Tunable Thermoelectric Power Factor in Semiconducting Single-Walled Carbon Nanotube Networks

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Single-walled carbon nanotubes (SWCNTs) are a versatile electronic material being explored as cost-effective, high-performance active materials in a variety of renewable energy applications such as transparent conducting or light-harvesting layers in photovoltaics and inclusions in thermoelectric composites. We present a series of experiments focused on understanding the thermoelectric performance of enriched semiconducting SWCNT networks dispersed in a semiconducting polymer matrix. Rational choice of the semiconducting polymer allows us to sensitively tune the s-SWCNT diameter and band gap distributions within the composites. We use a stable charge-transfer dopant to control the density of carriers in the s-SWCNT network, as determined by the bleach of the absorption corresponding to the S11 excitonic transition. The performance of these transparent conducting s-SWCNT composite networks is comparable to neat p-type and n-type s-SWCNT networks doped by either nitric acid or hydrazine treatments. By varying the carrier density we are able to probe the relationship between the electrical conductivity and Seebeck coefficient (thermopower) in the s-SWCNT networks as a function of the carrier density and position of the Fermi energy. Although the electrical conductivity of the s-SWCNT networks is poor at very low carrier densities we have measured a colossal thermopower as high as ~2,500 µV/K, which is more than an order of magnitude larger than has been previously reported for SWCNT-based material systems and is consistent with theoretical calculations that consider the density of electronic states in individual s-SWCNTs. As we tune the carrier density, we are able to maintain a thermopower above 200 µV/K over almost the entire range of hole densities, corresponding to conductivities up to 1885 S/m, resulting in a thermoelectric power factor of ~100 µW/m•K2. These studies suggest that the low dimensionality of the SWCNTs has a stronger impact on the electrical conductivity than the thermopower, implying that they are less strongly coupled in these systems than is observed for compound inorganic semiconductors. We also present a sensitive technique, based on a microfabricated silicon nitride thermal isolation platform, to probe the thermal conductivity in the s-SWCNT networks, allowing us to estimate the thermoelectric figure of merit (zT) for these materials to be ~ 0.01. These observations demonstrate the ability to exert exquisite control of the thermoelectric performance by tuning the carrier density and/or Fermi energy, and touts SWCNTs as an avenue for realizing thermally stable room temperature thermoelectric devices fashioned from inexpensive and abundant organic constituents.