1", "2" and "m" denote the particle-1, particle-2 and matrix, respectively. The following non-dimensional variables are used to convert Eq. (1) into a non-dimensional form.

$$x^*=\frac{x}{L};\quad y^*=\frac{y}{L};\quad z^*=\frac{z}{L};\quad k_i^*=\frac{k_i}{k_m};\quad T_i^*=\frac{T_i-T_{out}}{q''_{in}L/k_m} \tag{2}$$

where the characteristic length L, is the dimension of a unit cell, as shown in Fig. 1a. It is highlighted that the heat flux was specified at