

Supporting Information

A. TEM Microtome Sample Preparation



Figure S1. Polydimethyl siloxane mold used to prepare TEM samples

A polydimethylsilicone mold as shown in Figure S1 was first partially filled with Epon 862/W and then the epoxy was cured at 250°F for 3 hours. Next the graphite foil with nanotubes was placed in the mold and the mold was filled with epoxy resin and cured in a similar fashion as discussed earlier. Next TEM sections were microtomed from the potted specimen on a RMC Products Inc. PowerTome XL instrument. A 35° Diatome diamond knife was used to prepare the sections. The sections were collected on pre-cleaned 200 mesh copper grids with a lacey amorphous carbon layer. An FEI Titan 300, 80 - 300 KeV TEM was used for this study. The resolution of the microscope at 300 kV was ~ 1 Å. All the samples were investigated at 80 KeV.

B. Calculation of k_{eff}

Let us assume that the intrinsic conductivity of the nanotube be k . To calculate k_{eff} , we have to consider effective cross-sectional area perpendicular to the thermal transport which is significantly different than the ones that used in the prediction of k using modeling techniques such as molecular dynamics.

In MD simulations, the area that is used for thermal conductivity simulations corresponds to a cylindrical shell of width t and diameter D .

First we have to modify this area by actual cross-sectional area which is $\pi D^2/4$.

$$= k * \frac{\pi t}{\pi D}$$

$$(k_{NT})_a = k * \frac{\pi D t}{\pi D^2/4} = k * \frac{4t}{D}$$

This is the thermal conductivity which whole area of cylinder is taken into account. We have to also consider that because of the circular geometry, these cylinders can only pack up ~90% of the total surface area based on closest hexagonal packing criteria in 2 dimensions. This leads to

$$(k_{NT})_{pk} = 0.9 * (k_{NT})_a = k * \frac{4t}{D} * 0.9$$

We also have to consider that these cylinders are not well packed. In fact, only 20% the whole surface area is covered by these nanotube forests (please see Figure 3 in main text). Considering that as well, we find that effective value of conductivity becomes

$$k_{eff} = 0.2 * (k_{NT})_{pk} = k * \frac{4t}{D} * 0.9 * 0.2 = 0.72 * k * \frac{t}{D}$$

For our case, we find that the diameter (D) of the multi-wall CNTs was of the order of 20-30 nm. In addition, t should be less than $D/4$. Given $D \sim 20-30$ nm, we can assume $t \sim 2-3$ nm (corresponding to 7-10 layers in multi-wall CNTs). Incorporating these, into the above equation, we get

$$k_{eff} = 0.072 * k$$

Here, if we assume intrinsic value of MWCNTs to be $k = 100$ W/m-K, we get $k_{eff} \sim 7$ W/m-K. To compare, Kim et al [PRL, 2001, 87, 215502] reported the value of ~ 265 W/m-K for individual CNTs. When the nanotubes are bundled together in a forest like geometry, the phonon transport does get affected significantly due to inter-tube interactions, thus lowering the intrinsic thermal conductivity of the nanotubes. Given the number of defects present along its length (for example bamboo like structure, branching), its curvature, fusing (intermingling) with other

nanotubes, we believe that our assumption of $k = 100$ W/m-K is very safe and towards higher side.