Effective Thermal Conductivities of a Novel Fuzzy Fiber-Reinforced Composite Containing Wavy Carbon Nanotubes

This article deals with the investigation of the effect of carbon nanotube (CNT) waviness on the effective thermal conductivities of a novel fuzzy fiber-reinforced composite (FFRC). The distinctive feature of the construction of this novel FFRC is that wavy CNTs are radially grown on the circumferential surfaces of the carbon fibers. Effective thermal conductivities of the FFRC are determined by developing the method of cells (MOCs) approach in conjunction with the effective medium (EM) approach. The effect of CNT waviness is studied when wavy CNTs are coplanar with either of the two mutually orthogonal planes of the carbon fiber. The present study reveals that (i) if CNT waviness is parallel to the carbon fiber axis then the axial ($K_1$) and the transverse ($K_2$) thermal conductivities of the FFRC are improved by 86% and 640%, respectively, over those of the base composite when the CNT volume faction present in the FFRC is 16.5% and the temperature is 400 K, (ii) the effective value of $K_1$ of the FFRC containing wavy CNTs being coplanar with the carbon fiber axis is enhanced by 75% over that of containing straight CNTs for the fixed CNT volume faction when the temperature is 400 K, and (iii) the CNT/polymer matrix interfacial thermal resistance does not affect the effective thermal conductivities of the FFRC. The present work also reveals that for a particular value of the CNT volume fraction, optimum values of the CNT waviness parameters, such as the amplitude and the wave frequency of the CNT for improving the effective thermal conductivities of the FFRC can be estimated. [DOI: 10.1115/1.4028762]

Keywords: carbon nanotube waviness, nanocomposite, hybrid composite, thermal conductivity

1 Introduction

As nanoscale graphite structures, CNTs are of great interest not only for their mechanical properties but also for their thermal properties [1–4]. CNTs exhibit thermal properties that are remarkably different from other known materials and are expected to be a promising candidate in many advanced thermal management applications. The quest for utilizing such exceptional thermal properties of CNTs created enormous interest among the researchers for developing the two-phase CNT-reinforced composites [5–14]. Literature on the two-phase CNT-reinforced composites authenticate that the dispersion of highly conductive long CNTs into the matrix results in the enhancement in the effective thermal conductivities of the resulting nanocomposite. However, manufacturing of such two-phase nanocomposites reinforced with long CNTs has some challenging technical issues, such as agglomeration, misalignment, adhesion of CNTs in polymer matrix and difficulty in manufacturing long CNTs [15,16]. These difficulties can be alleviated by using CNTs as secondary reinforcements in the three-phase hybrid CNT-reinforced composites. In this case, CNTs are radially grown on the circumferential surfaces of the fiber reinforcements. For example, Thostenson et al. [17] synthesized CNTs on the circumferential surfaces of the carbon fibers using chemical vapor deposition technique and found that the presence of CNTs at the fiber/matrix interface improves the interfacial shear strength of the hybrid composite. Veedu et al. [18] fabricated the multifunctional composite in which the vertical arrays of CNTs are grown on the circumferential surfaces of the fibers. They reported that the presence of CNTs in the transverse direction of the composite enhances the effective thermal conductivity up to 51% compared to that of the base composite, and also alleviate the problem of agglomeration of CNTs. Fiber augmented with the radially grown CNTs on its circumferential surface is being called as “fuzzy fiber” [19] and the resulting composite is called as FFRC [19,20]. Recently, Kundalwal and Ray [21] predicted that the transverse effective thermal conductivities of the FFRC containing straight CNTs are significantly enhanced over their values without CNTs. Thus, the current status of progress in research on the CNT-reinforced composites brings to light that the three-phase hybrid CNT-reinforced composite, such as FFRC can be the promising candidate material for achieving the better thermal management benefits from the exceptionally conductive CNTs. CNTs embedded in nanocomposites are not straight but rather have certain degree of curvature or waviness that arises from their high aspect ratio, low bending stiffness, and processing induced effects [16]. Scanning electron microscopy images analyzed by Zhang et al. [22] and Yamamoto et al. [23] are shown in Figs. 1(a) and 1(b), respectively, which clearly demonstrate that CNTs are highly curved when dispersed in the polymer matrix. Waviness of CNTs is a prevailing factor in two- and three-phase CNT-reinforced composites, and has considerable effect on their effective properties [24–28]. It may be worth mentioning that the previously published work by the present authors [21] is concerned with the estimation of the effective thermal conductivities of the FFRC containing straight CNTs. Since the distribution
pattern and waviness of CNTs influence the effective properties of the CNT-reinforced composite, CNT waviness will also influence the thermal conductivities of the FFRC. Investigation of the effect of CNT waviness on the thermal conductivities of the FFRC is an important issue and has not yet been addressed—which provide the scope and the focus for the present study. It is therefore the objective of this work to investigate the effect of CNT waviness on the effective thermal conductivities of the FFRC. We account for CNT waviness by considering the plane of wavy CNTs is coplanar with either the longitudinal plane or the transverse plane of the carbon fiber.

2 Effective Thermal Conductivities of a Novel FFRC Containing Wavy CNTs

The schematic illustrated in Fig. 2 represents a lamina of the FFRC being studied here. In this novel composite, wavy CNTs are radially grown and uniformly spaced on the circumferential surfaces of the carbon fibers. In the present study, wavy CNTs are modeled as sinusoidal solid CNTs [25–28] while at any location along the length of the CNT wave, CNT is considered as the transversely isotropic [29]. The orientation of the plane of wavy CNTs will influence the effective properties of the FFRC [24]. Hence, two possible planar orientations of wavy CNTs are considered either in the transverse (2–3) planes or the longitudinal (1–3) planes of the carbon fiber, as shown in Figs. 3(a) and 3(b), respectively. In case of wavy CNTs being coplanar with the 2–3 planes, the amplitudes of the CNT waves are transverse to the axis of the carbon fiber while the amplitudes of the CNT waves being coplanar with the 1–3 planes are parallel to the carbon fiber axis. The radially grown wavy CNTs reinforce the polymer matrix surrounding the carbon fiber along the transverse direction to its length. Thus, the augmented fuzzy fiber can be viewed as a circular cylindrical composite fuzzy fiber (CFF) in which the carbon fiber is embedded in the CNT-reinforced polymer matrix nanocomposite (PMNC) and the radius of the CFF equals the sum of the radius of the carbon fiber and the linear distance between the ends of the CNT wave. The cross sections of such CFF containing wavy CNTs being coplanar with either the 2–3 planes or the 1–3 planes are shown in Figs. 4(a) and 4(b), respectively. Consequently, the representative volume element (RVE) of the FFRC can be treated as being composed of the two phases wherein the reinforcement is the CFF and the matrix is the polymer material.

At this juncture, it is important to mention that the CNT–CFF contact is not considered to be present in the PMNC material. Constructional feature of the PMNC is assumed to be such that wavy CNTs are uniformly dispersed in the polymer material and the gaps between them are filled up with the polymer. CNTs with random distributions in the matrix material may form a percolating network even at low volume fraction due to their high aspect ratio and inter-CNT contacts (see Fig. 1). Contact regions between CNTs affect the heat transfer performance of the CNT-reinforced composite. These contact points serve both as scattering sites for phonons propagating along contacting CNTs, reducing the individual CNT thermal conductivity, and as a route for heat conduction between the CNTs with additional thermal resistance across the interface [12]. Prasher et al. [30] indicated that the thermal conductivity of a randomly oriented bed of multiwalled CNTs can be controlled by the CNT–CFF thermal contact resistance which is orders of magnitude larger than the intrinsic thermal resistance of a CNT. They reported that if the interfacial thermal resistance between CNTs is small, as the volume fraction increases and more CNT–CFF contacts are formed, the composite thermal conductivity increases. Recently, progress has been made for manufacturing aligned CNT-reinforced composites by using novel fabrication techniques, such as chemical vapor deposition. Therefore, in the present study, only the effects of CNT waviness and CNT/matrix interfacial thermal resistance on the thermal conductivities of the FFRC are studied assuming that the inter-CNT contacts are not present in the PMNC material.

2.1 Models of the Wavy CNTs. The constructional feature of the CFF can further be viewed as concentric cylinders in which the carbon fiber is wrapped by a lamina of the PMNC material as illustrated in Fig. 5. The RVE of the unwound PMNC material containing a wavy CNT has been illustrated in Fig. 6. As shown in this figure, the RVE can be divided into infinitesimally thin slices of thickness dy. Averaging the effective thermal conductivities of these slices over the length $L_y$ of the RVE, the homogenized effective thermal conductivities of the unwound PMNC material can be estimated. The validity of such technique has been verified.
by several researchers [28,31,32] against experimental results. Now, the CNT wave is defined by

\[ x = A \sin(\omega y) \quad \text{or} \quad z = A \sin(\omega y); \quad \omega = \frac{n\pi}{L_{\text{nr}}} \]  

(1)

According to the CNT wave is coplanar with the 1–3 planes or the 2–3 planes, respectively. In Eq. (1), \( A \) and \( L_{\text{nr}} \) are the amplitude of the CNT wave and the linear distance between the ends of the CNT wave, respectively, and \( n \) represents the number of the CNT waves. The running length of the CNT wave \( (L_{\text{nr}}) \) can be expressed in the following form:

\[ L_{\text{nr}} = \int_{0}^{L_{\text{rf}}} \sqrt{1 + A^2 \omega^2 \cos^2(\omega y)} dy \]  

(2)

in which the angle \( \phi \) shown in Fig. 6 is given by

\[ \tan \phi = \frac{dx}{dy} = A\omega \cos(\omega y) \quad \text{or} \quad \tan \phi = \frac{dz}{dy} = A\omega \cos(\omega y) \]  

(3)

According to the wavy CNT is coplanar with the 1–3 or the 2–3 planes, respectively. Note that for a particular value of \( \omega \), the value of \( \phi \) varies with the amplitude of the CNT wave.

2.2 Modeling Approaches. This section deals with the procedures of employing two modeling approaches; namely, the MOC approach and the EM approach to predict the effective thermal conductivities of the FFRC. The various steps involved in the modeling of the FFRC are outlined as follows:
First, the effective thermal conductivities of the PMNC are to be determined by using either the MOC approach considering the perfect CNT/polymer matrix interface \( R_k = 0 \) or the EM approach incorporating the CNT/polymer matrix interfacial thermal resistance \( R_k \neq 0 \) where \( R_k \) is the CNT/polymer matrix interfacial thermal resistance.

Utilizing the thermal conductivities of the PMNC and the carbon fiber, the effective thermal conductivities of the CFF are to be determined by using the MOC approach.

Finally, using the thermal conductivities of the CFF and the polymer matrix, the effective thermal conductivities of the FFRC can be estimated by employing the MOC approach.

The FFRC and its constituent phases being studied here are made of aligned reinforcements. The MOC approach is an efficient analytical model to predict the effective orthotropic thermal conductivities of aligned fiber-reinforced composite considering the orthotropic constituent phases. On the other hand, an EM approach predicts the effective orthotropic thermal conductivities of aligned fiber-reinforced composite considering the isotropic constituents only. Hence, the MOC approach has been utilized to determine the effective thermal conductivities of the CFF and the FFRC—since the PMNC and the CFF are the transversely isotropic.

### 2.2.1 MOC Approach

This section presents the development of the MOC approach to estimate the effective thermal conductivities of the PMNC, the CFF, and the FFRC. The effective thermal conductivities at any point of the unwound PMNC lamina containing wavy CNTs can be estimated by transforming the effective thermal conductivities of the unwound PMNC lamina containing straight CNTs. Hence, the development of the MOC approach [33] for predicting the effective thermal conductivities of the unwound PMNC lamina containing straight CNTs will be presented first.

Let us first develop the micromechanics model using the MOC approach for the case of a doubly periodic representative unit cell, which represents a continuously reinforced PMNC. The continuum model for the unidirectional PMNC is based on the assumption that the continuous solid CNTs [5,6,25–28] extend in the \( x_3 \)-direction and are arranged in a doubly periodic array in the \( x_1 \)-and \( x_2 \)-directions (see Fig. 7). The cross section of the CNT is \( h_1 b_1 ; h_2 \) and \( b_2 \) represent its spacing in the polymer matrix. As a result of this periodic arrangement, it is sufficient to analyze the representative unit cell as shown in Fig. 7. This representative unit cell contains four subcells, and each rectangular subcell is labeled by \( \beta \gamma \), with \( \beta \) and \( \gamma \) denoting the location of the subcell along the \( x_1 \)-and \( x_2 \)-directions, respectively. Let four local coordinate systems \( (x_1^{(\beta)}, x_2^{(\gamma)}, x_3) \) be introduced, all of which have origins that are located at the centroid of each subcell. Since the average
behavior of the composite is sought, a first order theory in which the temperature deviation in the subcell is expanded linearly in terms of the distances from the center of the subcell \((x_1^{(p)})\) and \((x_2^{(p)})\) can be used. Following the first order theory, the deviation of the temperature from a reference temperature \(T_R\) (at which the material is stress free when its strain is zero), \(\Delta T^{(p)}\), is expanded in the following form:

\[
\Delta T^{(p)} = \Delta T + x_1^{(p)} \frac{\partial T}{\partial x_1} + x_2^{(p)} \frac{\partial T}{\partial x_2}
\]

(4)

where \(x_1^{(p)}\) and \(x_2^{(p)}\) characterize the linear dependence of the temperature on the local coordinates. The volume \((V_{\beta i})\) of each subcell is

\[
V_{\beta i} = h_b b_i l
\]

(5)

where \(h_b\), \(b_i\), and \(l\) denote the height, the width, and the length of the subcell, respectively, while the volume \((V)\) of the repeating unit cell is

\[
V = hbl
\]

(6)

The continuity conditions of the temperature at the interfaces of the subcells on an average basis lead to the following relations:

\[
h_1 x_1^{(1)} + h_2 x_1^{(2)} = (h_1 + h_2) \frac{\partial T}{\partial x_1} \quad \text{and} \quad b_2 x_2^{(2)} + b_2 x_2^{(1)} = (b_1 + b_2) \frac{\partial T}{\partial x_2}
\]

(7)

For the average heat flux in the subcell

\[
q_i^{(\beta)} = -K_i^{(\beta)} \frac{\partial T}{\partial x_i}; \quad i = 1, 2, \text{ and } 3
\]

(8)

where \(K_i^{(\beta)}\) denote the thermal conductivity coefficients of the subcells.

The average heat flux in the unwound PMNC material is determined from the following relation:

\[
q_i = \frac{1}{V} \sum_{\beta=1}^{2} V_{\beta i} q_i^{(\beta)}
\]

(9)

The continuity conditions of the heat flux at the interfaces of the subcells yield

\[
q_1^{(1)} = q_1^{(2)} \quad \text{and} \quad q_2^{(1)} = q_2^{(2)}
\]

(10)

The average heat flux components are related to the temperature gradients by the effective thermal conductivity coefficients \((K_{nc}^{(\beta)})\)

\[
q_i = -K_{nc}^{(\beta)} \frac{\partial T}{\partial x_i}
\]

(11)

By eliminating the microvariables \(x_1^{(\beta)}\) and \(x_2^{(\beta)}\), and using the continuity conditions, the effective thermal conductivities of unidirectional unwound PMNC lamina are given by [33]

\[
K_{nc}^{(1)} = \frac{K_p [K_p [b(V_{11} + V_{21}) + h_3 (V_{12} + V_{22})] + K_p h_1 (V_{12} + V_{22})]}{hbl [K_p h_1 + K_p h_2]}
\]

and

\[
K_{nc}^{(2)} = \frac{K_p [K_p [b(V_{11} + V_{12}) + b_2 (V_{21} + V_{22})] + K_p b_1 (V_{21} + V_{22})]}{hbl [K_p h_1 + K_p b_2]}
\]

(12)

In Eq. (12), the respective superscripts \(nc, n\), and \(p\) represent the unwound PMNC material containing straight CNTs, the CNT, and the polymer material. The effective thermal conductivities \((K_{nc}^{(R)})\) at any point in the unwound PMNC lamina where the CNT is inclined at an angle \(\phi\) with the \((3\text{-axis})\)-axis can be derived in a straightforward manner by employing the appropriate transformation law as follows [34]:

\[
[K^{nc}] = [T_1]^{-1} [K^{nc}_c] [T_1]^{-1} \quad \text{and} \quad [K^{nc}] = [T_2]^{-1} [K^{nc}_c] [T_2]^{-1}
\]

(13)

According to the wavy CNT is coplanar with the 1–3 or the 2–3 planes, respectively. The various matrices appeared in Eq. (13) are given by

\[
[K^{nc}_c] = \begin{bmatrix}
K^{nc}_1 & 0 & 0 \\
0 & K^{nc}_2 & 0 \\
0 & 0 & K^{nc}_3
\end{bmatrix}, \quad [T_1] = \begin{bmatrix}
\cos \phi & 0 & \sin \phi \\
0 & 1 & 0 \\
-\sin \phi & 0 & \cos \phi
\end{bmatrix}, \quad \text{and} \quad [T_2] = \begin{bmatrix}
1 & 0 & 0 \\
0 & \cos \phi & \sin \phi \\
0 & -\sin \phi & \cos \phi
\end{bmatrix}
\]

in which \(\cos \phi = \left[1 + \{n \pi A/L_w \cos(n \Delta y/L_w)\}^2\right]^{-1/2}\) and

\(\sin \phi = n \pi A/L_w \cos(n \pi y/L_w) \left[1 + \{n \pi A/L_w \cos(n \pi y/L_w)\}^2\right]^{-1/2}\)

It is now obvious that the effective thermal conductivities of the unwound PMNC lamina containing straight CNTs vary along the length of the CNT wave as the value of \(\phi\) vary over the length of the CNT. The average effective thermal conductivity matrix \([K^{nc}]\) of the lamina of such unwound PMNC material containing wavy CNTs can be obtained by averaging the transformed thermal conductivity coefficients over the linear distance between the CNT ends as follows [31]:

\[
[K^{nc}] = \frac{1}{\ell} \int_0^{\ell} [K^{nc}_c] dy
\]

(14)

It may also be noted that when the carbon fiber is viewed to be wrapped by such unwound lamina of the PMNC, the matrix \([K^{nc}]\) provides the effective thermal conductivities at a point located in the PMNC where the CNT axis (3-axis) is oriented at an angle \(\theta\) with the 3-axis in the 2–3 planes as shown in Figs. 4 and 5. Hence, at any point in the PMNC surrounding the carbon fiber, the effective thermal conductivity matrix \([K^{PMNC}]\) of the PMNC with respect to the principal coordinate system (1–2–3) turns out to be location dependant and can be determined by the following transformations [34]:

\[
[K^{PMNC}] = \begin{bmatrix}
1 & 0 & 0 \\
0 & \cos \theta & \sin \theta \\
0 & -\sin \theta & \cos \theta
\end{bmatrix}^{-1} \begin{bmatrix}
K^{nc}_1 & 0 & 0 \\
0 & K^{nc}_2 & 0 \\
0 & 0 & K^{nc}_3
\end{bmatrix}
\]

\[
\times \begin{bmatrix}
1 & 0 & 0 \\
0 & \cos \theta & \sin \theta \\
0 & -\sin \theta & \cos \theta
\end{bmatrix}
\]

(15)

Finally, a simple homogenization process can be carried out on the RVE of an annular cross section of the PMNC phase to compute its effective thermal conductivities [34,35] as follows:

\[
[K^{PMNC}] = \frac{1}{\pi (R^2 - a^2)} \int_0^{2\pi} \int_a^R [K^{PMNC}] r dr d\theta
\]

(16)
where $R$ and $a$ denote the outer and the inner radii of an annular cross section of the PMNC, respectively. It may be noted here that if the CNT volume fraction is homogenized in the annular portion of the RVE of the PMNC, the homogenized effective thermal conductivities of the PMNC will not be radially dependent (see Appendix A of Ref. [21] for details).

It is worthwhile to note that the thermal conductivity matrix of the homogenized PMNC $K_{\text{PMNC}}^{\text{eff}}$ is the transversely isotropic and its axis of symmetry is the 1- or $x_1$-axis. In order to model the CFF by the MOC approach, the CFF is considered to be composed of cells periodically arranged along the $x_2$- and $x_3$-directions while each cell consists of $\beta_1$ number of subcells as shown in Fig. 7. In this case, each repeating unit cell represents the CFF and the sub-cell is composed of either the carbon fiber or the PMNC. Now, considering the carbon fiber and the PMNC with its thermal conductivities given by Eq. (16), the MOC approach can be augmented to estimate the effective thermal conductivities of the CFF; such that

$$K_{\text{CFF}}^1 = \frac{K_{\text{CFF}}V_{11} + K_{\text{PMNC}}^1(V_{12} + V_{21} + V_{22})}{\text{hbl}},$$

$$K_{\text{CFF}}^2 = \frac{K_{\text{PMNC}}^2[K_{\text{CFF}}^2[h(V_{11} + V_{21}) + h_2(V_{12} + V_{22})] + K_{\text{PMNC}}^2h_1(V_{12} + V_{22})]}{\text{hbl}(K_{\text{PMNC}}^2h_1 + K^F_{\text{CFF}}h_2)},$$

$$K_{\text{CFF}}^3 = \frac{K_{\text{PMNC}}^2[K_{\text{CFF}}^2[b(V_{11} + V_{21}) + b_2(V_{12} + V_{22})] + K_{\text{PMNC}}^2h_1(V_{12} + V_{22})]}{\text{hbl}(K_{\text{PMNC}}^2b_1 + K^F_{\text{CFF}}b_2)},$$

In the above equation, the respective superscripts $f$ and $CFF$ denote the carbon fiber and the CFF.

Finally, considering the CFF as the cylindrical inclusion embedded in the polymer matrix, the effective thermal conductivities of the FFRC can be obtained by utilizing the MOC approach as follows:

$$K_{\text{1}} = \frac{K_{\text{CFF}}V_{11} + K_{\text{pol}}(V_{12} + V_{21} + V_{22})}{\text{hbl}},$$

$$K_{\text{2}} = \frac{K_{\text{pol}}[K_{\text{CFF}}^2[h(V_{11} + V_{21}) + h_2(V_{12} + V_{22})] + K_{\text{pol}}h_1(V_{12} + V_{22})]}{\text{hbl}(K_{\text{pol}}h_1 + K^F_{\text{CFF}}h_2)},$$

$$K_{\text{3}} = \frac{K_{\text{pol}}[K_{\text{CFF}}^2[b(V_{11} + V_{21}) + b_2(V_{12} + V_{22})] + K_{\text{pol}}h_1(V_{12} + V_{22})]}{\text{hbl}(K_{\text{pol}}b_1 + K^F_{\text{CFF}}b_2)}.$$  

### 3 Results and Discussion

In this section, the predictions by the MOC and the EM approaches are compared with the existing experimental results of aligned CNT-reinforced polymer composite. Subsequently, the effective thermal conductivities of the FFRC have been determined.

#### 3.1 Comparisons With the Experimental Results

The predictions by the MOC and the EM approaches developed in the present study are first compared with the experimental results by Marconnet et al. [12], Marconnet et al. [12] fabricated aligned CNT-polymer nanocomposite consisting of CNTs arrays infiltrated with an aerospace-grade thermostet epoxy. In their study, the axial ($K_a$) and the transverse ($K_T$) thermal conductivities of aligned CNT-polymer nanocomposite are reported considering an alignment factor of CNTs as 0.77. Reported values of ($K_a$) and ($K_T$) are also found to be in good agreement with those of the values estimated by using the EM approach. However, in this study, the angle between the material axis and the local CNT axis, and the distribution function describing the CNT orientation are not reported. To the best of authors’ knowledge, there are no experimental thermal conductivity data currently available for aligned CNT-reinforced composite other than this study. Hence, for the comparison purpose, it is assumed that CNTs are perfectly aligned (that is, alignment factor 1) in the polymer matrix. For such comparison, thermal conductivities of the multiwalled CNT ($K^w$) and the polymer material ($K^p$) are taken as 22.1 W/mK and 0.26 W/mK, respectively, as considered by Marconnet et al. [12].

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Figures 8(a) and 8(b) demonstrate the outcome of this comparison. In these figures, dotted line represents best fits obtained from the EM approach for the experimental results considering an alignment factor of CNTs as 0.77 [12]. Figure 8(a) reveals that the effective values of \( K_A \) predicted by the MOC and the EM approaches overestimate the experimental values of \( K_A \) by \( \approx 21\% \) and \( \approx 23\% \) when the CNT volume fractions are 0.07 and 0.16, respectively. On the other hand, the MOC and the EM approaches underestimate the values of \( K_T \) by 40% and 58% when the CNT volume fractions are 0.07 and 0.16, respectively. These differences between the results are attributed to the fact that the perfect alignments of CNTs are considered while computing the results by the MOC and the EM approaches whereas the value of alignment factor is 0.77 in Ref. [12]. Other possible reasons for the disparity between the analytical and the experimental results include the CNT/matrix interfacial thermal resistance, lattice defects within CNTs and modification of the phonon conduction within CNTs due to the interactions with the matrix [7,12]. It may also be observed from Fig. 8(a) that the prediction by the MOC and the EM approaches shows excellent agreement for the values of \( K_A \). This is attributed to the fact that these models provide a relation identical to the rule of mixtures [33,36]; see Eqs. (12) and (20). On the other hand, the marginal differences can be observed for the values of \( K_T \) predicted by the MOC and the EM approaches as shown in Fig. 8(b). Thus, it can be inferred from these comparisons that the MOC and the EM approaches can be reliably applied to predict the thermal conductivities of the FFRC and its constituent phases.

3.2 Analytical Modeling Results. To present the numerical results, the thermal conductivities of the constituent phases of the FFRC are considered to be temperature dependent. The thermal conductivities of the armchair (10,10) CNT, the carbon fiber and the polymer material are taken as \( 3.8 \times 10^3 \) W/mK, \( 3.1 \times 10^3 \) W/mK [3], \( 222 \) W/mK–912 W/mK [38], \( 0.16 \) W/mK–0.205 W/mK [39], respectively, for the temperature range 100 K–400 K. The thermal conductivities of these phases are nonlinear functions of change in the temperature and are demonstrated in Fig. 9. Following these figures, the relationships between the thermal conductivities of the constituent phases of the FFRC and the temperature can be written as follows:

\[
K_A = -2.3476 \times 10^{-18} T^{10} + 5.1847 \times 10^{-15} T^9 \\
- 4.9368 \times 10^{-12} T^8 + 2.6646 \times 10^{-9} T^7 \\
- 8.744 \times 10^{-7} T^6 + 1.8296 \times 10^{-4} T^5 - 0.02393 T^4 \\
+ 1.88883 T^3 - 85.36672 + 2256.47 W/mK
\]  \tag{21}

\[
K_T = 1.4235 \times 10^{-7} T^4 - 1.0199 \times 10^{-4} T^3 + 0.021449 T^2 \\
+ 0.8331 T W/mK
\]  \tag{22}

\[
K_p = -1.2805 \times 10^{-15} T^6 + 1.8231 \times 10^{-12} T^5 \\
- 1.0343 \times 10^{-9} T^4 + 2.9748 \times 10^{-7} T^3 \\
- 4.6272 \times 10^{-5} T^2 - 0.0040278 W/mK
\]  \tag{23}

In the FFRC, hexagonal packing array of the CFFs is considered as shown in Fig. 2 for evaluating the numerical results. It is obvious that the constructional feature of the FFRC imposes a constraint on the maximum value of the CNT volume fraction. The maximum value of the CNT volume fraction in the FFRC can be determined based on the surface-to-surface distance at the roots of two adjacent CNTs as 0.0017 \( \mu \)m [24]. CNT diameter (\( d_n \)), running length of sinusoidally CNT wave (\( L_w \)) and volume fraction of the carbon fiber (\( V_f \)) as follows [24]:

\[
V_{CNF} = \frac{\pi d^2 L_w V_f}{d^2 (d_n + 0.0017)^2}
\]  \tag{24}

The value of \( d_n \) of the armchair (10,10) CNT is considered as 0.00136 \( \mu \)m [29]. Unless otherwise mentioned, the effective thermal conductivities of the PMNC are computed by employing the MOC approach considering the perfect CNT/polymer matrix interface (that is, \( R_k = 0 \)). Subsequently, the estimated effective thermal conductivities of the PMNC are used to compute the effective thermal conductivities of the CFF. Table 1 summarizes the effective thermal conductivities of the PMNC and the CFF for the particular values \( T \). The effect of CNT waviness on the effective thermal conductivities of the FFRC is investigated when wavy CNTs are coplanar with either of the two mutually orthogonal planes. For such investigation, the volume fraction of the carbon fiber (\( V_f \)) in the FFRC, the wave frequency of the CNT (\( \omega \)) and the maximum amplitude of the CNT wave (\( A \)) are
considered as 0.5, 18π/L₀ µm⁻¹, and 100d₀ µm, respectively. Figure 10(a) illustrates the variation of the axial thermal conductivity \((K₁)\) of the FFRC with the temperature. This figure reveals that if the variations of the amplitude of wavy CNTs are in the 1–3 planes, the effective values of \(K₁\) are significantly improved over those of the FFRC containing either wavy CNTs being coplanar with the 2–3 planes or straight CNTs. It may also be noted that the trend of this result follows the trend of the thermal conductivity \((Kn)\) when wavy CNTs are coplanar with the 1–3 planes. This is due to the fact that the thermal conductivity \((Kₙ)\) of the polymer material \((Kₚ)\) of parts of wavy CNT length with the 1-axis leading to the increase in the axial thermal conductivity of the PMNC along the 1-axis and hence, the axial thermal conductivity of the FFRC increases. It may also be observed from Fig. 10(a) that the effective values of \(K₁\) decrease with the increase in the temperature when wavy CNTs are coplanar with the 1–3 planes. This is due to the fact that the thermal conductivity \((Kₚ)\) of the armchair CNT (10, 10) decreases with the increase in the temperature (see Fig. 9(a)) which eventually do not enhance the effective values of \(K₁\) of the FFRC.

The effective values of \(K₁\) of the FFRC containing either wavy CNTs being coplanar with the 2–3 planes or straight CNTs \((\omega = 0)\) are not improved over those of the base composite \((Vₜ = 0)\). In these cases, CNTs are coplanar with the transverse plane of the carbon fiber, and do not influence the effective values of \(K₁\) of the FFRC over those of the base composite. This is attributed to the fact that the effective axial thermal conductivities of the CFF \((K₁^{CFF})\) containing either wavy CNTs being coplanar with the 2–3 planes or straight CNTs are marginally improved which eventually do not enhance the effective values of \(K₁\) of the FFRC over those of the base composite (see Table 1 for details).

Figure 10(b) depicts that the effective transverse thermal conductivities \((K₂)\) of the FFRC are significantly improved over those of the base composite (that is, \(Vₜ = 0\)) irrespective of the planar orientations of wavy CNTs and the value of the CNT wave frequency. When compared with the results of the base composite (that is, \(Vₜ = 0\)), nearly 640% enhancement is occurred in the values of \(K₂\) for the temperature range 250K–400K if CNTs (straight or wavy) are present on the circumferential surfaces of the carbon fibers with minimum \(Vₜ = 0.054\) which is corresponding to the value of \(\omega = 0\) (that is, straight CNTs). Although not presented here, the computed effective transverse thermal conductivities \((K₃)\) of the FFRC are found to match identically with those of \(K₂\) corroborating the fact that the FFRC is the transversely isotropic about the 1-axis. Since the thermal conductivity \((K₃)\) of a CNT along the CNT axis within the range of angles considered is almost linear in the range of 0° to 60°.

Table 1 Effective thermal conductivity coefficients of the PMNC, the CFF, and the FFRC \((Rₜ = 0)\). The thermal conductivity coefficient of the \(i\)th phase \((W/mK)\), \(K_i\) and \(K_i'\) are the transverse thermal conductivity coefficients of the \(i\)th phase \((W/mK)\) and “a” indicates data for the base composite (that is, without CNTs)

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>nth phase</th>
<th>(V_{CNT} = 0°)</th>
<th>(\omega = 0) ((V_{CNT} = 0.054))</th>
<th>(\omega = 18\pi/Lₐ) ((1–3 planes, V_{CNT} = 0.165))</th>
<th>(\omega = 18\pi/Lₐ) ((2–3 planes, V_{CNT} = 0.165))</th>
</tr>
</thead>
<tbody>
<tr>
<td>300 K PMNC</td>
<td>—</td>
<td>—</td>
<td>2.077 × 1₀⁵</td>
<td>293.115</td>
<td>0.433</td>
</tr>
<tr>
<td>300 K CFF</td>
<td>—</td>
<td>—</td>
<td>334.936</td>
<td>520.892</td>
<td>335.019</td>
</tr>
<tr>
<td>300 K FFRC</td>
<td>—</td>
<td>—</td>
<td>303.755</td>
<td>435.834</td>
<td>303.772</td>
</tr>
<tr>
<td>350 K PMNC</td>
<td>—</td>
<td>—</td>
<td>2.077 × 1₀⁵</td>
<td>293.115</td>
<td>0.433</td>
</tr>
<tr>
<td>350 K CFF</td>
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<td>—</td>
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<td>520.892</td>
<td>335.019</td>
</tr>
<tr>
<td>350 K FFRC</td>
<td>—</td>
<td>—</td>
<td>303.755</td>
<td>435.834</td>
<td>303.772</td>
</tr>
<tr>
<td>400 K PMNC</td>
<td>—</td>
<td>—</td>
<td>2.077 × 1₀⁵</td>
<td>293.115</td>
<td>0.433</td>
</tr>
<tr>
<td>400 K CFF</td>
<td>—</td>
<td>—</td>
<td>334.936</td>
<td>520.892</td>
<td>335.019</td>
</tr>
<tr>
<td>400 K FFRC</td>
<td>—</td>
<td>—</td>
<td>303.755</td>
<td>435.834</td>
<td>303.772</td>
</tr>
</tbody>
</table>

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thermal conductivities of the PMNC and the FFRC with and without considering the CNT/polymer interfacial thermal resistance. $K_i$ is the axial thermal conductivity coefficient of the $i$th phase (W/mK), and $K_{i'}$ are the transverse thermal conductivity coefficients of the $i$th phase (W/mK).

![Image](http://heattransfer.asmedigitalcollection.asme.org/ on 11/07/2014 Terms of Use: http://asme.org/terms)

Table 2 Comparison of the effective thermal conductivity coefficients of the PMNC and the FFRC with and without considering the CNT/polymer interfacial thermal resistance. $K_i$ is the axial thermal conductivity coefficient of the $i$th phase (W/mK), and $K_{i'}$ are the transverse thermal conductivity coefficients of the $i$th phase (W/mK).

<table>
<thead>
<tr>
<th>$T$ (K)</th>
<th>$R_h$ = 0 (V$_{CNT}$ = 0.054)</th>
<th>$R_h = 20 \times 10^{-8}$ m$^2$K/W</th>
<th>$R_h$ = 0 (V$_{CNT}$ = 0.165)</th>
<th>$R_h = 20 \times 10^{-8}$ m$^2$K/W</th>
</tr>
</thead>
<tbody>
<tr>
<td>300 K</td>
<td>PMNC 0.247 434.446 0.158 434.397</td>
<td>2.077 $\times 10^3$ 293.115</td>
<td>2.077 $\times 10^3$ 293.089</td>
<td></td>
</tr>
<tr>
<td>350 K</td>
<td>FFRC 303.772 4.066 303.736 4.066</td>
<td>1.149 $\times 10^3$ 4.059</td>
<td>1.149 $\times 10^3$ 4.059</td>
<td></td>
</tr>
<tr>
<td>400 K</td>
<td>PMNC 0.251 278.522 0.160 278.474</td>
<td>1.331 $\times 10^4$ 188.01 4.059</td>
<td>1.331 $\times 10^4$ 187.699</td>
<td></td>
</tr>
<tr>
<td></td>
<td>FFRC 333.164 4.112 333.128 4.112</td>
<td>874.659 4.102</td>
<td>874.627 4.102</td>
<td></td>
</tr>
<tr>
<td></td>
<td>PMNC 0.247 203.809 0.158 203.762</td>
<td>973.770 137.642</td>
<td>973.695 137.337</td>
<td></td>
</tr>
</tbody>
</table>

temperature variation considered here is significantly larger than that of the polymer matrix and CNTs are radially grown on the carbon fiber, the effective value of $K_2$ of the FFRC is improved. Also, for a particular diameter of the CFF, the projected length of the radially grown CNTs along the radial direction remains constant irrespective of the wave frequency. Hence, increase in the effective value of $K_2$ does not depend on the CNT wave frequency and the planer orientations of wavy CNTs. Since the CNT waviness being coplanar with the 1–3 planes significantly improve the axial and the transverse thermal conductivities of the FFRC over those of the FFRC containing wavy CNTs being coplanar with the 2–3 planes, the subsequent results are presented in case of the FFRC containing either straight CNTs or wavy CNTs being coplanar with the 1–3 planes.

In the previous set of results, the effective thermal conductivities of the FFRC have been estimated considering the perfect CNT/polymer matrix interface ($R_h = 0$). However, the CNT/polymer matrix interfacial thermal resistance may affect the heat transfer characteristics of the FFRC. Researchers reported that the magnitude of $R_h$ between nanoparticles/CNTs and different matrices ranges from $0.77 \times 10^{-8}$ m$^2$K/W to $20 \times 10^{-8}$ m$^2$K/W [7,40]. To analyze the effect of the CNT/polymer matrix interfacial thermal resistance on the effective thermal conductivities of the FFRC, the EM approach incorporating such CNT/polymer matrix interfacial thermal resistance has been utilized and the value of $R_h$ is taken as $20 \times 10^{-8}$ m$^2$K/W. Table 2 summarizes the effective thermal conductivities of the PMNC and the FFRC with and without considering the CNT/polymer interfacial thermal resistance for the particular values of $T$. This Table demonstrates that the CNT/polymer interfacial thermal resistance does not affect the effective values of $K_3$, $K_2$, and $K_1$ of the FFRC containing either straight or wavy CNTs being coplanar with the 1–3 planes. Table 2 also demonstrates that the CNT/polymer thermal interfacial resistance affects the axial thermal conductivities ($K_1$) of the
PMNC containing straight CNTs. This finding is consistent with the previously reported results [6–8,11–14], where also it is found that the consideration of the CNT/polymer thermal interfacial resistance affects the effective thermal conductivities of aligned CNT-reinforced polymer composites. On the other hand, the CNT/polymer thermal interfacial resistance does not affect the effective values of $K_{eff,PMNC}$, $K_{eff,PMNC}$, and $K_{eff,PMNC}$ of the PMNC containing wavy CNTs being coplanar with the 1–3 planes. It may be recalled that the value of $K_{eff,PMNC}$ containing straight CNTs being coplanar with the 1–3 planes. In this case, the influence of the CNT/polymer thermal interfacial resistance is suppressed by the higher CNT volume fraction in the PMNC.

So far, in this work, the effect of CNT waviness on the thermal conductivities of the FFRC has been studied considering that the diameter of the CFF remains constant for different waviness parameters. This results into the increase in the CNT volume fraction in the FFRC. However, the investigations of the thermal conductivities of the FFRC for the fixed values of the CNT and the carbon fiber volume fractions irrespective of the values of $\omega$ would be an important study. As shown in Fig. 11, a straight CNT in the RVE of the PMNC can be replaced with the CNT wave in such a way that the CNT volume fraction in the FFRC remains same (that is, the running length of the CNT wave ($L_{nr}$) equals to the length of straight CNT ($L_{m}$)). For such investigation, the discrete values of $\omega$ are considered as 0, 6$\pi$/Ln, 10$\pi$/Ln, and 14$\pi$/Ln keeping the constant values of $V_{CNT} = 0.054$ and $V_{f} = 0.5$ in the FFRC containing either straight CNTs or wavy CNTs being coplanar with the 1–3 planes. It may be recalled that the value of $V_{CNT} = 0.054$ corresponds to the CNT volume fraction in the FFRC containing straight CNTs and the corresponding values of the length of straight CNT ($L_{m}$), the diameter of the carbon fiber ($d_f$) and the diameter of the CFF ($d_{CFR}$) are 1.734 $\mu$m, 10 $\mu$m, and 13.47 $\mu$m, respectively. Next, the values of $\omega$, $A$, and $L_{nr}$ of the CNT wave are varied in such a way that the CNT volume fraction in the FFRC remains constant (that is, keeping the value of $L_{nr}$ equals to the value of $L_{m}$). The outcome of these variations is shown in Table 3. Figures 12 and 13 demonstrate the variations of the effective values $K_1$ and $K_2$ of the FFRC with the temperature, respectively, considering the value of $K_1 = 20 \times 10^{-8}$ m$^2$K/W. Figure 12 reveals that the effective value of $K_1$ is significantly enhanced when the value of $\omega$ is 10$\pi$/Ln compared to all other values of $\omega$ considered as above. When compared with the results of the FFRC containing straight CNTs (that is, $\omega = 0$), it is observed that almost 75% and 9% improvements in the values of $K_1$ of the FFRC containing wavy CNTs ($\omega = 10\pi$/Ln and $A = 50d_f$) occur when the values of $T$ are 300 K and 400 K, respectively. These results are significant and reveal that keeping the same CNT and carbon fiber volume fractions in the FFRC, the axial thermal conductivities of the FFRC can be improved by optimizing the values of $\omega$ and $A$. On the other hand, Fig. 13 demonstrates that CNT waviness degrades the transverse thermal conductivities of the FFRC. In this case, straight CNTs being grown on the circumferential surfaces of the carbon fiber provide better estimates for the values of $K_2$ over those of the FFRC containing wavy CNTs. It may also be noted from Fig. 13 that the different values of $T$ do not influence the effective values $K_2$. Although not presented here, the optimum values of $\omega$ and $A$ should be determined for improving the values of $K_2$ of the FFRC containing wavy CNTs being coplanar with 2–3 planes over those of the FFRC containing straight CNTs. However, this involves a wide range of geometrical and material parameters. Therefore, investigations of such parameters for optimizing the effective thermal conductivities of the FFRC are not presented.

<table>
<thead>
<tr>
<th>$\omega$ ($\mu$m$^{-1}$)</th>
<th>$A$ ($\mu$m)</th>
<th>$L_{nr}$ ($\mu$m)</th>
<th>$d_{CFR}$ ($\mu$m)</th>
<th>$\nu_{f}$</th>
<th>$\nu_{f}$</th>
<th>$\nu_{f}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>1.734</td>
<td>13.47</td>
<td>0.1322</td>
<td>0.5513</td>
<td>0.907</td>
</tr>
<tr>
<td>$6\pi$/Ln</td>
<td>75$d_f$</td>
<td>0.480</td>
<td>12.28</td>
<td>0.2116</td>
<td>0.6631</td>
<td>0.827</td>
</tr>
<tr>
<td>$10\pi$/Ln</td>
<td>50$d_f$</td>
<td>0.975</td>
<td>11.95</td>
<td>0.2513</td>
<td>0.7003</td>
<td>0.805</td>
</tr>
<tr>
<td>$14\pi$/Ln</td>
<td>25$d_f$</td>
<td>1.395</td>
<td>12.79</td>
<td>0.1694</td>
<td>0.6113</td>
<td>0.861</td>
</tr>
</tbody>
</table>

Note: $\omega$, $A$, $L_{nr}$, and $L_{nr}$ denote the CNT wave frequency, amplitude of the CNT wave, linear distance between the CNT ends, running length of the CNT wave, respectively; $d_{CFR}$ and $d_{CFR}$ are the diameters of the carbon fiber and the CFF, respectively; $\nu_{f}$ and $\nu_{f}$ represent the CNT volume fractions in the PMNC and the FFRC, respectively; $\nu_{f}$ and $\nu_{f}$ represent the carbon fiber volume fractions in the CFF and the FFRC, respectively; $\nu_{f}$ is the CFF volume fraction in the FFRC.
here and can be considered as the future scope of the present study.

4 Conclusions

In this article, the effect of CNT waviness on the effective thermal conductivities of the FFRC has been studied. The distinctive feature of the construction of this composite is that the amplitudes of sinusoidally wavy CNTs radially grown on the circumferential surfaces of the carbon fibers are either parallel or transverse to the axes of the carbon fibers. Analytical models based on the MOC and the EM approaches are developed to determine the effective thermal conductivities of this novel composite. The following major conclusions are drawn from the work carried out in this paper:

(1) If the amplitudes of sinusoidally wavy CNTs are parallel to the carbon fiber axis and the diameter of the CFF is constant for different waviness parameters then the axial ($K_{ax}$) and the transverse ($K_{tx}$ and $K_{ts}$) thermal conductivities of the FFRC are significantly improved over those of the FFRC containing either straight CNTs or wavy CNTs being coplanar with the transverse plane of the carbon fiber.

(2) The effective values $K_1$ and $K_2$ of the FFRC containing wavy CNTs being coplanar with the carbon fiber axis are improved by 86% and 640%, respectively, over those of the base composite (that is, without CNTs) when the values of $V_{CNT}$ and $T$ are 16.5% and 400 K.

(3) The present study reveals that the CNT waviness parameters, such as the wave frequency and the amplitude of the CNT should be optimized for enhancing the thermal conductivities of the FFRC keeping the CNT and carbon fiber volume fractions in the FFRC constant.

(4) The CNT/polymer matrix interfacial thermal resistance does not affect the overall effective thermal conductivities of the FFRC.

(5) For the constraint on the thickness of the FFRC lamina, wavy CNTs can be used keeping the diameter of the CFF constant for increasing the CFF volume fraction in the FFRC resulting in significant enhancement in the values of the thermal conductivities of the FFRC. Thus this study establishes that the wavy CNTs can be exploited for developing truly multifunctional composites with improved heat transfer performance characteristics for advanced thermal management applications.

Acknowledgment

The authors would like to thank the (anonymous) referees for their valuable comments and suggestions.

Nomenclature

- $a$ = radius of the carbon fiber (m)
- $A$ = amplitude of the CNT wave (m)
- $a_k$ = Kapitza radius (m)
- $b$ = width of the cell (m)
- $b_c$ = width of the subcell (m)
- $d_{CF}$ = diameter of the CFF (m)
- $d_f$ = diameter of the carbon fiber (m)
- $d_{bc}$ = diameter of the CNT (m)
- $h$ = height of the representative unit cell (m)
- $h_{bc}$ = height of the subcell (m)
- $K_i$ = effective thermal conductivities of the FFRC ($W/mK$)
- $K_{cf}$ = thermal conductivity of the carbon fiber ($W/mK$)
- $K^n$ = thermal conductivity of the CNT ($W/mK$)
- $K^p$ = thermal conductivity of the polymer ($W/mK$)
- $K^{CF}$ = effective thermal conductivities of the CFF ($W/mK$)
- $K^{NC}$ = effective thermal conductivities of the unwound PMNC lamina containing straight CNTs ($W/mK$)

Superscripts and Subscripts

- $I$ = length of the subcell (m)
- $L$ = length of the representative unit cell (m)
- $L_{av}$ = straight distance between the two ends of the CNT wave (m)
- $L_{a}$ = length of straight CNT (m)
- $L_{ar}$ = running length of the CNT wave (m)
- $n$ = number of sinusoidally CNT waves
- $q$ = heat flux in the unwound PMNC ($W/m^2$)
- $q_{av}$ = average heat flux in the $\beta$th subcell ($W/m^2$)
- $R$ = radius of the CFF (m)
- $R_{k}$ = interfacial thermal resistance between the CNT and the polymer ($m^2K/W$)
- $T$ = temperature (K)

\[ T_1, T_2 = \text{transformation matrices} \]
\[ V = \text{volume of the representative unit cell (m)}^3 \]
\[ V_{CF} = \text{CF volume fraction in the FFRC} \]
\[ V_{CNT} = \text{CNT volume fraction in the FFRC} \]
\[ V_f = \text{carbon fiber volume fraction in the CFF} \]
\[ V_{nc} = \text{CNT volume fraction in the PMNC} \]
\[ V_p = \text{polymer volume fraction in the PMNC} \]
\[ V_{bc} = \text{volume of the } \beta \text{th subcell (m)}^3 \]
\[ V_{i} = \text{volume of the } i \text{th phase (m)}^3 \]
\[ \lambda_a = \text{angle between the radial axis (3-axis) along which the CNT wave is grown and the 3-axis in the 2–3 planes} \]
\[ \lambda_{bc} = \text{interfacial thermal resistance between the CNT and the polymer} \]
\[ \phi = \text{angle between the CNT axis at any point and the 3 or 3'-axis which is varying over the linear distance between the CNT ends} \]
\[ \omega = \text{wave frequency of the CNT wave (m$^{-1}$)} \]

**References**
