ADVANCED MATERIALS

Supporting Information

for *Adv. Mater.*, DOI: 10.1002/adma.201300059

Thermal and Electrical Transport in Ultralow Density Single-Walled Carbon Nanotube Networks

*Ke Jia Zhang, Abhishek Yadav, Kyu Hun Kim, Youngseok Oh, Mohammad F. Islam, Ctirad Uher, and Kevin P. Pipe**

Supporting Information for adma.201300059

Blackbody radiation losses

Blackbody radiation losses from the sample can be expressed by $Q_{\mathrm{radiation\_env}} = A_{\mathrm{side}}\sigma_{\mathrm{b}}\varepsilon(T^4\text{-}T_{\mathrm{a}}^4)$, where A_{side} is the area of the exposed (side) surface of the sample, σ_{b} is the Stefan-Boltzman constant, $\varepsilon \approx 1$ is the emissivity (estimated from data for a CNT array[9]), $T_{\mathrm{a}} = (T_3+T_4) / 2$ is the sample mean temperature, and T is the environmental temperature.

Parasitic conduction from thermocouples to environment

Parasitic heat conduction from the thermocouples to the environment can be found by calculating $Q_{\mathrm{conduction\_loss}} = (k_{\mathrm{copper}}+k_{\mathrm{cons}})A_{\mathrm{TC}}\Delta T / l_{\mathrm{TC}}$ for thermocouples TC3, TC4, TC5, and TC6, where k_{copper} and k_{cons} are the thermal conductivities of copper and constantan, A_{TC} is the cross-sectional area of the thermocouple wires (which have a diameter of 10μm), $l_{\mathrm{TC}} \approx 14$ cm is the length of the thermocouple wires, and ΔT is the change in temperature from the sample to the environment.

Effect of interface thermal resistance

Letting ΔT_{int} be the temperature drop at the SWCNT aerogel/silver paste interface and ΔT_{S} be the temperature drop across the 3mm-thick aerogel sample, the measured temperature drop is given by $2\Delta T_{\mathrm{int}} + \Delta T_{\mathrm{S}}$, and has a value in the range of 30 – 35 K. For a SWCNT/metal interface, the highest reported thermal boundary resistance (TBR) is 9.2 mm^{2}K/W.[10] To account for the difference in volume fraction (ϕ) for the SWCNT array in reference 10 (2-3%) and that of our SWCNT aerogels (0.63%), we use the relation $R_{\mathrm{int,i}} = R_{\mathrm{int}} \times \phi$, where $R_{\mathrm{int,i}}$ is the TBR of an interface between a metal and an individual SWCNT. This yields $R_{\mathrm{int}} = 9.2\times(2.5/0.63) = 36.5$ mm^{2}K/W for an aligned SWCNT array with the same volume concentration as the aerogels. Using $\kappa \approx 0.05$ and 0.02 W/mK (for the as-grown and Gr-coated aerogel, respectively), we find $2\Delta T_{\mathrm{int}} / (2\Delta T_{\mathrm{int}} + \Delta T_{\mathrm{S}}) = 1.2\times10^{-3}$ and 4.8×10^{-4} for an aligned SWCNT array with the volume concentration of the as-grown and Gr-coated aerogel, respectively.

We next consider the effect of SWCNT alignment on TBR. For an isotropic 3D network of rods (here, SWCNTs), the average number of SWCNTs crossing a plane can be calculated as:[11]

$$\left\langle N_{\mathrm{S}}\right\rangle = \frac{A}{ld}\frac{\overline{n_V}}{2}\left(1+4\frac{d}{l}\right); \overline{n_V} = n_V l^2 d \qquad (1)$$

where A is the sample cross-sectional area (diameter 10 mm), l is the tube length (1 μm), d is the tube diameter (0.93 nm), and $n_{\mathrm{v}} = \phi / (\delta dl)$ is the volume number density of SWCNTs derived from the volume fraction ϕ. For an aligned SWCNT array, the average number of SWCNTs at the top surface can be calculated as $N_{\mathrm{A}} = A\phi / (\pi\delta d)$. The ratio $N_{\mathrm{S}} / N_{\mathrm{A}}$ is $(1+4d/l) / 2$, which is very close to 0.5 for the SWCNT aerogels. Based on the results of aligned arrays above, we therefore find $2\Delta T_{\mathrm{int}} / (2\Delta T_{\mathrm{int}} + \Delta T_{\mathrm{S}}) = 2.4\times10^{-3}$ and 9.6×10^{-4} for an isotropic (non-aligned) SWCNT array corresponding to the as-grown and Gr-coated SWCNT aerogel, respectively.

Radiation within the aerogels

SWCNT aerogels have optical properties similar to carbon aerogels, including optical thickness,[12, 13] making photon diffusive theory an appropriate means to calculate radiation transfer within the SWCNT aerogel.[13] The effective radiative conductivity of the aerogel can be expressed by $k_{\mathrm{r}} = 16n^2\sigma_{\mathrm{b}}T_{\mathrm{a}}^3 / 3\rho e_{\mathrm{temp}}$,[13] where the refractive index n can be calculated using the Clausius-Mossotti formula,[15] $(n^2\text{-}1) / (n^2+2) = \phi(n_{\mathrm{carbon}}^2\text{-}1) / (n_{\mathrm{carbon}}^2+2) + (1\text{-}$

ϕ)(n_{vacuum}^2-1) / (n_{vacuum}^2+2), for which n_{carbon} = 2 is the solid backbone refractive index,[14] n_{vacuum} = 1 is the pore refractive index, and ϕ is the SWCNT volume fraction. The temperature-dependent specific extinction coefficient e_{temp} can be derived from the frequency-dependent specific extinction coefficient using the Rosseland weighting function[13] and ranges between 550 m^2/kg and 800 m^2/kg over the temperature range of 100 – 300 K.

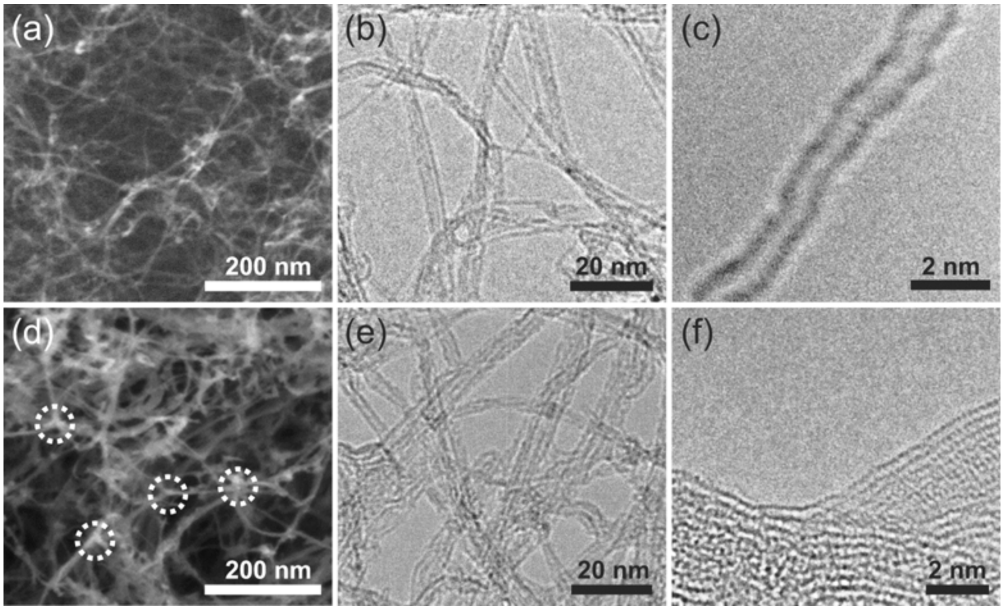

Figure S1. Microstructure of as-grown and Gr-coated SWCNT aerogels. Images (a-c) of as-grown SWCNT aerogels were obtained by (a) scanning electron microscopy and (b) low- and (c) high-resolution transmission electron microscopy, and show three-dimensional, isotropic networks of nanotubes. Similarly obtained images for Gr-coated SWCNT aerogels (d-f) show an accumulation of graphene at the nodes between the nanotubes, highlighted by white circles in (d). The graphene also coats ~25-40% of the nanotube surfaces.

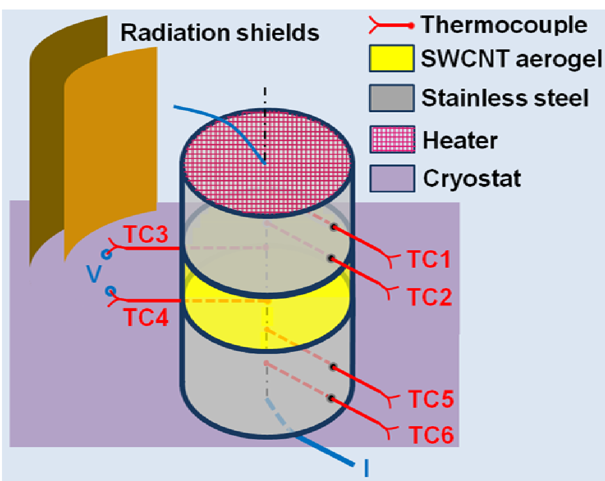

Figure S2. Experimental setup. The aerogel sample is sandwiched between two stainless steel plates, allowing the heat flux to be measured. Measurements are performed in vacuum to prevent parasitic convection, with two radiation shields used to reduce parasitic radiation losses.

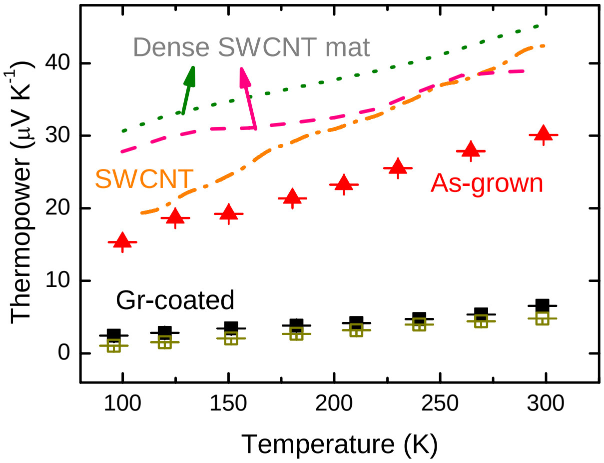

Figure S3. Thermopower. ▲: as-grown, ■(black and olive): Gr-coated, dash-dot line: single (10,0) SWCNT,[20] dashed line: laser-synthesized dense SWCNT mat,[41] dotted line: arc-synthesized dense SWCNT mat.[41]