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Contact mechanics and thermal conductance of carbon nanotube array interfaces

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ABSTRACT

A model is developed in this work to predict the thermal contact resistance of carbon nanotube (CNT) array interfaces with CNT arrays synthesized directly on substrate surfaces. An analytical model for contact mechanics is first developed in conjunction with prior data from load–displacement experiments to predict the real contact area established in CNT array interfaces as a function of applied pressure. The contact mechanics model is utilized to develop a detailed thermal model that treats the multitude of individual CNT–substrate contacts as parallel resistors and considers the effects on phonon transport of the confined geometry that exist at such contacts. The influence of CNT array properties, *e.g.* diameter and density, are explicitly incorporated into the thermal model, which agrees well with experimental measurements of thermal resistances as a function of pressure for different types of interfaces. The model reveals that: (1) ballistic thermal resistance dominates at the CNT array interface; (2) the overall performance of CNT array interfaces is most strongly influenced by the thermal resistance at the contacts between free CNT ends and the opposing substrate surface (one-sided interface) or the opposing CNT array (two-sided interface); and (3) dense arrays with high mechanical compliance reduce the thermal contact resistance of CNT array interfaces by increasing the real contact area in the interface.

1. Introduction

Carbon nanotubes (CNTs) are hollow tubular structures that can have single or multiple layers of ideally seamless graphene rolls. With lengths of several micrometers or more, these nanostructures can achieve aspect ratios as high as 10^4 [1]. Because of these unique structural features and strong carbon-to-carbon bonding, CNTs possess many exceptional vibrational, optical, mechanical, and thermal properties [1,2]. One of these attractive properties is an extremely high intrinsic thermal conductivity that is comparable to that of diamond [1–8]. Also, CNT arrays can exhibit high mechanical compliance and resilience [9–11]. Consequently, CNT arrays can be effective in reducing thermal interface resistance, potentially satisfying the increasing power dissipation challenge in microelectronics, and significant efforts have focused on using CNT and carbon nanofiber (CNF) arrays for this purpose [12–26]. While excellent performance of these materials has been observed experimentally, detailed modeling of heat flow through these arrays has not been reported, and the present work seeks to address this need by developing a combined thermo-mechanical model of CNT array thermal interfaces.

Low-to-moderate thermal interface resistance of CNT arrays has been measured by several techniques. Xu and Fisher [14] reported a resistance value of 20 mm^2 K/W at a contact pressure of 450 kPa for a Si-supported CNT array interfaced with Cu and tested with a reference bar method. Tong et al. [21] used an all-optical pump and probe phase sensitive transient thermo-reflectance method to measure a thermal resistance value of 12 mm^2 K/W for a Si–CNT–glass interface, which is very close to the result presented in [15]. Employing a photoacoustic technique, Amama et al. [22] and Cola et al. [23] measured thermal resistance values as low as 8 mm^2 K/W at a pressure 350 kPa for Si–CNT–Ag interfaces. Using the same photoacoustic technique, Cola et al. [20] measured thermal resistances near 4 mm^2 K/W at moderate pressures for an interface consisting of two CNT arrays in contact.

To complement experiments on CNT arrays used to reduce contact resistance, a thermal model is needed to explain and predict the resistance of CNT array interfaces with different properties for engineering applications and to optimize CNT array properties for improved performance. However, heat transfer through CNT array interfaces is very complicated. Using a variety of simplifying assumptions, Xu [27] developed an initial framework for describing heat transfer through CNT array interfaces. Neglecting the contributions of convection and radiation, the total thermal resistance of CNT array interfaces consists of three primary components [27]: (i) the resistances at interfaces of CNTs to their growth substrate, (ii) the resistances at interfaces of free CNT ends to the opposing substrate surface (one-sided interface) or the opposing CNT array

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Nomenclature

A	nominal contact area (m^2)
A_r	real contact area (m^2)
A'_r	real contact area for two CNT arrays in contact (m^2)
a	radius (or size) of contact area (m)
$\bar{a}$	average radius (or size) of contact area (m)
b	radius of cylinders (or CNTs) (m)
$\bar{b}$	average radius of cylinders (or CNTs) (m)
B	effective bulk modulus of a CNT array (Pa)
c_1	adjustable parameter that captures variation in CNT array characteristics
c_2	parameter that determines the magnitude of saturation pressure
$C/$	volumetric lattice specific heat [$J/(m^3K)$]
E_b	bending modulus of an individual CNT (Pa)
E_r	radial compressive modulus of an individual CNT (Pa)
F	force (N)
h	Planck's constant (6.626068×10^{-34} m^2 kg/s)
Kn	Knudsen number (l_{mfp}/a)
k	thermal conductivity [W/mK]
k_B	Boltzmann constant (1.38066×10^{-23} J/K)
l_{mfp}	phonon mean free path (m)
N	number density of CNTs in contact with growth substrate (CNTs/mm^2)
n	number density of CNTs in contact with opposing substrate (CNTs/mm^2)
n'	number density of CNTs in contact with opposing CNT array (CNTs/mm^2)
P	nominal contact pressure (Pa)
P_f	effective pressure on CNT array (Pa)
q''	heat flux (W/m^2)
R	thermal resistance (K/W)
R''	area-normalized thermal resistance (mm^2 K/W)
$\bar{R}''$	area-normalized thermal resistance based on average CNT radius (mm^2 K/W)
R_z	average peak-to-valley height of surface profile (m)
T	temperature (K)
t	thickness (or length) (m)
t'	incompressible thickness of CNT array (m)
t_o	mathematically extrapolated CNT array thickness at zero pressure (m)
U	lattice vibration energy with the Debye model (J/m^3)
v_g	frequency-independent phonon group velocity (m/s)

Greek symbols

Γ	averaged phonon transmissivity
λ_d	dominant phonon wave length (m)
σ_R	CNT array's resistance to compression (Pa)
Φ	volume ratio of carbon nanotubes in an array
Ψ	constriction alleviation factor
υ	Poisson's ratio

Subscripts

Ag	silver
array	CNT array
b	ballistic resistance
c	total resistance at individual CNT contacts
CNT	carbon nanotube or CNT array
Cu	copper
cd	circular disc in half space
cs	constriction
cy	cylinder
eff	effective
free ends	opposing substrate or opposing CNT array interface
GS	growth substrate
i	arbitrary index corresponding to CNTs of different radius within an array
m	mean or average
Ni	nickel
OS	opposing substrate
pl	plane
root	growth substrate interface
Ti	titanium
vdW	van der Waals contact
x	contact width
y	contact length

(two-sided interface), and (iii) the resistances within the CNT array itself (which can, in turn, include intra- and inter-CNT elements).

Molecular dynamics simulations and Boltzmann transport theory have been used for thermal modeling of solid–fluid interfaces [28,29] as well as solid–solid interfaces [30–34]. The sheer size and inherent variabilities of dense, vertically oriented CNT arrays make such approaches too expensive to use for CNT arrays; however, these approaches can provide useful sub-models to the overall interface model. In a temperature range of 10–100 K, Prasher [35] calculated the thermal boundary resistance between an individual multiwalled CNT and a Pt contact with CNTs vertically and horizontally oriented to the Pt surface using analytical approximations for phonon transport. In an equally low temperature range, Prasher et al. [36] calculated the thermal boundary resistance between an individual single-walled CNT and a Si substrate. The complementary challenge of estimating the total thermal resistance – and total contact area – achieved by the multitude of contacts that exists in a CNT array interface is addressed in the present work.

Before the application of a thermal model, the contact mechanics of an interface must be understood. Inspired by observations of CNT deformation at interfaces, a semi-empirical wool fiber compression theory [37,38], which was developed for the textile industry more than 60 years ago, is applied here to describe the deformation of substrate-supported CNT arrays under loading. Employing classical contact mechanics principles and considering the effect of van der Waals forces at the nanoscale, we extend the theory to predict the contact area established in CNT array interfaces based on data from recent load–displacement experiments [39–41]. The CNT array contact mechanics model and detailed constriction and ballistic resistance analysis at individual CNT–substrate contacts are integrated in a thermal resistance model that describes heat transfer across CNT array interfaces. The model includes the effects of CNT array properties, *e.g.* diameter, density, and distribution, with the aim of providing useful information for optimizing CNT array thermal contact resistance.

2. CNT array contact mechanics model

2.1. CNT roots: contact at the growth substrate

Fig. 1 contains schematics of a vertically oriented CNT array directly synthesized on a flat substrate. Fig. 1a and c illustrate the section and plan views of CNT contacts to their growth substrate. To facilitate the thermal resistance analysis presented in later sections, the planar growth substrate is represented as a cluster of cylinders (sometimes called 'flux tubes') with radius $b_{GS,i}$ [42]. Centered on the axis of each cylinder at the surface is a protruding carbon nanotube with radius $b_{CNT,i}$. The sum of the substrate cylin-