

Reactive Metal Bonding of Carbon Nanotube Arrays for Thermal Interface Applications

Michael T. Barako, Yuan Gao, Yoonjin Won, Amy M. Marconnet,
Mehdi Asheghi, and Kenneth E. Goodson, *Fellow, IEEE*

Abstract—Vertically aligned carbon nanotube (CNT) arrays can offer an attractive combination of high thermal conductance and mechanical compliance for thermal interface applications. These arrays require a reliable, thermally conductive bonding technique to enable integration into devices. This paper examines the use of a reactive metal bonding layer to attach and transfer CNT arrays to metal-coated substrates, and the thermal performance is compared with CNT arrays bonded with indium solder. Infrared microscopy is used to simultaneously measure the intrinsic thermal conductivity of the CNT array and the thermal boundary resistance of both the bonded and growth CNT interfaces over a range of applied compressive stresses. A coarse-grained molecular simulation is used to model the effects of compressive pressure on the CNT array thermal conductivity. Reactive metal bonding reduces the thermal boundary resistance to as low as $27 \text{ mm}^2 \cdot \text{K} \cdot \text{W}^{-1}$, which is more than an order of magnitude less than the nonbonded contact.

Index Terms—Carbon nanotubes (CNTs), infrared (IR) microscopy, reactive metal bonding, thermal interface material (TIM).

I. INTRODUCTION

THERMAL interface materials (TIMs) require a combination of high thermal conductance and mechanical compliance for efficient heat transfer and long operating lifetimes. Vertically aligned carbon nanotube (CNT) arrays have received considerable attention for thermal applications due to the high axial thermal conductivity of individual CNTs, which has been measured to be as high as $100\text{--}3500 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ [1]–[7]. The constituent CNTs are nominally vertically aligned to provide a low resistance

thermal pathway across the array, while the low transverse elastic modulus [8]–[10] enables the TIM to alleviate thermo-mechanical stresses. These stresses often arise in the presence of temperature gradients across materials with dissimilar coefficients of thermal expansion, which can lead to degradation and failure of the interface. Thermal conductivity and mechanical compliance are critical to both the performance and the reliability of TIMs for electronics packaging applications, such as in die attachment to heat spreaders and in thermoelectric generators used in waste heat recovery.

There has been extensive work in the last decade focused on improving the intrinsic thermal conductivity of vertically aligned CNT arrays, which ranges from $\sim 10^{-1}$ to $10^2 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ [7], [11]–[15]. The effective thermal conductivity of CNT arrays is governed by the CNT volume fraction, chirality, length, defects, morphology, and CNT–CNT interactions; these properties may be tuned through the CNT growth conditions and postgrowth treatments [7], [13], [16]–[27]. As the intrinsic thermal conductivity of bulk CNT arrays improves, the boundary resistance between the CNT array and the adjacent surface becomes the limiting resistance to heat transfer [11], [12], [28]–[30]. It is challenging to grow CNT TIMs directly on device surfaces since the high-required growth temperature damages many sensitive electronic components. Using an independent bonding step, a CNT array can be grown using a conventional synthesis technique on a sacrificial substrate and later transferred to the device. This decouples the optimization of the CNT array growth from the development of efficient bonding techniques.

The thermal boundary resistance at the interface between the base of the CNT array and the growth substrate is often significantly different than at the interface between the tips of the CNTs and the substrate to which the array is bonded. Several experimental techniques have been used to measure the thermal properties of CNT arrays, including thermoreflectance [11], [12], photoacoustic [31], [32], and steady state [14], [16], [28], [30], [33], [34] methods. Bonding has been shown to improve the adhesion of the CNTs to the target substrate and to reduce the tip interface resistance by more than an order of magnitude, which is likely due to the increasing number of engaged CNTs that participate in conduction across the interface. Previous bonding techniques, include metallic [12], [30], [32], [35], anodic [28], and thermocompressive [30], [31], [33], [36] bonding.

In this paper, CNT arrays are bonded using a commercially available reactive metal film that can be processed at low

Manuscript received May 20, 2014; revised October 13, 2014; accepted November 7, 2014. Date of publication November 19, 2014; date of current version December 5, 2014. This work was supported in part by the National Science Foundation/U.S. Department of Energy Partnership on Thermoelectric Devices for Vehicle Applications under Grant 1048796, in part by the National Defense Science and Engineering Graduate Fellowship Program, in part by the National Science Foundation Graduate Research Fellowship Program, in part by the Stanford Graduate Fellowship Program, and in part by the Office of Naval Research, Arlington, VA, USA under Grant N00014-09-1-0296-P00004. Recommended for publication by Associate Editor A. Bhattacharya upon evaluation of reviewers' comments.

M. T. Barako, Y. Won, M. Asheghi, and K. E. Goodson are with the Department of Mechanical Engineering, Stanford University, Stanford, CA 94305 USA (e-mail: mbarako@stanford.edu; yoonjin@stanford.edu; masheghi@stanford.edu; goodson@stanford.edu).

Y. Gao was with Stanford University, Stanford, CA 94305 USA. She is now with Oracle America, Inc., Santa Clara, CA 95054 USA (e-mail: yuangao@stanford.edu).

A. M. Marconnet was with Stanford University, Stanford, CA 94305 USA. She is now with the Department of Mechanical Engineering, Purdue University, West Lafayette, IN 47907 USA (e-mail: amarconn@purdue.edu).

Color versions of one or more of the figures in this paper are available online at <http://ieeexplore.ieee.org>.

Digital Object Identifier 10.1109/TCPMT.2014.2369371

temperatures but provides a short pulse of localized high temperature to form the bond. Here, reactive metal bonding is compared to indium solder bonding, which is a standard technique used in electronics packaging applications [37]. Comparative infrared (IR) thermometry is used to simultaneously and independently measure the intrinsic thermal conductivity and surrounding thermal interface resistances for CNT arrays bonded to metal-coated substrates. The thermal properties are measured over a range of applied compressive pressures to investigate the pressure dependence of CNT thermal properties and to verify the stability of the bond.

II. SAMPLE PREPARATION

Vertically aligned multiwall CNT arrays (Nanostructured and Amorphous Materials, Inc.) are first grown by chemical vapor deposition to a thickness of $500\ \mu\text{m}$ on silicon substrates. The CNT number density is 10^{10} – 10^{11} CNT/cm², and the individual tubes have an inner diameter of 5–8 nm and an outer diameter of 7–10 nm. These arrays are coated with an adhesion layer, diffusion barrier, and active bonding surface (50-nm Cr, 20-nm Ni, and 150-nm Au, respectively) using electron beam evaporation. The metallization of the surfaces facilitates thermally conductive metallic bonding, while preserving the mechanical compliance of the CNT film.

The CNT arrays are bonded to metal-coated substrates using two methods. The first utilizes a nanostructured reactive metal foil (Nanofoil [38]) as a bonding layer between the metal-coated CNT array and the target substrate, as shown in Fig. 1. This commercially available reactive metal foil contains a 20- μm -thick multilayered core comprised of many thin, periodic Al/Ni bilayers that is plated with 10- μm Sn. The reaction is initiated using a pulsed electrical current, which causes the Al/Ni layers to react and exothermically alloy into $\text{Al}_{0.5}\text{Ni}_{0.5}$. This releases heat and welds the Sn to the adjacent Au surface. The reaction proceeds over a few milliseconds, during which the temperature at the bonding interface can reach $1500\ ^\circ\text{C}$ [38]. Despite the high energy density at the point of reaction, the total energy released during bonding is low ($\sim 1\ \text{kJ/g}$), and this prevents an excessive temperature rise at the device layer of the chip. This temperature rise is estimated to be under $100\ ^\circ\text{C}$ using a previously developed analysis for thermoreflectance characterization [17], [39], [40] and modeling this situation as a heat pulse generated at the interface between a silicon substrate and a semiinfinite copper heat sink.

The reactive metal bonding layer is first placed between the metal-coated CNT surface and a fused silica substrate that is coated with the same metals as the CNT array. The thermal boundary resistance is then measured at the interface prior to bonding. Without removing the sample from the measurement apparatus, the reactive metal is ignited after applying $\sim 250\ \text{kPa}$ compressive pressure to ensure good contact between the Au surfaces and the Sn-plating. This *in situ* bonding process minimizes the uncertainty that would be inherently introduced by variations in sample preparation and contact conditions caused by reassembling the TIM in the

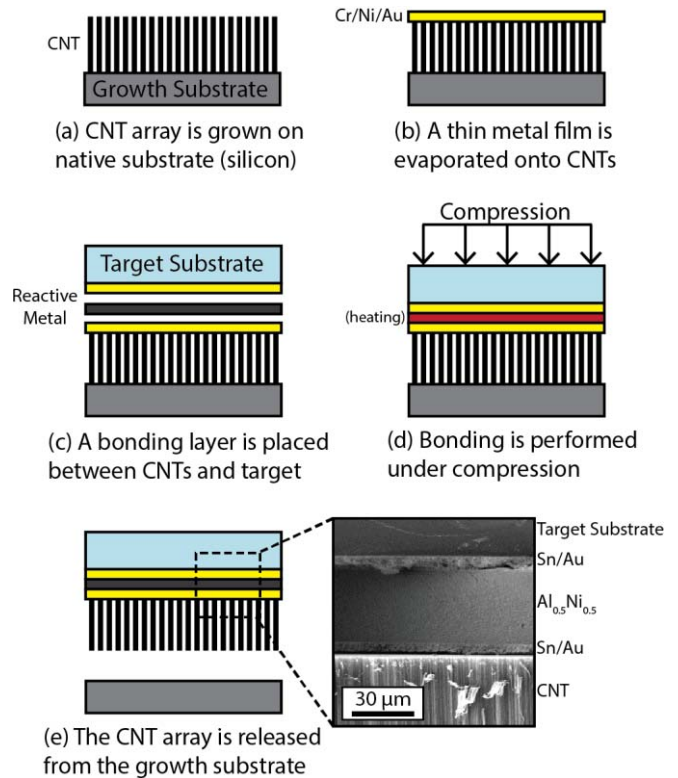


Fig. 1. Schematic of the reactive metal bonding process. (a) Vertically aligned CNT array is grown via chemical vapor deposition onto a silicon substrate. (b) Coated with metal using electron beam evaporation. (c) Bonding layer (reactive metal or indium foil) is placed between the CNT array and a metal-coated target substrate. (d) Bonding is performed under compression. (e) CNT array can then be released from the growth substrate. The SEM shows the substrate-bonding layer-CNT structure after bonding.

experimental setup after bonding [28]. The thermal boundary resistance is measured again after bonding to isolate the effect of bonding on thermal interface resistance.

This technique is promising as a simple, scalable process for bonding CNT-based TIMs and other nanostructured films. Unlike many common eutectic metals (e.g., In, Pb, and Ag), the materials that constitute the reactive metal layer are inexpensive, abundant, and nontoxic. Due to the high temperature of the reaction, reactive metal can also be used for brazing or other processes requiring local high temperatures.

The thermal boundary resistance of a reactive metal bond is compared with a second bonding technique which uses indium solder as the bonding layer. Indium is a common bonding material used in electronics packaging applications, and it adheres well to Au-plated surfaces with the aid of solder flux [37]. In this paper, 25- μm -thick indium foil (Indium Co.) is cleaned with acetone and deionized water and coated with a thin layer of solder flux (Indium Co., Flux 5R). The foil is then placed between the CNT array and a gold-coated fused silica substrate and the entire stack is heated on a hot plate set to $180\ ^\circ\text{C}$. While a thinner layer of indium would further reduce the interface resistance, Tong *et al.* [12] found that a 1- μm -thick layer did not provide complete and uniform contact between the rough CNT surface and the target substrate. In this paper, compressive pressure ($\sim 250\ \text{kPa}$) is applied to ensure good contact and wetting of the indium to the

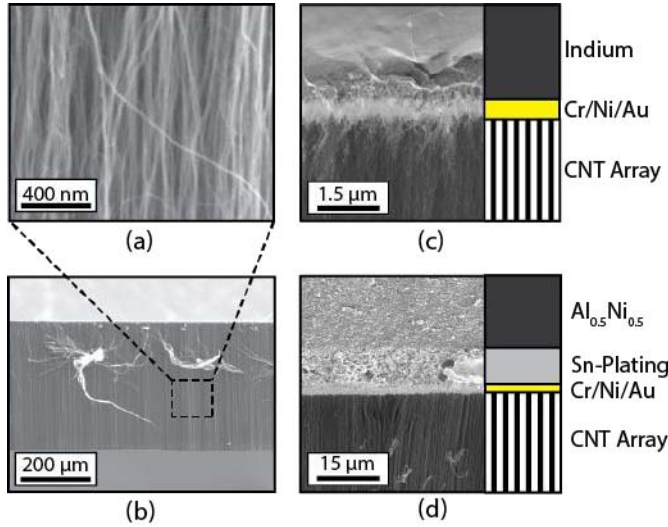


Fig. 2. (a) SEM image of vertically aligned CNTs highlighting the morphology and (b) corresponding CNT array on a silicon growth substrate. (c) Higher magnification image showing the interface of a CNT array bonded using indium foil and (d) reactive metal.

gold surfaces. The scanning electron microscopy (SEM) images of the metal-coated CNT array and the bonded interfaces are shown in Fig. 2.

III. THERMAL CHARACTERIZATION

A. Cross-Sectional Infrared Thermometry

Comparative IR microscopy is used to simultaneously measure the intrinsic thermal conductivity of the CNT array (k_{CNT}) and the thermal interface resistance on each side of the CNT array (R''_{int}). The sample is placed in thermal series between two reference layers having the same cross-sectional area (fused silica with thermal conductivity $k_{\text{ref}} = 1.4 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), and the entire stack is compressed between a heat source and a heat sink (Fig. 3). The surface of the sample stack is spray-coated with a thin layer of graphite (Sprayon Products, LU204) to provide a uniform emissivity close to unity. The emissivity of the IR image is then calibrated by raising the temperature of the sample stack at both ends of the field of view to 70°C as measured by thermocouples. Using this independently measured temperature and a known isothermal field of view, an emissivity map is generated that is used to correlate the measured radiance at each pixel to the temperature. The IR microscope (quantum focus instruments) is then used to generate a high resolution (spatial resolution of $5\text{-}\mu\text{m}/\text{pixel}$), 2-D temperature map (temperature resolution of $\sim 0.1 \text{ K}$) of the surface.

A steady heat flux is then applied to the sample stack that is approximately 1-D. Pressure is measured during the measurement using a load cell (Omega LCMKD-10N) that is affixed in series with the micrometer actuator used to control the compressive strain of the stack. The 2-D temperature field perpendicular to the conduction heat flux vector is reduced to a 1-D temperature profile by averaging the temperature in each cross section (Fig. 3). A least squares linear fit is applied to the temperature profile in each layer to compute

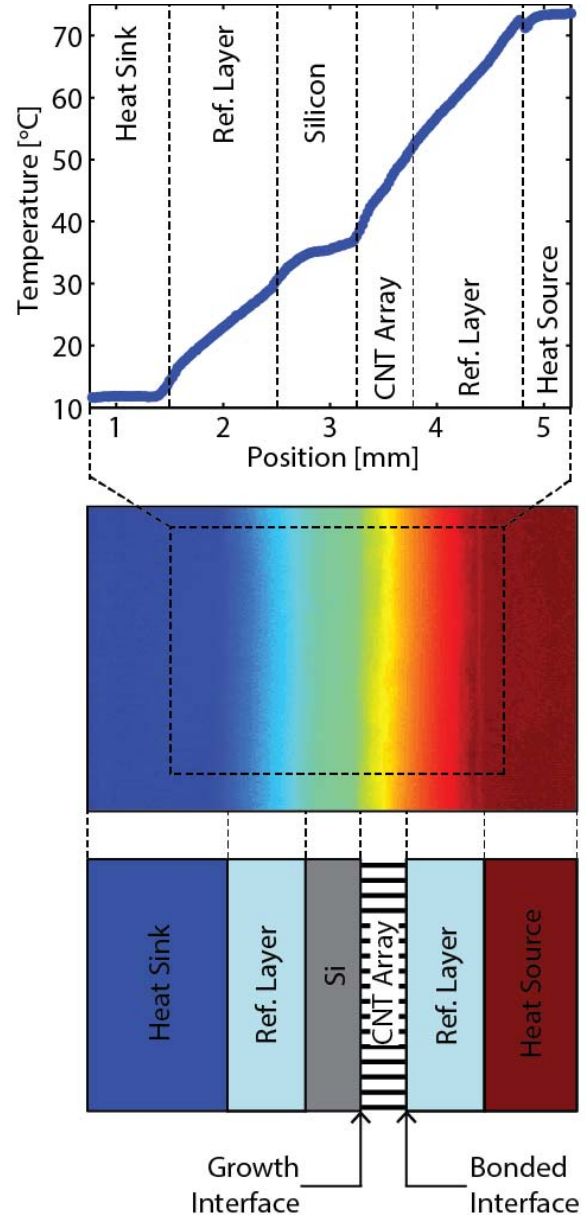


Fig. 3. Schematic, temperature map, and temperature profile used in a comparative IR microscopy measurement. The sample stack is placed between a heat source and a heat sink and the resulting temperature map is imaged along the direction of heat propagation. A 1-D temperature profile is extracted by averaging the temperature in each cross section.

the corresponding temperature gradients. The temperature in each layer is governed by Fourier's law

$$q'' = -k_i \frac{dT_i}{dx} \quad (1)$$

where q'' is the heat flux, k_i is the thermal conductivity, and dT_i/dx is the temperature gradient in the i^{th} layer. The simultaneous measurement of the heat flux through the hot- and cold-side reference layers provides an upper and lower bound, respectively, for the conduction heat flux through the CNT array. The difference between these two heat flux values quantifies the heat dissipated by convection and radiation from the surface of the stack, which is found to be $<10\%$ for all measurements.

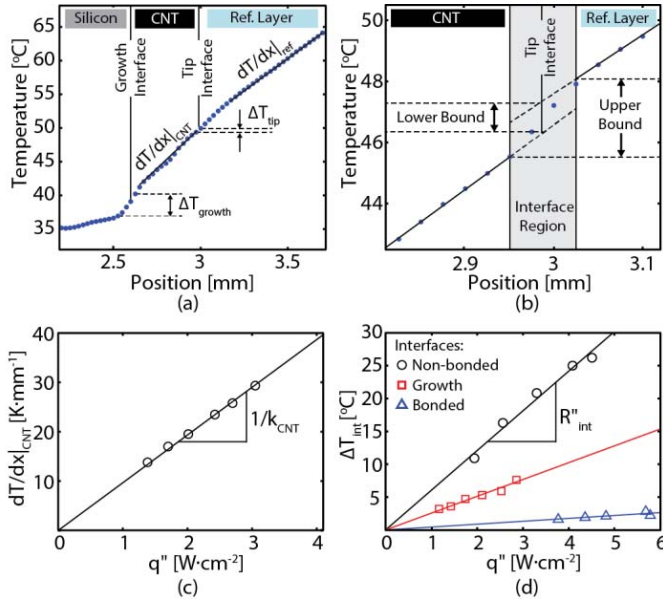


Fig. 4. (a) Extraction of temperature gradients and (b) interfacial temperature drops using the temperature profile measured by IR microscopy. By comparing these measured values to the imposed heat flux, a linear regression is used to extract (c) thermal conductivity and (d) thermal interface resistance.

The thermal conductivity of the CNT array is calculated by equating the heat flux given by Eq. (1) in each layer

$$\frac{k_{\text{CNT}}}{k_{\text{ref}}} = \frac{\left. \frac{dT}{dx} \right|_{\text{ref}}}{\left. \frac{dT}{dx} \right|_{\text{CNT}}} \quad (2)$$

The interfacial temperature drop is measured at both the growth and tip interfaces of the CNT film. This measurement provides a platform for isolating and optimizing the bonding procedure by decoupling the contribution of the thermal boundary resistances R''_{int} from the volumetric resistance of the bulk CNT array. At each interface, R''_{int} is computed using the interfacial temperature drop ΔT_{int} and the heat flux conducting through the stack

$$R''_{\text{int}} = \frac{\Delta T_{\text{int}}}{k_{\text{ref}} \left. \frac{dT}{dx} \right|_{\text{ref}}} \quad (3)$$

The subscript of R''_{int} is used to indicate the nature of the specific interface being measured, such as the interface between the base of the CNTs and the growth substrate (R''_{growth}) and the interface between the tips of the CNTs and the adjacent substrate before (R''_{tip}) and after bonding with indium ($R''_{\text{tip,In}}$) or reactive metal ($R''_{\text{tip,RM}}$).

Interfacial temperature drops are measured by extrapolating the linear fits in adjacent layers to the exact location of the interface. This becomes challenging as R''_{int} becomes small since ΔT_{int} is reduced to a value comparable with the signal noise, which is the case for both indium- and reactive metal-bonded interfaces. As a result, the linear regression in both layers adjacent to the interface can be extrapolated to provide upper and lower bounds for the estimated interfacial temperature difference [Fig. 4(a) and (b)].

The uncertainty in R''_{int} for bonded interfaces, includes appreciable contributions from both the interfacial temperature

difference ΔT_{int} and the heat flux. The uncertainty in ΔT_{int} can be as large as $\pm 0.5^\circ\text{C}$ for the highest heat flux measurements. For interfaces with high thermal resistance (e.g., R''_{growth} , R''_{tip}), the uncertainty in ΔT_{int} is found to be negligibly small compared with the uncertainty in the heat flux. To improve the accuracy of this analysis, data are taken over a range of applied heat flux values. The heat flux is then compared with the temperature gradient in the CNT layer and to ΔT_{int} to calculate k_{CNT} and R''_{int} , respectively, as shown in Fig. 4(c) and (d). Each data point is typically averaged over a minimum of six measurements per pressure (each measurement having a different heat flux). The uncertainty in the measured heat flux is predominately due to losses between the hot- and the cold-side reference layers caused by convection and radiation. Additional uncertainty is introduced by variation in the experimental setup, such as misalignment of the layers, sample variation, and a reduced sensitivity of the IR signal at lower temperatures due to reduced emission.

B. Simulation of Compressed Carbon Nanotube Arrays

Won *et al.* [9] developed a coarse-grained molecular simulation tool to predict the mechanical properties of vertically aligned CNT films using the actual array morphology obtained through image analysis. In this coarse-grained model, each nanotube is represented by a set of nodes connected by many short, elastic nanotube segments, which behave as Euler–Bernoulli beams. The simulation results are consistent with previous experimental measurements of the elastic modulus of CNT arrays using resonators and nanoindentation [8], [10], [41]. In addition to accurately predicting the elastic modulus of CNT films, the simulation also defines the energetically relaxed 3-D morphology of the CNT network. These relaxed structures better describe the experimentally observed CNT array morphologies, which include self-organized bundles and entangled nanotubes.

This geometry is then used to predict thermal transport across the array, including axial CNT conduction as well as CNT–CNT thermal contact resistances. A temperature difference between the top and bottom surfaces (along the axial direction of the CNTs) is imposed to generate a planar heat flux boundary condition. Then, the temperature of each node is calculated for the 3-D network based on the structural information, which assumes a constant thermal conductance of $1.2 \times 10^{-6} \text{ K} \cdot \text{W}^{-1}$ along each segment and a constant thermal conductance of $1.0 \times 10^{-6} \text{ K} \cdot \text{W}^{-1}$ between nodes in contact between adjacent nanotubes. The heat flux and corresponding average surface temperatures are computed, and the thermal conductivity of the CNT network is then calculated using (1).

IV. RESULTS

The thermal properties of the CNT arrays and their adjacent interfaces are first characterized prior to bonding as a baseline for assessing the effectiveness of different bonding techniques. Both k_{CNT} and R''_{tip} vary with compression due to the pressure-dependent degree of CNT engagement at the interface. Two distinct regions are identified in the measured pressure dependence of k_{CNT} . Below $\sim 300 \text{ kPa}$, increasing

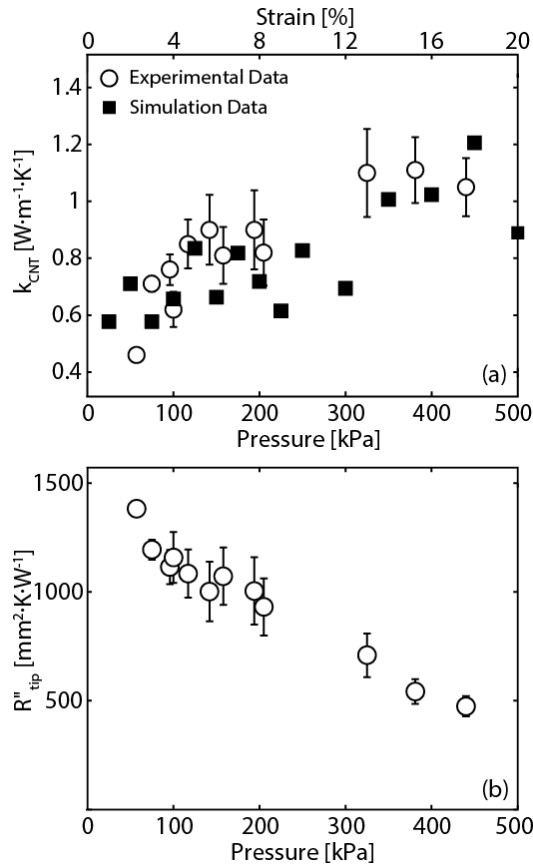


Fig. 5. (a) Thermal conductivity and (b) thermal interface resistance of a nonbonded CNT array in contact with an adjacent metal-coated substrate. Data indicated by circles were obtained experimentally using IR microscopy while data indicated by squares were obtained via the coarse-grained simulation. Each simulation data point is constructed from a compressed CNT unit cell representing a small volume of one possible strained CNT array configuration.

pressure engages more individual CNTs in conduction, which likely causes k_{CNT} to increase and R''_{tip} to decrease. While the surface planarity of the CNT array varies by $<1\%$, only a fraction of the constituent CNTs engage the surface under low compression due to the complex CNT tip morphology that gives rise to a crust layer in the top region of the array [8], [10]. Above ~ 300 kPa, k_{CNT} remains constant, which could correspond to the point when all of the accessible CNTs are engaged. The predicted thermal conductivity values agree well with the experimental results shown in Fig. 5 and exhibit an increasing trend with increasing pressure. In addition, R''_{tip} continues to decrease with pressure as there may be more thermal contact area between the CNTs and the adjacent substrate as the CNTs flatten against the opposing surface [29]. The weak engagement between the nonbonded tips of the CNTs and the adjacent surface creates a large thermal resistance across the interface.

Reactive metal bonding significantly reduces the interfacial thermal resistance and corresponding temperature drop. Fig. 6(a) shows the sharp temperature drop at the CNT tip interface before bonding compared with the temperature drop after reactive metal bonding under constant heat flux conditions. The data recorded prior to bonding includes the unreacted reactive metal layer at the tip interface. Indium bonding

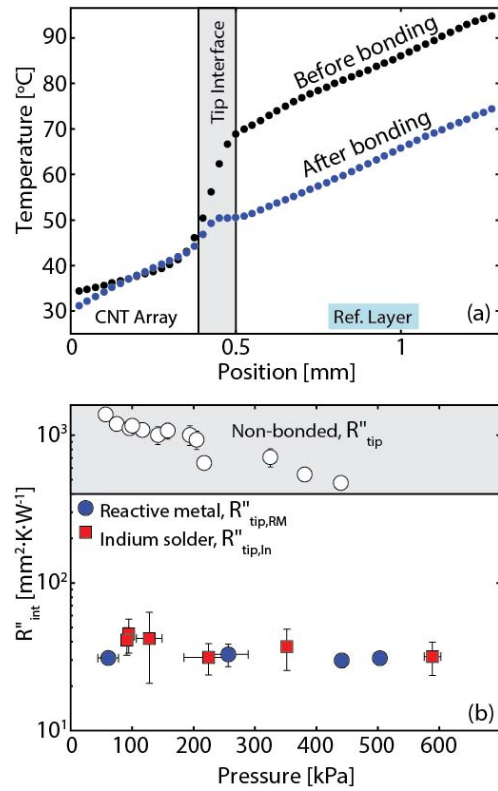


Fig. 6. (a) Comparison of CNT tip interface temperature drop across a reactive metal bonding layer before and after *in situ* bonding. (b) Thermal boundary resistances for reactive metal bonding and indium solder bonding as a function of applied compressive pressure. The pressure independence of the thermal interface resistance is indicative of good mechanical adhesion. Error bars: spread in data across different measurements.

similarly yields more than an order of magnitude reduction in R''_{tip} . Fig. 6(b) shows that while the nonbonded interface exhibits decreasing thermal resistance with pressure, both reactive metal and indium bonding layers exhibit a pressure-independent thermal resistance, which is characteristic of a high-quality metallic bond since the bonding process should inherently engage the maximum number of CNTs. The indium and reactive metal layers are thin and have high thermal conductivity ($k_{In} = 80 W \cdot m^{-1} \cdot K^{-1}$ and $k_{RM} = 40 W \cdot m^{-1} \cdot K^{-1}$ [38]) and do not contribute significantly to the total thermal resistance of the interface. The quality of the bond depends primarily on the degree to which the indium and Sn-plating on the reactive metal wet and adhere to the rough surface of the CNT array and the adjacent substrate. Voids in both the indium and the Sn layers are observed to be present in low concentrations via SEM images after bonding, and this additionally contributes to the measured thermal interface resistance. For the interface bonded with indium, $R''_{tip, In}$ is measured to be $38 \pm 8 mm^2 \cdot K \cdot W^{-1}$, while the interface bonded with reactive metal is measured to be $R''_{tip, RM} = 34 \pm 7 mm^2 \cdot K \cdot W^{-1}$ over the range of applied pressures (Table I). Each bonding method produces a substantially reduced thermal interface resistance compared to both the nonbonded interface and the growth interface.

Both indium bonding and reactive metal bonding provide mechanical adhesion of the CNT array to the bonded substrate.

TABLE I

THERMAL INTERFACE RESISTANCES BETWEEN CNTS AND SUBSTRATES

Symbol	Interface Configuration	R''_{int} [$\text{mm}^2 \cdot \text{K} \cdot \text{W}^{-1}$]
R''_{tip}	Unbonded contact	500-1500
R''_{growth}	Growth interface	130-185
$R''_{\text{tip,In}}$	Indium bonding	30-47
$R''_{\text{tip,RM}}$	Reactive metal bonding	27-41

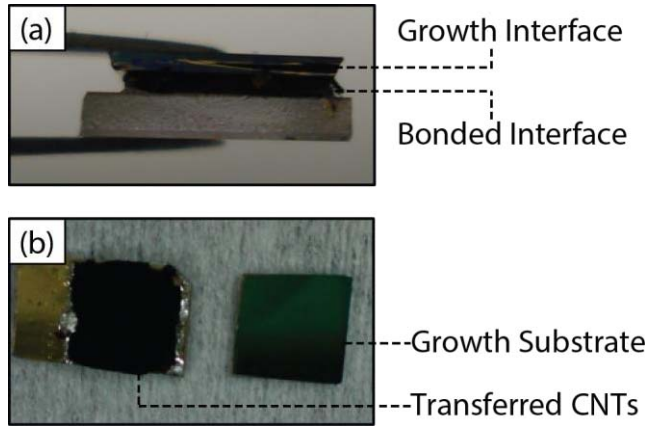


Fig. 7. (a) Side view of a CNT array bonded to a fused silica substrate prior to removing the growth substrate. (b) Top view of the detached CNTs and clean growth silicon substrate after completing the transfer process.

The growth substrate can be released from the bonded CNT array due to the weak adhesion at the growth interface, completing the transfer process, as shown in Fig. 7. The full implementation of a CNT array as a TIM requires a second bonding step, where the newly exposed CNT surface (previously the growth interface) is metal-coated and the bonding process is repeated with a second substrate. The final TIM structure consists of a CNT array with metallic bonds on both sides between two different substrates. These bonds can be reworked similar to conventional eutectic bonding by heating beyond the melting point of the bonding layer and removing the TIM. For the reactive metal bond, the $\text{Al}_{0.5}\text{Ni}_{0.5}$ layer remains solid at high temperatures but can be removed after melting the adjacent Sn layer.

The performance may be improved and the working temperature range expanded by replacing the Sn layer with a higher melting point metal. Bonding processes that do not require solders, such as welding and brazing, could take advantage of the local high temperatures available by reactive metal reactions to provide this extended temperature range. These results demonstrate that metallic bonding with indium or reactive metal can form a thermally conductive bond between a CNT array and a substrate for TIM applications.

V. CONCLUSION

Reactive metal films enable the use of high-temperature bonding processes without exposing sensitive devices to high processing temperatures. In this paper, the reactive metal layer provides the high temperature, localized heat source

to form Sn-Au bonds between metal-coated CNT arrays and substrates. Both indium and reactive metal bonding reduce the interfacial resistance by more than an order of magnitude and the bonded interfaces are independent of the applied pressure after bonding. Subsequent efforts should attempt to utilize the high temperatures provided by reactive metal films to braze and/or weld CNT films to metal-coated substrates, which would significantly extend the operating temperature range of CNT-based TIMs. Additional analysis techniques should be employed to better assess the morphological features of the metallic bond itself, such as the quantity and dispersity of voids present in the bonding layer and the morphological effects of thermal cycling and reworking on both the CNT array and the CNT-metal interface. Further improvements in bond quality may be realized through optimization of the exact bonding conditions, such as flux selection to minimize void formation or bonding in an inert atmosphere to mitigate undesired oxidation effects.

REFERENCES

- [1] E. Pop, D. Mann, Q. Wang, K. Goodson, and H. Dai, "Thermal conductance of an individual single-wall carbon nanotube above room temperature," *Nano Lett.*, vol. 6, no. 1, pp. 96-100, 2006.
- [2] M. T. Pettes and L. Shi, "Thermal and structural characterizations of individual single-, double-, and multi-walled carbon nanotubes," *Adv. Funct. Mater.*, vol. 19, no. 24, pp. 3918-3925, 2009.
- [3] M. Fujii *et al.*, "Measuring the thermal conductivity of a single carbon nanotube," *Phys. Rev. Lett.*, vol. 95, p. 065502, Aug. 2005.
- [4] T.-Y. Choi, D. Poulidakos, J. Tharian, and U. Sennhauser, "Measurement of the thermal conductivity of individual carbon nanotubes by the four-point three-omega method," *Nano Lett.*, vol. 6, no. 8, pp. 1589-1593, 2006.
- [5] P. Kim, L. Shi, A. Majumdar, and P. L. McEuen, "Thermal transport measurements of individual multiwalled nanotubes," *Phys. Rev. Lett.*, vol. 87, p. 215502, Oct. 2001.
- [6] Q. Li, C. Liu, X. Wang, and S. Fan, "Measuring the thermal conductivity of individual carbon nanotubes by the Raman shift method," *Nanotechnology*, vol. 20, no. 14, p. 145702, 2009.
- [7] A. M. Marconnet, M. A. Panzer, and K. E. Goodson, "Thermal conduction phenomena in carbon nanotubes and related nanostructured materials," *Rev. Modern Phys.*, vol. 85, pp. 1296-1327, Aug. 2013.
- [8] Y. Gao, T. Kodama, Y. Won, S. Dogbe, L. Pan, and K. E. Goodson, "Impact of nanotube density and alignment on the elastic modulus near the top and base surfaces of aligned multi-walled carbon nanotube films," *Carbon*, vol. 50, no. 10, pp. 3789-3798, 2012.
- [9] Y. Won *et al.*, "Mechanical characterization of aligned multi-walled carbon nanotube films using microfabricated resonators," *Carbon*, vol. 50, no. 2, pp. 347-355, 2012.
- [10] Y. Won *et al.*, "Zipping, entanglement, and the elastic modulus of aligned single-walled carbon nanotube films," *Proc. Nat. Acad. Sci. USA*, vol. 110, no. 51, pp. 20426-20430, Dec. 2013.
- [11] Y. Gao *et al.*, "Nanostructured interfaces for thermoelectrics," *J. Electron. Mater.*, vol. 39, no. 9, pp. 1456-1462, 2010.
- [12] T. Tong, Y. Zhao, L. Delzeit, A. Kashani, M. Meyyappan, and A. Majumdar, "Dense vertically aligned multiwalled carbon nanotube arrays as thermal interface materials," *IEEE Trans. Compon. Packag. Technol.*, vol. 30, no. 1, pp. 92-100, Mar. 2007.
- [13] X. Wang, Z. Zhong, and J. Xu, "Noncontact thermal characterization of multiwall carbon nanotubes," *J. Appl. Phys.*, vol. 97, no. 6, p. 064302, 2005.
- [14] K. Zhang, Y. Chai, M. M. F. Yuen, D. G. W. Xiao, and P. C. H. Chan, "Carbon nanotube thermal interface material for high-brightness light-emitting-diode cooling," *Nanotechnology*, vol. 19, no. 21, p. 215706, 2008.
- [15] Y. Gao, A. M. Marconnet, R. Xiang, S. Maruyama, and K. E. Goodson, "Heat capacity, thermal conductivity, and interface resistance extraction for single-walled carbon nanotube films using frequency-domain thermoreflectance," *IEEE Trans. Compon., Packag., Manuf. Technol.*, vol. 3, no. 9, pp. 1524-1532, Sep. 2013.

- [16] A. M. Marconnet, N. Yamamoto, M. A. Panzer, B. L. Wardle, and K. E. Goodson, "Thermal conduction in aligned carbon nanotube-polymer nanocomposites with high packing density," *ACS Nano*, vol. 5, no. 6, pp. 4818–4825, 2011.
- [17] M. A. Panzer *et al.*, "Thermal properties of metal-coated vertically-aligned single-wall nanotube arrays," *ASME J. Heat Transf.*, vol. 130, no. 5, p. 052401, 2008.
- [18] H. Cui *et al.*, "Growth behavior of carbon nanotubes on multilayered metal catalyst film in chemical vapor deposition," *Chem. Phys. Lett.*, vol. 374, nos. 3–4, pp. 222–228, 2003.
- [19] S. Maruyama, R. Kojima, Y. Miyauchi, S. Chiashi, and K. Masamichi, "Low-temperature synthesis of high-purity single-walled carbon nanotubes from alcohol," *Chem. Phys. Lett.*, vol. 360, nos. 3–4, pp. 229–234, Jul. 2002.
- [20] E. Einarsson, Y. Murakami, M. Kadowaki, and S. Maruyama, "Growth dynamics of vertically aligned single-walled carbon nanotubes from *in situ* measurements," *Carbon*, vol. 46, no. 6, pp. 923–930, 2008.
- [21] Y. Li *et al.*, "Preferential growth of semiconducting single-walled carbon nanotubes by a plasma enhanced CVD method," *Nano Lett.*, vol. 4, no. 2, pp. 317–321, 2004.
- [22] H. Zhu, K. Suenaga, J. Wei, K. Wang, and D. Wu, "A strategy to control the chirality of single-walled carbon nanotubes," *J. Crystal Growth*, vol. 310, no. 24, pp. 5473–5476, 2008.
- [23] A. J. Hart and A. H. Slocum, "Rapid growth and flow-mediated nucleation of millimeter-scale aligned carbon nanotube structures from a thin-film catalyst," *J. Phys. Chem. B*, vol. 110, no. 16, pp. 8250–8257, 2006.
- [24] Y. C. Choi *et al.*, "Controlling the diameter, growth rate, and density of vertically aligned carbon nanotubes synthesized by microwave plasma-enhanced chemical vapor deposition," *Appl. Phys. Lett.*, vol. 76, no. 17, p. 2367, 2000.
- [25] G. Zhang *et al.*, "Ultra-high-yield growth of vertical single-walled carbon nanotubes: Hidden roles of hydrogen and oxygen," *Proc. Nat. Acad. Sci. USA*, vol. 102, no. 45, pp. 16141–16145, 2005.
- [26] P. M. Parthangal, R. E. Cavicchi, and M. R. Zachariah, "A generic process of growing aligned carbon nanotube arrays on metals and metal alloys," *Nanotechnology*, vol. 18, no. 18, p. 185605, 2007.
- [27] M. Chhowalla *et al.*, "Growth process conditions of vertically aligned carbon nanotubes using plasma enhanced chemical vapor deposition," *J. Appl. Phys.*, vol. 90, no. 10, p. 5308, 2001.
- [28] S. V. Aradhya, S. V. Garimella, and T. S. Fisher, "Electrothermally bonded carbon nanotube interfaces," in *Proc. ITherm*, pp. 1071–1077, 28–31 May 2008.
- [29] B. A. Cola, J. Xu, and T. S. Fisher, "Contact mechanics and thermal conductance of carbon nanotube array interfaces," *Int. J. Heat Mass Transf.*, vol. 52, nos. 15–16, pp. 3490–3503, 2009.
- [30] J. R. Wasniewski *et al.*, "Characterization of metallically bonded carbon nanotube-based thermal interface materials using a high accuracy 1D steady-state technique," *J. Electron. Packag.*, vol. 134, no. 2, p. 020901, 2012.
- [31] R. Cross, B. A. Cola, T. Fisher, X. Xu, K. Gall, and S. Graham, "A metallization and bonding approach for high performance carbon nanotube thermal interface materials," *Nanotechnology*, vol. 21, no. 44, p. 445705, 2010.
- [32] S. L. Hodson, T. Bhuvana, B. A. Cola, X. Xu, G. U. Kulkarni, and T. S. Fisher, "Palladium thiolate bonding of carbon nanotube thermal interfaces," *J. Electron. Packag.*, vol. 133, no. 2, p. 020907, 2011.
- [33] A. Hamdan *et al.*, "Evaluation of a thermal interface material fabricated using thermocompression bonding of carbon nanotube turf," *Nanotechnology*, vol. 21, no. 1, p. 015702, 2010.
- [34] J. Xu and T. S. Fisher, "Enhanced thermal contact conductance using carbon nanotube arrays," in *Proc. 9th ITherm*, Jun. 2004, pp. 549–555.
- [35] X. Hu, L. S. Pan, G. Gu, and K. E. Goodson, "Superior thermal interfaces made by metallically anchored carbon nanotube arrays," in *Proc. InterPACK*, 2009, pp. 597–603.
- [36] R. D. Johnson *et al.*, "Thermocompression bonding of vertically aligned carbon nanotube turfs to metalized substrates," *Nanotechnology*, vol. 20, no. 6, p. 065703, 2009.
- [37] S. S. Too, M. Touzelbaev, M. Khan, R. Master, J. Diep, and K.-H. Keok, "Indium thermal interface material development for microprocessors," in *Proc. 25th Annu. IEEE SEMI-THERM*, Mar. 2009, pp. 186–192.
- [38] *NanoFoil Made by Indium Corporation* [Online]. Available: <http://www.indium.com/nanofoil/>, accessed Mar. 24, 2012.
- [39] A. Feldman, "Algorithm for solutions of the thermal diffusion equation in a stratified medium with a modulated heating source," *High Temperatures-High Pressures*, vol. 31, no. 3, pp. 293–298, 1999.
- [40] D. G. Cahill, "Analysis of heat flow in layered structures for time-domain thermoreflectance," *Rev. Sci. Instrum.*, vol. 75, no. 12, p. 5119, 2004.
- [41] Y. Won, Y. Gao, R. G. de Villoria, B. L. Wardle, T. W. Kenny, and K. E. Goodson, "Crust removal and effective modulus of aligned multi-walled carbon nanotube films," in *Proc. 13th IEEE ITherm*, May/Jun. 2012, pp. 1070–1076.



Michael T. Barako received the B.S. degree in mechanical engineering from Carnegie Mellon University, Pittsburgh, PA, USA, in 2010, and the M.S. degree in mechanical engineering from Stanford University, Stanford, CA, USA, in 2012, where he is currently pursuing the Ph.D. degree with the NanoHeat Laboratory.

He was an Undergraduate Research Assistant with the Nanoscale Transport Phenomena Laboratory, Carnegie Mellon University.

Mr. Barako was a recipient of the National Defense Science and Engineering Graduate Fellowship.



Yuan Gao received the B.S. degree in mechanical engineering from Columbia University, New York, NY, USA, in 2007, and the M.S. and Ph.D. degrees in mechanical engineering from Stanford University, Stanford, CA, USA, in 2009 and 2012, respectively.

She was a Research Assistant with the NanoHeat Laboratory, Stanford University, from 2007 to 2012. She has interned at Google, Inc., Mountain View, CA, USA, and InSynergy Engineering, Inc., Honolulu, HI, USA. She is currently a Hardware Engineer with Oracle America, Inc., Santa Clara, CA, USA.

Her current research interests include thermal and mechanical properties of vertically aligned carbon nanotube films.

Dr. Gao was a recipient of the National Science Foundation Graduate Research Fellowship.



Yoonjin Won received the B.S. degree in mechanical engineering from Seoul National University, Seoul, Korea, in 2005, and the M.S. and Ph.D. degrees in mechanical engineering from Stanford University, Stanford, CA, USA, in 2007 and 2011, respectively.

She is currently a Post-Doctoral Researcher with the NanoHeat Laboratory, Stanford University. Her current research interests include thermal and mechanical properties of nanostructured materials for energy conversion and electronics cooling.



Amy M. Marconnet received the B.S. degree in mechanical engineering from the University of Wisconsin—Madison, Madison, WI, USA, in 2007, and the M.S. and Ph.D. degrees in mechanical engineering from Stanford University, Stanford, CA, USA, in 2009 and 2012, respectively.

She was a Graduate Research Assistant with the NanoHeat Laboratory, Stanford University, an Undergraduate Research Assistant with Prof. J. Booske with the University of Wisconsin—Madison, and an Intern and a Consultant with

the Electron Devices Division, L-3 Communications, San Carlos, CA, USA. She was a Post-Doctoral Associate with the NanoEngineering Group, Massachusetts Institute of Technology, Cambridge, MA, USA, from 2012 to 2013. Since 2013, she has been an Assistant Professor with the School of Mechanical Engineering, Purdue University, West Lafayette, IN, USA. Her current research interests include micro and nanoscale thermal transport and energy conversion.

Prof. Marconnet is a member of the American Society of Mechanical Engineers, the Materials Research Society, and the Tau Beta Pi Association. She was a recipient of the National Science Foundation's Graduate Research Fellowship and the Stanford Graduate Fellowship.



Mehdi Asheghi received the B.S. degree in mechanical engineering from the Sharif University of Technology, Tehran, Iran, in 1991, the M.S. degree from California State University, Long Beach, CA, USA, in 1993, and the Ph.D. degree in mechanical engineering from Stanford University, Stanford, CA, USA, in 1999.

He is currently a Consulting Associate Professor of Mechanical Engineering with Stanford University. He is focusing on the development of nanoengineered embedded convective and passive two-phase (vapor chamber) cooling solutions, thermoelectric energy harvesting at room/high temperatures, and advanced thermal phase change energy storage. In addition, he is involved in thermal engineering of high electron mobility transistors on SiC/diamond substrates, phase-change random access memory, and silicon nanoporous structures. He led a well-known and funded research program at Carnegie Mellon University, Pittsburgh, PA, USA, from 2000 to 2006, that focused on nanoscale thermal phenomena in semiconductor and data storage devices. He has authored over 150 book chapters, journal publications, and fully reviewed conference papers.

Dr. Asheghi has served as the General and Program Chair of the 2012 and 2014 IEEE Intersociety Conference on Thermal and Thermomechanical Phenomena in Electronic Systems and the Program Chair of the 2015 American Society of Mechanical Engineers InterPACK Conference.



Kenneth E. Goodson (F'13) received the B.S. and Ph.D. degrees in mechanical engineering from the Massachusetts Institute of Technology (MIT), Cambridge, MA, USA, in 1989 and 1993, respectively.

He was with the Materials Group, DaimlerBenz, Ulm, Germany. He co-founded Cooligy Inc., Mountain View, CA, USA, which developed heat sinks for workstations and was acquired by Emerson, St. Louis, MO, USA, in 2006. He is currently the Robert Bosch Chairman of the Department of Mechanical Engineering with Stanford University, Stanford, CA, USA, where he currently holds the Davies Family Provostial Professorship. He is also a Heat Transfer Specialist with interests ranging from electronics thermal management to vehicle waste heat recovery. His laboratory works closely with a broad variety of industrial collaborators and brings fundamental science to applications in heat management and energy conversion. His laboratory has highly-cited papers on thermal interfaces, microfluidic heat sinks, electronic packaging thermal and mechanical metrology, phase change memory, and diamond heat spreaders. His 38 Ph.D. alumni include 14 Professors at Stanford University, MIT, the University of California at Berkeley, Berkeley, CA, USA, and other schools.

Prof. Goodson is a fellow of the American Society of Mechanical Engineers and the American Association for the Advancement of Science. He received the Kraus Medal and the Thermi Award and delivered plenary lectures at the IEEE Intersociety Conference on Thermal and Thermomechanical Phenomena in Electronic Systems, InterPack, ThermoMinic, SemiTherm, and PHONONS.