Carbon 114 (2017) 15–22

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

Understanding thermal conductance across multi-wall carbon nanotube contacts: Role of nanotube curvature

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ARTICLE INFO

Article history:
Received 17 August 2016
Received in revised form
26 October 2016
Accepted 21 November 2016
Available online 22 November 2016

Keywords:
Molecular dynamics
MWCNTs
Thermal transport
Interface thermal conductance
Phonon focusing

ABSTRACT

Thermal energy transfer at the interconnects in carbon based nanoelectronic devices plays a crucial role towards their performance as well as their reliability. In this study, we investigate such thermal energy transfer across physically interacting multi-wall carbon nanotubes (MWCNTs) as a function of their diameter, length, number of walls, inter-layer chirality differences, and different angular orientation of the cross-contact. Using non-equilibrium molecular dynamics simulations for phonon energy transfer, we predict that MWCNTs' curvature and their number of walls emerge as two critical factors, with each of them determining the limiting value of the thermal conductance across MWCNT contacts in different diameter regimes. For thinner MWCNTs, the curvature determines the limiting value of the conductance and leads to an interesting non-monotonic character, while the number of walls dominates the contact conductance for large diameter MWCNTs. We discuss their respective origins and distinguish their governing regimes using several arguments —focusing of phonons, and confinement of the phonon focusing cone, large mean free path of graphite— and how they modulate radial thermal transport, leading to observed trends of thermal conductance across MWCNT contacts.

1. Introduction

Interfaces play a pertinent role in thermal transport related structure-property relationships in many different areas of scientific interests, including composites and electronics reliability. As the system dimensions reduce to sub-micron/nanometer length scales, their role becomes even more critical due to increase in surface-area to volume ratio. Within the specialized field of nanoelectronics, fabrication of carbon nanotube (CNT) based devices have attained consistent attention [1], partly due to their excellent electrical and thermal transport properties, which can be further tuned through chemical and structural modifications [2–5]. For example, metallic CNTs (especially multi-wall carbon nanotubes (MWCNTs)) have been used as percolation constituents in flexible traces, conductive solders and nanocomposites for desired electronic transport [6–13]. However, despite a large number of potential nanoelectronic applications, the use of CNT devices has still been limited because of their unknown reliability and variation in performance, thus advocating the necessity of understanding the energy transfer and loss at device interconnects. As electrons pass through and hop across different CNTs within these device components, inevitable heating due to dissipative losses at the interconnects (and/or the physical contacts) emerges as a performance-limiting bottleneck. Of particular interest to this study is the issue of thermal conductance across CNT contacts, which are often experimentally encountered in nanotube based flexible solders, adhesives and polymeric nanocomposite materials tailored for electronics applications [6–14].

To date, most experimental work towards measuring interface

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http://dx.doi.org/10.1016/j.carbon.2016.11.056
0008-6223/

thermal conductance (ITC) across CNT contacts has been done on ensembles of CNTs, where ITC is indirectly estimated as an average over numerous contacts [15–18]. Recently, Yang et al. reported an interesting observation through one of the first direct measurements of ITC between individual MWCNTs of ~42–68 nm diameter [19]. The experimental data indicated that the area-normalized thermal conductance between two MWCNT segments (broken from one long MWCNT) increases approximately linearly with tube diameter. Interestingly, even for the largest MWCNTs, the reported value was still an order of magnitude lower than that of inter-layer thermal conductance in bulk graphite (~18 GWm^{-2} K^{-1}) [3]. This behavior is surprising with common expectation that the normalized ITC values should be independent of tube diameter. Through semi-quantitative analyses, the authors attributed their observation to three coupled mechanisms: phonon reflection at inner tube surface, long phonon mean free path along *c*-axis of graphite, and significant phonon focusing in graphitic materials because of its high thermal anisotropic nature. Despite their numerical analyses were based on simple slit contact model between two semi-infinite graphitic slabs, the study provided valuable physical and intuitive insights into the observed phenomenon [19].

The planar graphitic model, as employed by Yang et al. appears adequate to model thermal exchange between large diameter (>40 nm) MWCNTs, where local curvature effects are marginal. However, for thinner CNTs, where intrinsic curvature can become significant, it is pertinent to ask following questions: a) Does the *flat*-graphitic model of contact thermal conductance still valid for such MWCNTs where their intrinsic curvature starts playing a role?; b) If so or not, how does curvature modulate or alter the aforementioned mechanisms towards governing interface thermal transfer?; c) In addition to diameter, what role does the number of inner walls play in determining overall thermal energy exchange?; d) Do chirality differences between consecutive walls play any role in inter-wall thermal energy exchange?; and e) How does the differences in angular orientation at MWCNT contact affect thermal transport across the contact?

Currently, there exists a disconnection between experimental and simulation efforts from the perspective of addressing questions projected above. While most thermal conductance experiments have been performed on MWCNTs (diameter > 20 nm up to 100s of nm) [15–19] primarily due to experimental challenges, almost all numerical modeling has been performed on single-wall CNTs (SWCNTs) (diameter < 2 nm) because of the limitations associated with computational costs [20–34]. In this study, we aim to bridge this gap by performing comprehensive molecular dynamics (MD) simulations with more realistic models for the first time, i.e., modeling contact thermal conductance between MWCNTs up to 8 nm diameter and up to 10 walls along with modeling their flat graphitic counterparts, i.e., multi-layer graphene nano-ribbons (MLGNRs). The obtained results disclose interesting phenomena beyond what have been reported so far. The new observations, together with the underlying mechanism provide a more complete physical picture of contact thermal conductance between graphitic nanomaterials and help determining the efficiency of energy transport across device interconnects/junctions and thus, dictating their performance.

2. Results and discussion

2.1. Thermal conductance across multi-layer graphene nanoribbons

We first verify the physical insights based on graphene stacks by performing non-equilibrium molecular dynamics (NEMD) simulations with MLGNRs. The modeled system is represented in Fig. 1a with two ~8 nm wide, ~25 nm long, 4-layer GNRs forming a cross contact. The simulation details and further specifics are discussed in the Methods section. We should point out that our simulations are notably different from what has been reported by Yang et al. in that (a) no periodic boundary conditions have been applied so this model truly represents two MLGNRs making a cross contact, unlike the previous study in which parallel, periodic GNR stacks were used; and (b) MLGNRs up to 32 layers (4 times to what is reported in Ref. [19]) were modeled, resulting in the effective thickness up to ~11 nm.

Fig. 2a shows the resultant heat transfer rate and the steady-state temperature distribution (in the inset) across MLGNRs as a function of the number of GNR layers (N_L), while Fig. 2b plots the derived contact thermal conductance as a function of N_L. It is evident from Fig. 2 that both heat transfer rate and contact thermal conductance increase as N_L gets larger, which is consistent with the experimental observation based on MWCNTs and associated numerical analysis [19]. It should be noted that the maximum ribbon thickness (~11 nm) is comparable to the wall thickness values of the MWCNTs used by Yang et al. (~14–26 nm); and as indicated in Fig. 2b, the escalation of the contact thermal conductance with the ribbon thickness does not slow down significantly till 32 layers, suggesting that the observed trend is expected to persist for much thicker GNRs.

It is important to point out that without periodic boundary conditions, our simulation better represents two graphitic nanomaterials making a point contact, while the modeling results in Ref. [19] are actually for two flat plates making a slit contact in a periodic geometry. The consistency between these two approaches, i.e. observation of increasing trend in conductance with N_L, is suggestive of marginal differences associated with applying different boundary conditions. The differences in absolute values of predicted interface thermal conductance (a factor of ~2 with respect to Ref. [19]) are attributed to differences in the modeling geometry, but more importantly to the differences in aspect ratio of modeled MLGNRs (lower aspect ratios were modeled in this study). It has been recently reported that GNR aspect ratio do play an important role towards out-of-plane interface thermal conductance and lead to larger values at higher aspect ratios [35].

2.2. Thermal conductance across multi-wall carbon nanotubes

After discussing and validating the cases where effects of tube curvature can be neglected and inter-tube thermal energy exchange in large MWCNTs can be simplified using their flat counterparts, i.e. flat graphene stacks, we switch our focus to regime of thinner MWCNTs (<10 nm), where curvature is not marginal and should not be neglected or simplified by modeling flat graphene-like cases. In this context, we perform a series of comprehensive MD simulations to calculate interface thermal conductance across MWCNT cross-contacts, where we vary the diameter of MWCNTs from 1.6 to 8 nm, number of concentric walls ranging from 1 to 10, and model MWCNT lengths of 25 and 50 nm. A representative cross-contact geometry of interacting MWCNTs is shown in Fig. 1b. The specific details of simulation setup, their execution and thermal transport methodology are explained in Methods section.

One important issue to be discussed and appreciated is how to determine the contact area between interacting MWCNTs. Since the CNTs are intrinsically cylindrical and yet flexible, the 'effective' interaction area for thermal coupling at the contact between two tubes is not trivial to calculate and several methodologies have been employed in the literature to explore it [21,23,25,28,36]. The