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International Journal of Thermal Sciences 46 (2007) 842–847

International Journal of Thermal Sciences

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Effect of interface on the thermal conductivity of carbon nanotube composites ☆

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Received 17 May 2006; received in revised form 7 November 2006; accepted 7 November 2006

Available online 4 December 2006

Abstract

This paper presents the effect of interface on the equivalent thermal conductivity of the carbon nanotube composites. The element free Galerkin method has been utilized as a numerical tool to evaluate the thermal conductivity of the composites. The numerical results have been obtained using continuum mechanics approach for a model composite problem, and it was found that the interface has a major effect on the thermal conductivity of the composites. The effect of interface on the effective conductivity of the composite is small for short nanotubes as compared to long nanotubes. Interface thickness also plays an important role on the effective thermal conductivity of the composite. Nanotube anisotropy has got a small effect on effective thermal conductivity of the composites. Transverse thermal conductivity of the composite has got nearly linear variation with nanotube length.

Keywords: Carbon nanotube; Nano-composites; Longitudinal and transverse thermal conductivity; Anisotropy; Interface; Meshless; Element free Galerkin method

1. Introduction

For more than ten years, carbon nanotubes (CNTs) have been extensively studied by many researchers due to their excellent properties and numerous applications [1–5]. Carbon nanotube composite is one of their many applications. In recent years, CNT composites have attracted ever increasing attention of many scientists and researchers [6–9]. CNTs are much stronger, have high conductivity and larger aspect ratio as compared to conventional carbon fibers. Many believe that the reinforcement of CNTs in polymer matrix may provide us an entirely new class of materials. Therefore, thermal behavior prediction of nano-composite materials is very essential to know the amount of heat dissipation from their surfaces. Few numerical simulations have been carried out to predict the thermal properties of nano-composites using continuum mechanics approach [10–14]. Nishimura and Liu [10] applied the boundary integral equation method for the thermal analysis of CNT based nano-composites. They solved a heat conduction problem in 2-D infinite domain embedded with many rigid inclusions with the help of a fast multipole boundary element method. Zhang et al. [11,12] used the meshless hybrid boundary node method for the heat conduction analysis of carbon nanotube composites. They coupled their method with the fast multipole method to solve large scale problems. Song and Youn [13] evaluated the effective thermal conductivity of the carbon nanotube/polymer composites by control volume finite element method. Singh et al. [14] applied the element free Galerkin method to evaluate the equivalent thermal conductivity of CNT-composites, and found that the thermal conductivity of the composite is the function of nanotube dimensions.

It has been observed that the interface plays an important role in heat conduction through carbon nanotube composites [15–18]. Nan et al. [15] developed an empirical formula to predict the effective thermal conductivity of carbon nanotube composites with interface resistance, and found that the interface resistance greatly affects the thermal conductivity of the composites. Shenogin et al. [16] measured the effect of thermal boundary resistance on the heat flow in carbon nanotube composites. They found that the effective thermal conductivity of nanotube polymer composites is limited by interface resistance. A theoretical model was proposed by Xue [17] to evaluate the

☆ This work was supported by the CLUSTER of Ministry of Education, Culture, Sports, Science and Technology, Japan.

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doi:10.1016/j.ijthermalsci.2006.11.003

Nomenclature

k_{cx}, k_{cy}, k_{cz}	thermal conductivities of nanotube in x-, y- and z-directions respectively .. $\mathrm{W\,m^{-1}\,K^{-1}}$
k_e	equivalent thermal conductivity of composite in longitudinal direction .. $\mathrm{W\,m^{-1}\,K^{-1}}$
k_{eT}	equivalent thermal conductivity of composite in transverse direction .... $\mathrm{W\,m^{-1}\,K^{-1}}$
k_i	thermal conductivity of interface material equivalent to interface resistance R_i .... $\mathrm{W\,m^{-1}\,K^{-1}}$
k_{mx}, k_{my}, k_{mz}	thermal conductivities of matrix in x-, y- and z-directions respectively .. $\mathrm{W\,m^{-1}\,K^{-1}}$
L	length of square RVE....................... µm
L_c	nanotube length........................... µm
m'	number of terms in basis
n	number of nodes in the domain of influence
$p_j(\mathbf{x})$	monomial basis function
q_n	normal heat flux........................ $\mathrm{W\,m^{-2}}$
r_c	nanotube outer radius .... nm
R_i	interface resistance .... $\mathrm{m^2\,K\,W^{-1}}$
t_c	nanotube thickness......................... nm
t_i	interface thickness .... nm
$T^h(\mathbf{r})$	MLS approximation function for temperature
w	weight function used in MLS approximation
$\bar{w}$	weighting function used in weighted integral form
V_m	matrix domain
V_i	interface domain
V_c	CNT domain
$\Phi_I(\mathbf{r})$	shape function
α	penalty parameter

Subscript

$m,\ i,\ c$	denote the matrix, interface and nanotube respectively

thermal conductivity of the composite with interface resistance, and it was found that the interface plays an important role in the conduction through carbon nanotube composites. Ju and Li [18] recently measured the effect of interface thermal conductance on the overall conductivity of the composites. They found that the interface conductance has a major effect on the overall thermal properties of the composites. On the other hand, Bagchi and Nomura [19] noticed that the interface conductance has a small effect on the overall conductivity of the composites.

So far, in continuum mechanics based numerical simulations, the effect of interface has been ignored while evaluating the overall thermal properties of nano-composites. Therefore, in the present work, the effect of interface has been considered to evaluate the equivalent thermal conductivity of the composites. A meshless approach known as element free Galerkin method has been used as a tool for the numerical simulation. A nanoscale square representative volume element (square RVE) containing single CNT has been taken to evaluate the thermal properties of the composites.

2. Review of element free Galerkin method

The element free Galerkin (EFG) method requires the moving least square (MLS) approximation function for the discretization of a governing equation. These MLS approximation function consist of three components: a weight function associated with each node, a basis function, and a set of non-constant coefficients. Using MLS approximation scheme, an unknown function of temperature $T(\mathbf{x})$ is approximated with $T^h(\mathbf{x})$ given by [14]

$$T^h(\mathbf{x}) = \sum_{I=1}^{n} \Phi_I(\mathbf{x}) T_I = \boldsymbol{\Phi}(\mathbf{x})\mathbf{T} \tag{1}$$

where $\mathbf{x}^{\mathrm{T}} = [x \quad y \quad z]$, T_I are the nodal parameters, and $\Phi_I(\mathbf{x})$ is the shape function, which is defined as

$$\Phi_I(\mathbf{x}) = \sum_{j=1}^{m'} p_j(\mathbf{x}) \big(\mathbf{A}^{-1}(\mathbf{x})\mathbf{B}(\mathbf{x})\big)_{jI} = \mathbf{p}^{\mathrm{T}}\mathbf{A}^{-1}\mathbf{B}_I \tag{2a}$$

where

$$\mathbf{A} = \sum_{I=1}^{n} w(\mathbf{x}-\mathbf{x}_I)\mathbf{p}(\mathbf{x}_I)\mathbf{p}^{\mathrm{T}}(\mathbf{x}_I) \tag{2b}$$

$$\mathbf{B}(\mathbf{x}) = \big[w(\mathbf{x}-\mathbf{x}_1)\mathbf{p}(\mathbf{x}_1), w(\mathbf{x}-\mathbf{x}_2)\mathbf{p}(\mathbf{x}_2), \ldots, w(\mathbf{x}-\mathbf{x}_n)\mathbf{p}(\mathbf{x}_n)\big] \tag{2c}$$

The rational weight function [20] has been used in this work, which is given as

$$w(s) = \begin{cases} \frac{1}{s^{\bar{n}}+C} & 0 \leqslant s \leqslant 1 \\ 0 & s > 1 \end{cases} \tag{3}$$

where $2 \leqslant \bar{n} \leqslant 7$, $0.01 \leqslant C \leqslant 0.1$, $s = \|\mathbf{x}-\mathbf{x}_I\|/d_{mI}$ is the normalized radius, $d_{mI} = d_{\max}c_I$ and $d_{\max}$ = scaling parameter. The full details of EFG method can be found in [21].

3. Numerical implementation

A square RVE containing a single nanotube (Fig. 1) has been taken to evaluate the thermal conductivity of the composite. The carbon nanotube has been placed symmetrically at the center of the square RVE such that the axis of the RVE coincides with the axis of the nanotube. The 2-D left and right square surfaces of the RVE are maintained at two different constant temperatures T_1 and T_2 respectively, while other rectangular surfaces are kept insulated. The governing heat conduction equation in Cartesian coordinate system is given as

$$\frac{\partial}{\partial x}\left(k_x \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(k_y \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(k_z \frac{\partial T}{\partial z}\right) = 0 \tag{4a}$$

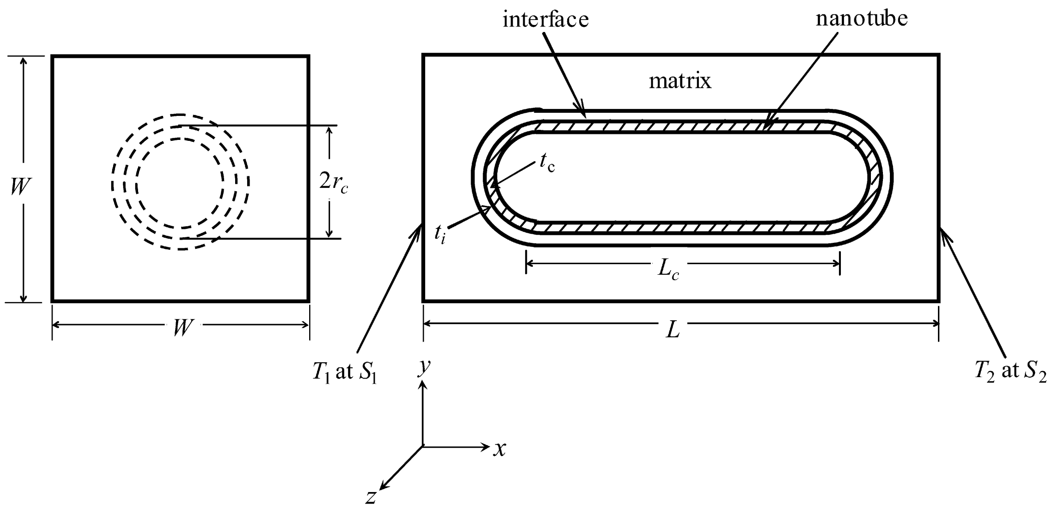

Fig. 1. Nano scale representation of a nano-composite.

The essential boundary conditions are

$$T(0,y,z)=T_1 \tag{4b}$$

$$T(L,y,z)=T_2 \tag{4c}$$

Continuity of temperature at the contact of matrix and interface materials requires

$$T|_m=T|_i \tag{4d}$$

Continuity of temperature at the contact of CNT and interface materials requires

$$T|_i=T|_c \tag{4e}$$

Continuity of normal heat flux at the contact of matrix and interface materials requires

$$q_n|_m=q_n|_i \tag{4f}$$

Continuity of normal heat flux at the contact of CNT and interface materials requires

$$q_n|_i=q_n|_c \tag{4g}$$

The weighted integral form of Eq. (4a) is given as

$$\sum_{j=m,i,c}\int_{V_j}\bar{w}\left\{\frac{\partial}{\partial x}\left(k_{jx}\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial y}\left(k_{jy}\frac{\partial T}{\partial y}\right)+\frac{\partial}{\partial z}\left(k_{jz}\frac{\partial T}{\partial z}\right)\right\}dV=0 \tag{5}$$

Using divergence theorem, the weak form of Eq. (5) is obtained as

$$\sum_{j=m,i,c}\int_{V_j}\left\{\frac{\partial\bar{w}}{\partial x}k_{jx}\frac{\partial T}{\partial x}+\frac{\partial\bar{w}}{\partial y}k_{jy}\frac{\partial T}{\partial y}+\frac{\partial\bar{w}}{\partial z}k_{jz}\frac{\partial T}{\partial z}\right\}dV=0 \tag{6}$$

From Eq. (6), the functional $I(T)$ can be obtained as

$$I(T)=\sum_{j=m,i,c}\int_{V_j}\frac{1}{2}\left\{k_{jx}\left(\frac{\partial T}{\partial x}\right)^2+k_{jy}\left(\frac{\partial T}{\partial y}\right)^2+k_{jz}\left(\frac{\partial T}{\partial z}\right)^2\right\}dV \tag{7}$$

Enforcing essential boundary conditions using Lagrange multiplier method, the functional $I^*(T)$ is obtained as

$$I^*(T)=\sum_{j=m,i,c}\int_{V_j}\frac{1}{2}\left\{k_{jx}\left(\frac{\partial T}{\partial x}\right)^2+k_{jy}\left(\frac{\partial T}{\partial y}\right)^2+k_{jz}\left(\frac{\partial T}{\partial z}\right)^2\right\}dV+\frac{\alpha}{2}\int_{S_1}(T-T_1)^2\,dS+\frac{\alpha}{2}\int_{S_2}(T-T_2)^2\,dS \tag{8}$$

Variation of $I^*(T)$ is given by

$$\delta I^*(T)=\sum_{j=m,i,c}\int_{V_j}\left\{k_{jx}\left(\frac{\partial T}{\partial x}\right)^{\mathrm{T}}\delta\left(\frac{\partial T}{\partial x}\right)+k_{jy}\left(\frac{\partial T}{\partial y}\right)^{\mathrm{T}}\delta\left(\frac{\partial T}{\partial y}\right)+k_{jz}\left(\frac{\partial T}{\partial z}\right)^{\mathrm{T}}\delta\left(\frac{\partial T}{\partial z}\right)\right\}dV+\alpha\int_{S_1}(T-T_1)\delta T\,dS+\alpha\int_{S_2}(T-T_2)\delta T\,dS \tag{9}$$

Setting $\delta I^*(T)=0$ for arbitrary δT in Eq. (9), results in the following set of linear equations

$$[\mathbf{K}]\{\mathbf{T}\}=\{\mathbf{f}\} \tag{10a}$$

where

$$K_{IJ}=\sum_{j=m,i,c}\int_{V_j}\begin{bmatrix}\Phi_{I,x}\\\Phi_{I,y}\\\Phi_{I,z}\end{bmatrix}^{\mathrm{T}}\begin{bmatrix}k_{jx}&0&0\\0&k_{jy}&0\\0&0&k_{jz}\end{bmatrix}\begin{bmatrix}\Phi_{I,x}\\\Phi_{I,y}\\\Phi_{I,z}\end{bmatrix}dV+\int_{S_1}\alpha\Phi_I^{\mathrm{T}}\Phi_J\,dS+\int_{S_2}\alpha\Phi_I^{\mathrm{T}}\Phi_J\,dS \tag{10b}$$

$$f_I=\int_{S_1}\alpha T_1\Phi_I\,dS+\int_{S_2}\alpha T_2\Phi_I\,dS \tag{10c}$$

Assuming the material properties as homogeneous, anisotropic and independent of temperature, the thermal conductivity of

Table 1
Data for CNT based composite problem

Parameter	Value of parameter
RVE length, L	10 µm
RVE cross-sectional dimension, W	40 nm
Nanotube length, L_c	2, 3, 4, ..., 8 µm
Nanotube outer radius, r_c	10 nm
Nanotube thickness, t_c	4 nm
Interface thickness, t_i	1, 2 and 4 nm
Thermal conductivity of matrix, $k_{mx} = k_{my} = k_{mz} = k_m$	0.27 W m^{-1} K^{-1}
Isotropic thermal conductivity of CNT, $k_{cx} = k_{cy} = k_{cz}$	3000 W m^{-1} K^{-1}
Temperature at S_1, T_1	300 K
Temperature at S_2, T_2	100 K

the aligned composite in longitudinal direction of nanotube has been evaluated as

$$k_e = -\frac{q_n L}{\Delta T} \tag{11}$$

where, k_e denotes the equivalent thermal conductivity of the composite, L is the length of RVE, q_n is the average normal heat flux, and ΔT is the temperature difference between left and right ends of RVE.

The percentage volume fraction of nanotube [14] has been calculated by the following expression

$$\upsilon = \left(\frac{V^c}{V^m + V^c}\right) \times 100 \tag{12}$$

where, υ is the volume fraction of CNT in composite, V^m is the volume of polymer matrix, and V^c is the volume of CNT including cavity.

4. Numerical results and discussion

The data required for the model composite problem have been tabulated in Table 1. Penalty approach has been used to enforce the constant temperatures at two square surfaces of RVE. Nanotube as well as interface domains have been discretized using uniform nodal distribution schemes. Three point Gauss quadrature scheme has been used for the numerical integration of Galerkin weak form.

4.1. *Effect of interface* (k_i)

Various values of interface conductivity (k_i) have been taken to study the effect of interface on the thermal conductivity of the composite. Fig. 2 shows the variation of effective thermal conductivity (k_e) of the composites with nanotube length (L_c) for various values of k_i and $t_i = t_c$. The effect of k_i on the effective thermal conductivity of the composite has been presented in Fig. 3 for $L_c = 5$ µm i.e. 9.83% of CNT. The variation of effective thermal conductivity of the composite with interface thickness (t_i) is shown in Fig. 4 for $L_c = 5$ µm i.e. 9.83% of CNT. From the results presented in Figs. 2–4, it can be seen that the interface thickness has a small effect on the effective thermal conductivity of the composite. For small nanotubes, the effect of interface is very small, whereas for long nanotubes, interface plays a significant role on the thermal conductivity of

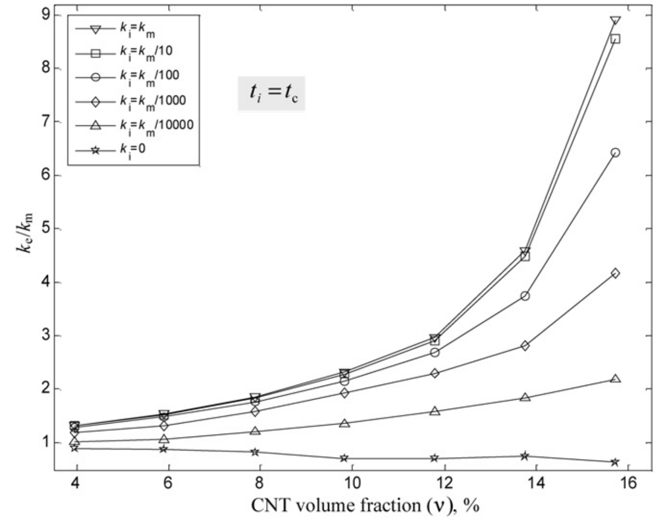

Fig. 2. Variation of effective thermal conductivity (k_e) with CNT volume fraction (ν).

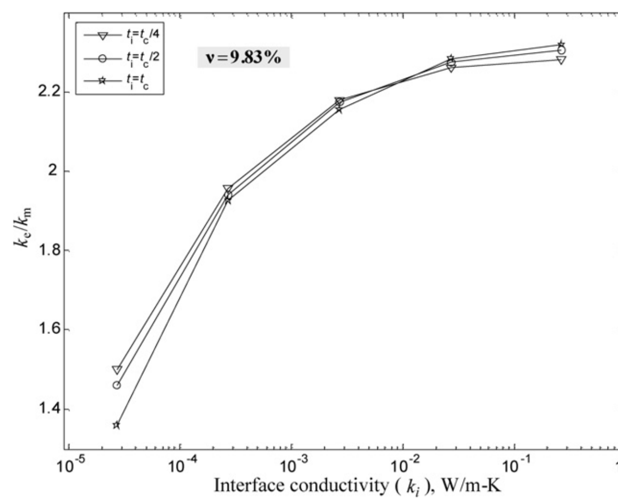

Fig. 3. Effect of interface conductivity (k_i) on the effective thermal conductivity (k_e) of the composite.

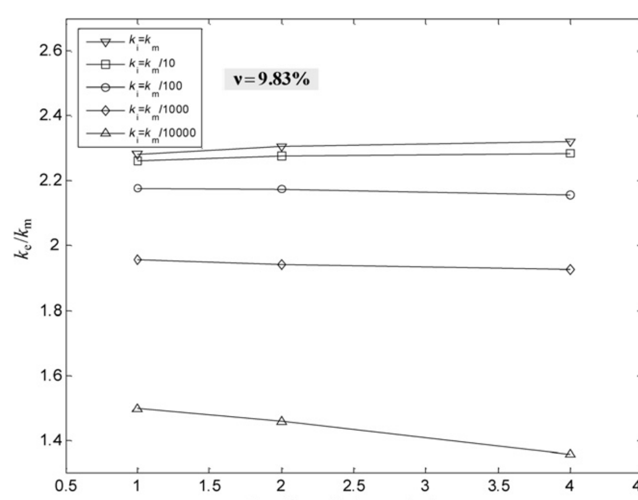

Fig. 4. Variation of effective thermal conductivity (k_e) with interface thickness (t_i).

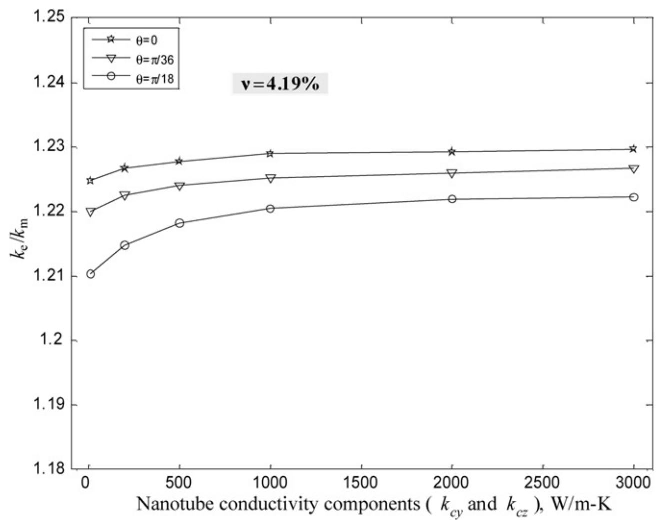

Fig. 5. Variation of nanotube anisotropy on thermal conductivity (k_e).

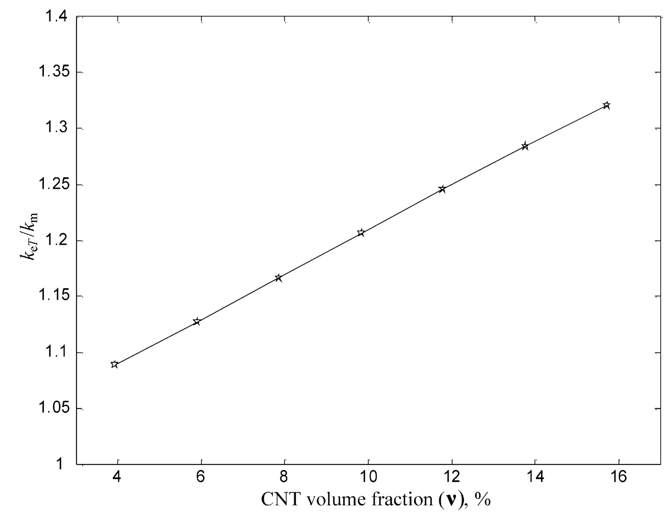

Fig. 6. Variation of effective transverse thermal conductivity (k_{eT}) with CNT volume fraction (ν).

the composite. Interface with zero conductivity decreases the overall conductivity of the composite.

4.2. *Effect of nanotube anisotropy*

The lengths of RVE and CNT have been taken as 1 μm and 200 nm respectively for this simulation. Various values of k_{cy} or k_{cz} have been taken to study the effect of nanotube anisotropy on the equivalent thermal conductivity of the composite. To get a proper effect of anisotropy, nanotube has been placed in RVE such that it makes an angle θ with the axis of RVE. The variation of effective thermal conductivity (k_e) of the composites with nanotube length for various values of k_{cy} or k_{cz} is shown in Fig. 5 for $L_c = 200$ nm i.e. 4.19% of CNT. The equivalent thermal conductivity of the composite is shown in Fig. 5 for three different values of θ. From Fig. 5, it can be concluded that the effect of nanotube anisotropy is almost negligible when nanotube axis coincides with the axis of RVE i.e. $\theta = 0$, whereas the effect of anisotropy is very small when the nanotube axis makes a non-zero angle with the axis of RVE (i.e. say $\theta = \pi/18$).

4.3. *Transverse thermal conductivity* (k_{eT})

To evaluate the transverse properties of the composites, two different constant values of temperature (i.e. 300 K and 100 K) have been applied at two opposite rectangular faces of the RVE. Various parameters used to evaluate the transverse thermal conductivity of the composite have been provided in Table 1. The effective transverse thermal conductivity of the composite (k_{eT}) evaluated by Eq. (11) has been presented in Fig. 6 for the various values of nanotube length. 3.95% of CNT addition (i.e. equivalent to $L_c = 2$ μm) increases the transverse thermal conductivity of the composites by 8.9%, whereas 15.7% of CNT addition (i.e. equivalent to $L_c = 8$ μm) increases the transverse thermal conductivity of the composite by 32%. From the results presented in Fig. 6, it is observed that the variation of transverse thermal conductivity is almost linear with nanotube length.

Table 2
Variation of equivalent thermal conductivity (k_e) of composite with number of CNTs

Parameters	Number of CNTs in RVE			
	one	two	three	four
% volume fraction of CNT in RVE (ν)	11.31	22.62	33.93	45.24
Equivalent thermal conductivity (k_e)	2.6727	2.6766	2.6820	2.6866
Increase of conductivity (k_e/k_m)	9.90	9.91	9.93	9.95
Effectiveness of enhancement (k_e/ν)	23.63	11.83	7.94	5.94

4.4. *Numerical results with multiple CNTs inside RVE*

The length as well as the radius of all CNTs has been kept constant for this simulation, therefore as the number of CNTs increases; the amount of CNT in RVE also increases. The CNTs have been enforced in a fixed portion of RVE. Table 2 shows a variation of equivalent thermal conductivity of the composite with the number of CNTs. The equivalent thermal conductivity of the composite increases to 2.6727 $\mathrm{W\,m^{-1}\,K^{-1}}$ i.e. 9.90 times that of polymer matrix for single CNT (11.31%), 2.6766 $\mathrm{W\,m^{-1}\,K^{-1}}$ i.e. 9.91 times of polymer matrix for two CNTs (22.62%), 2.6820 $\mathrm{W\,m^{-1}\,K^{-1}}$ i.e. 9.93 times of polymer matrix for three CNTs (33.93%), and 2.6820 $\mathrm{W\,m^{-1}\,K^{-1}}$ i.e. 9.95 times of polymer matrix for four CNTs (45.24%) respectively. From the results presented in Table 2, it can be noted that there is a very minor improvement in the equivalent thermal conductivity with the increase of CNTs. The main reason for this minor improvement is that the effective resistance between two ends of RVEs remains almost constant until the same length CNTs are enforced inside a fixed portion of RVE.

From above analysis, it can be inferred that the interface plays a significant role in the effective conductivity of the composites. It can be stated that the interface resistance is the principal factor for lower thermal conductivity of the composite as expected by many researchers. Anisotropy of nanotube makes a very small impact on the effective thermal conductivity of the composite. The variation of transverse thermal conductivity of the composite with nanotube length has found linear. Moreover, the amount of CNTs inside RVE may not always increase the equivalent thermal conductivity of the composite.

5. Conclusions

In this paper, the effect of interface on the thermal conductivity of the composite was studied by element free Galerkin method. The numerical results were obtained for a model problem using continuum mechanics approach. The present analysis shows that the effect of interface on the overall conductivity of composite is small for short nanotubes, whereas interface has a significant effect on the overall thermal conductivity of the composite for long nanotubes. The interface thickness has got small effect on the thermal conductivity of the composites. Effect of nanotube anisotropy is very small on the overall thermal conductivity of the composite in longitudinal direction. Transverse thermal conductivity has got linear variation with nanotube length. The addition of CNTs inside RVE may not increase the equivalent thermal conductivity of the composite without proper reinforcement.

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