# EPOXY-FILLED MULTIWALL CARBON NANOTUBE ARRAYS FOR THERMAL INTERFACE APPLICATION

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## Introduction

Advances in technology and further miniaturization of microelectronics have led to serious thermal management issues. Many electronic components are comprised of multilayer systems, creating a need for thermal interface materials (TIMs) for thermal management. Decreasing the interfacial thermal contact resistance can drastically increase the thermal performance of the component [1-3]. Multiwall carbon nanotube (MWCNT) arrays possess superior thermal conductivity, along the direction of the MWCNT axes. Reported values for a MWCNT are shown to be as high as 3000 W/mK [5]. In this work, we have thermally characterized sandwich-bonded aluminum discs utilizing adhesive-infiltrated MWCNT arrays. We have observed interfacial thermal contact resistances down to 20 mm$^2$K/W using a laser flash apparatus (Netzsch LFA 427).

## Experimental

The MWCNTs were grown by chemical vapor deposition (CVD) on a quartz substrate to produce continuous arrays of uniform thickness and length. Utilizing a vacuum bag technique, a thin film epoxy, designated SR-4-3, was infiltrated into the MWCNT arrays. The harvested arrays were free standing, completely unattached to the quartz substrate. Packing of the MWCNT arrays was 15-18% close-packing with areal densities of MWCNTs between 2-3.5 mg/cm$^2$ (0.3-0.35 g/cc bulk densities).

LFA tests of epoxy-infiltrated MWCNT arrays, designated AIA-number, were performed to demonstrate the thermal performance of the material under conditions in an actual interface. Aluminum 6061 discs of 12.7 mm (0.5 inch) diameter and 3.12 mm (0.125 inch) thickness were machined from sheets with a mirror polished and mill finish side ($r_A$=0.642 µm). To replicate an actual interface, mill finish surfaces were bonded with the epoxy-infiltrated MWCNT array TIM under a constant pressure of 75 psi at 150 °C for 2 hrs.

The multilayer systems that were compared to the array sandwiches include aluminum substrate material joined with no adhesive (Al-rough), SR-4-3 epoxy film (baseline), commercial adhesives of Ther-O-Bond 1500 (TOB 1500), TOB 1600, TOB 4949, Hysol EA 9396, T412DST (double-sided electrically conductive tape), and silver-filled epoxy resin.

## Results and Discussion

Each of the interface materials were tested for interfacial thermal contact resistance ($R_c$), effective thermal conductivity ($k_{eff}$), and overall thermal diffusivity ($\alpha$) of the sandwich (as if a solid). The thermal diffusivity with respect to contact resistance is plotted on a log-log scale in Figure 1.

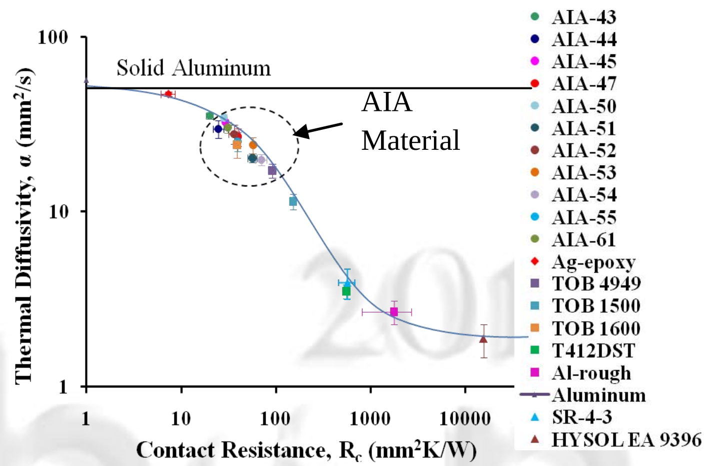


**Fig. 1** Thermal properties of interface materials bonding mill-finish Al substrates.

Thermal diffusivity is just one of the relevant properties when considering materials for possible interface applications. It is useful to find the overall thermal diffusivity of the assembly to compare it against the thermal diffusivity of the substrate material. Multilayer systems that diffuse heat as effectively as solid aluminum are desired for a composite assembly of two aluminum substrates bonded by a TIM.

As shown in Figure 1, the two aluminum substrates, stacked on top of each other without a TIM (Al-rough) showed a very low thermal diffusivity value (2.682 mm$^2$/s). The rough surfaces of the aluminum create voids in the contacting surfaces, causing poor contact. The reduction in contact area results in a reduced rate of heat conduction through the thickness [5]. Bonding the Al substrates with a commercial epoxy adhesive (HYSOL EA 9396) resulted in even poorer thermal performance. Here the polymer adhesive essentially functioned as a thermal insulating barrier for conduction. The introduction of a thermal interface material resulted in significantly higher diffusivity values. The MWCNT arrays outperformed the commercial thermal adhesives, with the exception of TOB 1600, which was comparable, and the silver-filled epoxy interface with the highest overall thermal diffusivity values of an assembly. Specimens bonded with SR-4-3, HYSOL EA 9396 epoxy resin and the T412DST (double-sided electrically conductive tape) had the lowest thermal diffusivity across the assembly.

The overall diffusivity of the assemblies versus effective thermal contact resistance in Figure 1 formed a clear S-curve, on the log-log scale, within the results. This suggests that, for moderate contact resistances, there exists a linear relationship between log contact resistance and log diffusivity (i.e. temperature half-rise time measured by the LFA). It is intuitive that the maximum diffusivity value is the solid aluminum sample. The MWCNT array TIM approached this value following the outline of the curve. Also, on the low end, the epoxy resins exhibited lower diffusivity values and higher contact resistance values. The curve clearly approached this minimum value (as shown in Figure 1).

Diffusivity is a useful thermal property to describe the multilayer systems; however the most important property to define the ability of a material to serve as a thermal interface material is interfacial thermal contact resistance, $R_c$. Thermal contact resistance is the ratio of the virtual temperature jump, to the heat flux, across the interface ($R_c=\Delta T/\vec{q}$). Therefore, the goal was to reduce the interfacial contact resistance to increase the heat flux. The lowest contact resistance was found through the interface composed of silver-filled epoxy and the epoxy-infiltrated MWCNT arrays. For the arrays, the lowest interfacial thermal contact resistance observed for the epoxy-infiltrated CNT arrays was down to 20 $mm^2K/W$ for the AIA 43 array. This suggests the epoxy-infiltrated MWCNT array could function as a TIM in many applications, especially where light weight adhesives are preferred.

Effective thermal conductivity ($k_{eff}$), an intrinsic property of the TIM, is independent of interface parameters. It essentially normalizes the thermal performance of the interface to the bondline thickness ($t_i$) as $k_{eff}= t_i/R_c$ [6]. To compare some CNT arrays to each other as well as to other TIMs, effective thermal conductivity values are seen in Figure 2.

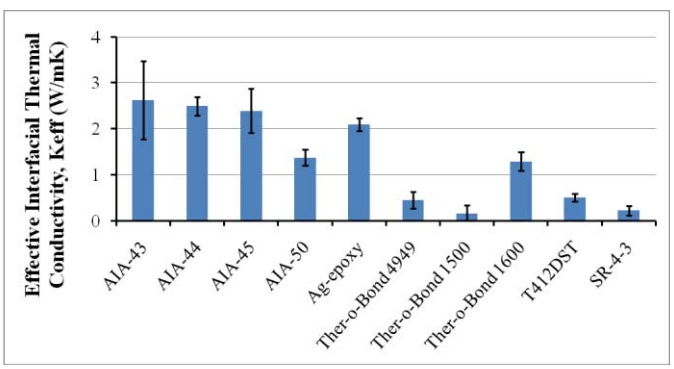


**Fig. 2** Effective interfacial thermal conductivity of sandwich assemblies

The values of the effective thermal conductivity of the MWCNT arrays are higher than the commercial materials, and some samples, such as AIA 43, with an $k_{eff}$ of 2.62 W/mK, exceeded the effective conductivity value of silver epoxy resin (2.00 W/mK). The larger thickness of the arrays (~100 µm) is the factor that limits the thermal performance of the diffusivity and contact resistance. To optimize the CNT arrays for thermal transport, the arrays should be reduced in length for a thinner bond-line. Also, with longer nanotubes there is more opportunity for defects that create phonon scattering and reduce the thermal transport capabilities.

## Conclusions

Multiwall carbon nanotube (MWCNT) arrays possess superior thermal conductivity along the direction of the MWCNT axes. Harvested epoxy-infiltrated MWCNT arrays with adhesive properties as well as thermal transport capabilities make them a great candidate for TIMs. The arrays presented here out-perform most commercial adhesives used in thermal interface material applications. When the MWCNT arrays bond two aluminum discs, simulating actual interface conditions, the thermal interface contact resistance values were as low as 20 $mm^2K/W$. Commonly used commercial adhesives only reached 38.68 $mm^2K/W$. Decreasing contact resistance will drastically increase the thermal performance of a component.

The aligned MWCNT arrays, with diffusivity values higher than isotropic graphite, exhibits excellent thermal transport performance in many applications. However, there is much area for improvement in the development of MWCNT arrays. The thermal performance can be drastically increased by reducing the thickness of the interface layer while increasing the concentration of carbon nanotubes within the contact area, which greatly influences the effective thermal conductivity. Further research will focus on the delicate balance of these parameters, while obtaining continuous arrays for large, commercial applications.

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