

Fine-tuned carbon nanotubes scavenge for waste heat

Thermoelectric materials serve key roles in electronics, transportation, and manufacturing. They can recover energy from waste heat produced by automobiles or other engines. Conversely, electrical energy applied to these materials can effectively pump heat away from sensitive electronics to actively cool high-power computers and communications systems. The performance of a thermoelectric for either process depends on the power factor, which scales with electronic conductivity and an intrinsic (material-dependent) Seebeck coefficient. Existing materials have, to date, delivered insufficient performance. Carbon nanotubes have demonstrated promising thermoelectric properties; however, their electronic structure has remained under-optimized for this application. The largest reported samples constituted only tiny flakes; such a scale is insufficient to meet the challenges of most heat-generating systems.

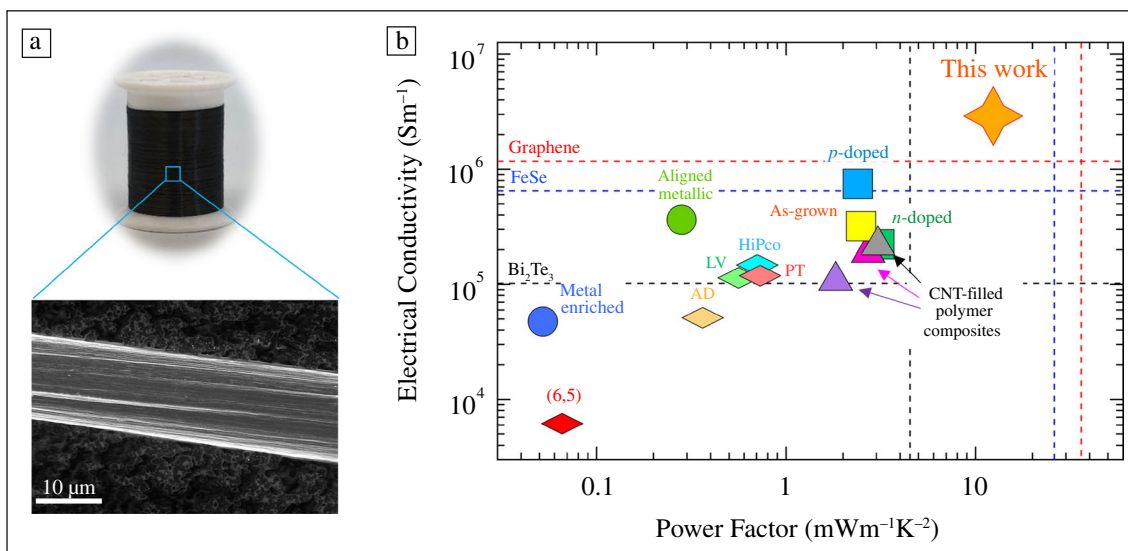
To break through these limitations, researchers from Rice University, along with collaborators from the Tokyo Metropolitan University, developed carbon nanotube weaves that exhibited two crucial properties. First, carbon nanotubes in these weaves were highly aligned, pure, dense, and electronically conductive. Second, the electronic band structure of these nanotubes was fine-tuned to maximize the Seebeck coefficient. The research team's effort yielded carbon nanotube weaves with a power factor of $14 \text{ mW m}^{-1} \text{ K}^{-2}$, a record for carbon nanotube thermoelectrics. With a 60-nanotube thermoelectric proof-of-concept prototype, the team, which is led by Rice University's Junichiro Kono, successfully harvested $5 \text{ } \mu\text{W}$ from a 60°C temperature gradient, which was then amplified to power a green-colored light-emitting diode. Their experimental approach and findings, and the corresponding theoretical foundation behind their work, appeared in a recent issue of *Nature Communications* (<https://doi.org/10.1038/s41467-021-25208-z>).

Natsumi Komatsu, a graduate student in Kono's group and first author on the article, told *MRS Bulletin*,

"Low-dimensional materials have exhibited large thermoelectric power factors, but such demonstrations have been limited to micrometer-scale samples. It is exciting that we were able to preserve giant power factor in meter-long fibers, which are also weavable and washable. Our fibers will make promising building blocks for the emerging technologies of fiber and textile electronics."

The team turned to commercially sourced double-walled carbon nanotubes to develop the nanoscale building blocks of their high-power thermoelectric concept. Matteo Pasquali, a co-author of the article, and his team dissolved the 1.8-nm-wide nanotubes in chlorosulfonic acid, extruded the resulting slurry, and spun it into a fiber weave. The fibers, which were $9\text{-}\mu\text{m}$ wide each, maintained a high electrical conductivity of $>10 \text{ MS/meter}$ and a tensile strength of 4.2 GPa . The resulting mats exhibited high Seebeck coefficients of $>60 \text{ } \mu\text{V/K}$ and thermal conductivities that reached $580 \text{ W/m}\cdot\text{K}$.

The main source of these exceptional values stemmed from the team's chemical pretreatment of the weaved carbon nanotube fibers with chlorosulfonic acid



(a) Photograph and electron microscope image of carbon nanotube (CNT) fiber. (b) Comparison of reported power factor values for various CNT samples as a function of electrical conductivity, including from this work. Credit: *Nature Communications*. AD, arc discharge; LV, laser vaporization; PT, plasma torch.



or iodine monochloride, as well as thermal annealing at 350–500°C. These treatments maximized hole (*p*-type) doping of the nanostructures, and, subsequently, shifted the materials' Fermi energy levels to approach that of one of the one-dimensional van Hove singularities. At these critical inflection points in the material's density of states, electron wave equations exhibit “kinks” that translate into

observable changes of optoelectronic properties. One-dimensional materials, such as carbon nanotubes, benefit from quantum confinement at this critical density of states intersection of energy bands to exhibit the maximum Seebeck coefficient. At the same time, the team's efforts to improve the sample morphology sustained the nanotube fibers' electrical conductivities.

This careful, fine-tuned balance enabled the resulting thermoelectric mat to exhibit unprecedented power factors. The coupling of this fundamental breakthrough with the high scalability of this approach opens new opportunities for waste-heat-to-energy generators and active coolants of supercomputers.

Boris Dyatkin

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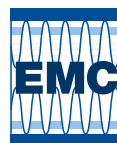
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