

Thin-Film Thermoelectric Conversion Devices for Direct Thermal-to-Electric Conversion for DC and Pulse Power

Rama Venkatasubramanian, Jonathan M. Pierce, Meiyong Himmtann, Geza Dezsi, and Yo-Rhin Rhim

ABSTRACT

New propulsion technologies are critical to developing new capabilities in Department of Defense platforms. An innovative approach taking nuclear heat and directly generating DC electric power with solid-state thermoelectric devices, without the need for a steam power plant, can lead to reliable and compact systems while offering several system-level advantages. These advanced thermal-to-electric device developments are also applicable to efficient radioisotope thermoelectric generators (RTGs) for space outer-planetary missions. Similarly, many platforms, and special operations in particular, need very compact (lightweight, small volume) pulse electric power sources with high specific power density in the range of $\sim 1,000$ kW/kg and a long shelf life. This article describes progress with fundamental scientific advances relevant to these thermal-to-electric conversion applications leveraging recent advances in nano-engineered thin-film thermoelectric materials.

NEEDS AND REQUIREMENTS

Direct Thermal-to-Electric Conversion for DC Power

New nuclear propulsion technologies, especially affordable solutions to increase a fleet's capability to address emerging threats, are critical to ensuring the performance of US Department of Defense (DOD) missions. Switching from nuclear-powered steam power to direct thermal-to-electric conversion (DTEC) is expected to reduce the size of the power plant significantly, creating unprecedented opportunities for new capabilities in future Navy vessels as well as in other DOD platforms. Implementing innovative solutions that also improve naval capability and safety would benefit the US Navy's strategic deterrent missions.

Past efforts aimed toward achieving this objective have proposed a design for a prototypical large-scale (~ 1 MW) DTEC power system based on advanced modular thermoelectric conversion devices. Key requirements for implementing thermoelectric conversion at the fundamental heat-to-electric conversion device level in such DTEC systems include (1) higher efficiencies ($>12\%$ with temperature differentials of $\sim 350^\circ\text{C}$) and (2) electric power densities >2 W/cm². In our early research efforts leveraging recent advances in nano-engineered thin-film thermoelectric materials, we have begun to address these two metrics to set the stage for

further advanced development of DTEC systems. These results are discussed below.

Microscale Energetic Thermoelectric Generators for Pulse Power

Several DOD platforms, like the chip-scale satellite system¹ shown in Figure 1a, need highly compact (light-weight, small volume) pulse electric power sources with high specific power in the $\sim 1,000$ kW/kg range and a long shelf life. Similarly, many special operations need compact pulse power sources, such as those requiring high-power RF communication (Figure 1b). Supercapacitors (with 10 kW/kg) and electrolytic capacitors (50 kW/kg) can self-discharge and offer shorter shelf lives. To address these needs, we combined the controlled fast release (~ 10 ms) of heat from an oxidizer (chemical energy) stored in a nickel-aluminum (Ni-Al) wafer or a nanoscale-porous-silicon (NPS) wafer with a high-speed (~ 10 ms) thin-film thermoelectric heat-to-electric power device to demonstrate a highly compact peak power source on demand.

DEVELOPMENT HIGHLIGHTS

We have developed nano-engineered thin-film thermoelectric materials² to improve the figure of merit (ZT)

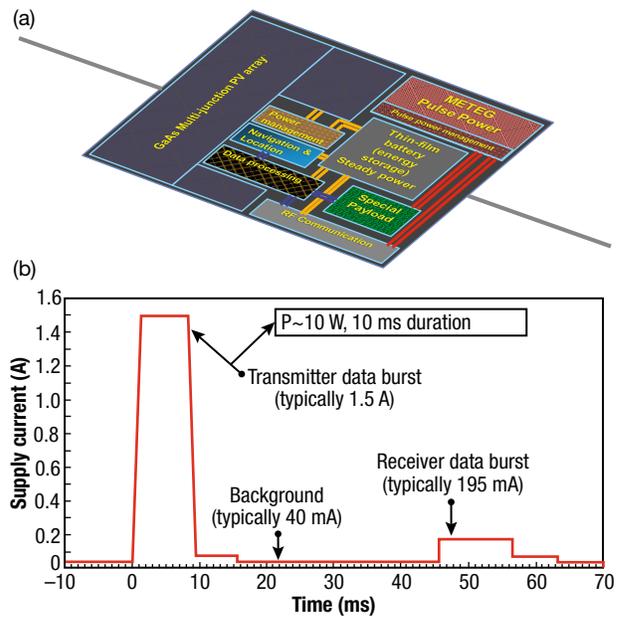


Figure 1. Microscale energetic thermoelectric generators (METEG) applications. (a) New-generation small-scale satellite systems like chip-scale satellite systems¹ and (b) RF communication devices in ground systems can benefit from METEG pulse power sources.

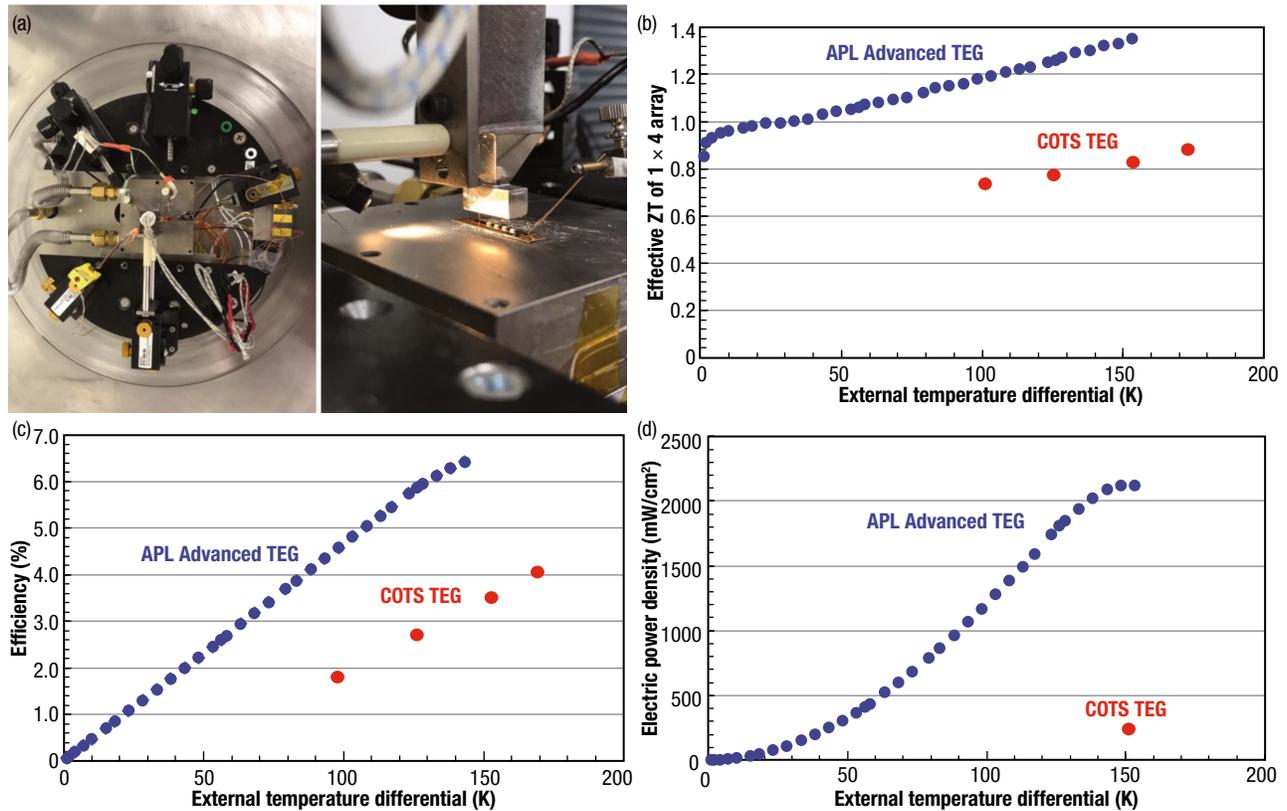


Figure 2. Setup and results. Experimental setup for testing thin-film thermoelectric devices (a) and performance curves that describe the advancements achieved over commercial off-the-shelf (COTS) thermoelectric devices, including (b) effective ZT as a function of temperature, (c) thermal-to-electric conversion efficiency, (d) and electric power density.

of both p-type and n-type materials to values approaching ~ 3 and ~ 2 , respectively, at 300 K. In addition, with these materials the device-level p-n couple ZT has been improved to ~ 1.5 at 300 K. These are significant advances to the state of the art.³ Such high ZT values are fundamentally required for achieving the 15% efficiency targets in DTEC applications; in addition, other advances in thermal interfaces and thermal management are necessary. We evaluated these advanced materials for both steady-state and pulse thermal-to-electric power conversion devices.

The thin-film nano-engineered thermal-to-electric conversion devices, built as a 1×4 p-n couple array, were tested with a thermally managed heat sink and a calibrated heat source, as shown in Figure 2a. The effective device ZT (from efficiency measurements), the efficiency versus temperature differential (DT) across the thin-film device, and the power density are shown in Figure 2, b–d, respectively. The data from the best commercial off-the-shelf (COTS) bulk Bi_2Te_3 -alloy-based devices are also indicated. We observe that (1) the thin-film module-level device ZT approaching ~ 1.4 is about 70% better than state-of-the-art COTS bulk Bi_2Te_3 -alloy materials; (2) heat-to-electric conversion efficiencies are 65% better than COTS materials in the temperature range of 32–200°C; (3) and electric power densities are significantly better than those achievable with COTS materials. These are critical and enabling results with thin-film nano-engineered thermoelectric materials and devices applicable to eventual DTEC applications. Such high efficiencies can add onto the high-temperature and mid-temperature portions of a three-stage cascade.⁴ If the total efficiencies of heat-to-electric conversion are higher, then for a fixed electric power, less heat needs to be rejected at the heat-sink/radiator, so lower cold-side temperatures can be tolerated, further improving the efficiencies.

The Ragone plot in Figure 3a shows the comparison of the proposed METEG devices for two different types of energetics discussed above (i.e., NPS and Ni-Al) with other state-of-the-art pulse power sources such as lithium thin-film batteries, supercapacitors, carbon onions micro-supercapacitors, and electrolytic

supercapacitors. Whereas energy storage devices like supercapacitors and electrolytic capacitors have varying degrees of an inverse relationship between power density and energy density, METEGs do not show such an inverse relationship. This is because they are essentially electrical pulse power generation devices from ultrarapid conversion of stored chemical energy to heat and then to electric power. Hence, improving the efficiency of heat-to-electric conversion and/or heat transfer from the chemical-to-heat process to then electric, while keeping the volume of the thin-film thermoelectric device about the same, allows for favorable scaling of power density without sacrificing energy density.

A cross-sectional schematic showing the essential aspects of the METEG is shown in Figure 3b. The details of the various layers that constitute the METEG are also shown in Figure 3c. The METEG includes the NPS energetic array, which serves as a micro-combustion heat generator. The NPS layer is formed out of the top 25 μm of a typical silicon wafer, as shown in Figure 3c; in the case of Ni-Al energetics, the NPS layer is replaced by a 5- to 10- μm alternating layers of Ni-Al (each about 1 μm). In both cases, the energetics or heat production

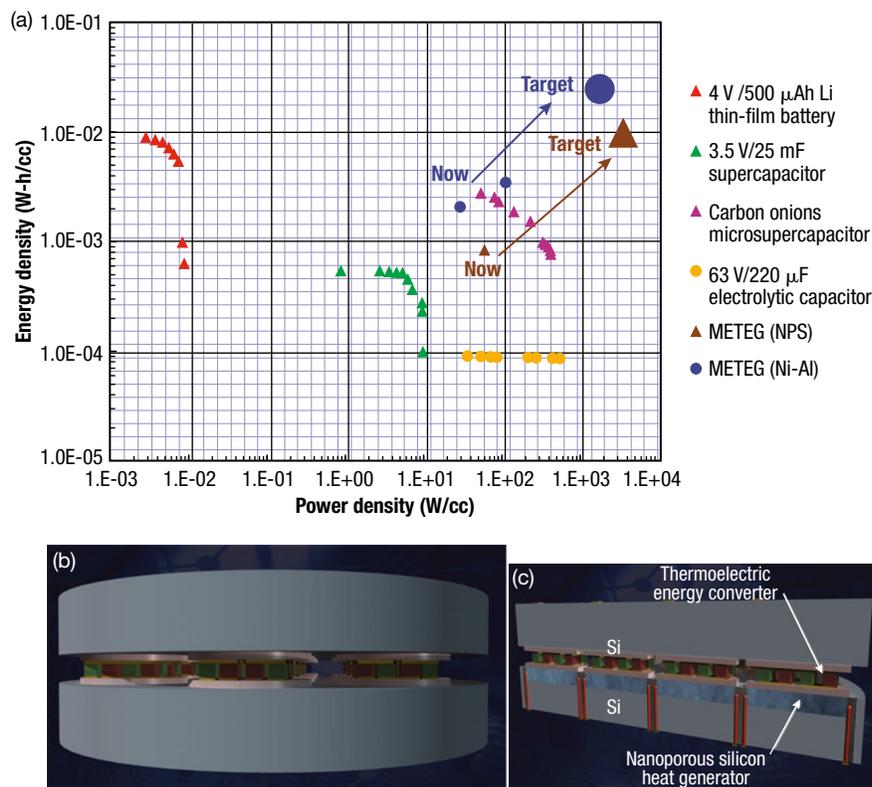


Figure 3. Capabilities and details of the METEG concept. Top, Plot of energy density versus power density for a variety of technologies such as lithium-ion batteries, electrolytic capacitors, supercapacitors,⁵ and METEG technologies developed in this effort; bottom left, schematic of a compact, packaged METEG device that can be developed based on the reported results; bottom right, inner details of the METEG device showing the thermoelectric converter and other aspects.

is locally initiated by electrically heated elements that can be photolithographically processed onto the silicon wafer, shown schematically in Figure 3c.

The top half of the METEG is the thermoelectric energy converter, which converts the heat from combustion into electrical energy for providing bursts of power on demand for a wide variety of applications—for instance, pulse radio frequency transmissions, as shown in Figure 1b; for taking a high-power radar snapshot; and for charging a supercapacitor bank. Making the micro-combustion portion and the energy converter on separate wafers provides more flexibility in optimizing each part of the system. Just as important, this allows the potential reuse of the same top half, the thermoelectric converter wafer, when all of the heat-generating NPS elements or Ni-Al elements on a silicon wafer have been used up; the bottom half can be replaced with a new silicon wafer containing the energetics elements. Such multiuse applications can be important for small satellite and chipsat systems that require on-demand pulse power for long periods of operation.

In summary, we have described significant steps toward validating advanced nano-engineered thin-film thermoelectric materials and devices for both steady-state (DC) and pulse power thermal-to-electric conversion applications. The developments in DTEC power devices show early promise and, with further development, could be attractive for efficient Navy propulsion systems converting nuclear heat to electric power, offering significant benefits in weight, volume, agility, and cost. These DTEC developments are also of interest for radioisotope thermoelectric generators (RTGs)

operating on extremely cold planets and moons, as well as for recovering some of the vast amount of unutilized waste heat in today's world of energy-intense applications and living.^{6,7} The pulse power applications based on the METEG technology are expected to be of value in man-portable or deployable unattended power systems in space or on the ground as well as in micro-robots⁸ with peak power demand needs.

REFERENCES

- ¹M. Peck, "Exploring space with chip-sized satellites," *IEEE Spectr.*, Jul. 28, 2011, <https://spectrum.ieee.org/aerospace/satellites/exploring-space-with-chipsized-satellites>.
- ²R. Venkatasubramanian, J. Pierce, and G. Dezzi, Superlattice Structures for Thermoelectric Devices, US Patent No. 10,903,139 (filed Sep. 11, 2017, and published Jan. 26, 2021).
- ³R. Venkatasubramanian, E. Siivola, T. Colpitts, and B. O'Quinn, "Thin-film thermoelectric devices with high room-temperature figures of merit," *Nature*, vol. 413, pp. 597–602, 2001, <https://doi.org/10.1038/35098012>.
- ⁴B. Cook, T. E. Chan, G. Dezzi, P. Thomas, C. C. Koch, et al., "High-performance three-stage cascade thermoelectric devices with 20% efficiency," *J. Electron. Mater.*, vol. 44, pp. 1936–1942, 2015, <https://doi.org/10.1007/s11664-014-3600-9>.
- ⁵F. Wang, X. Wu, X. Yuan, Z. Liu, Y. Zhang, et al., "Latest advances in supercapacitors: From new electrode materials to novel device designs," *Chem. Soc. Rev.*, vol. 46, no. 22, pp. 6816–6854, 2017, <https://doi.org/10.1039/C7CS00205J>.
- ⁶The Energy Flow Chart Released by Lawrence Livermore National Laboratory, Estimated Energy Consumption in the US 2016, Mar. 2017, https://flowcharts.llnl.gov/content/assets/images/charts/Energy_Energy_2016_United-States.png.
- ⁷R. Venkatasubramanian, "Power from nano-engineered wood," *Nat. Mater.*, vol. 18, pp. 536–537, 2019, <https://doi.org/10.1038/s41563-019-0352-1>.
- ⁸W. A. Churaman, L. J. Currano, J. Rajkowski, C. J. Morris, and S. Bergbreiter, "The first launch of an autonomous jumping microrobot using nanoporous energetic silicon," *J. Microelectromech. Syst.*, vol. 21, no. 1, pp. 198–205, 2012, <https://doi.org/10.1109/JMEMS.2011.2174414>.



Rama Venkatasubramanian, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Rama Venkatasubramanian is a project manager, team leader for energy and thermal management, and a member of the Principal Professional Staff in APL's

Research and Exploratory Development Department. He has a BS in electrical engineering from the Indian Institute Technology (IIT, Madras) and an MS and a PhD in electrical engineering from Rensselaer Polytechnic Institute. He has 30 years of progressive experience in the research and development of solid-state energy conversion materials and devices and has led multidisciplinary programs in the development of atomically engineered thin-film superlattices and nano-engineered bulk thermoelectric materials and devices for electronics and photonics thermal management, energy harvesting, and power conversion. Other areas of contribution and expertise include compound semiconductor photovoltaics, topological insulators, and infrared detectors. Rama is a Fellow of IEEE (2011) and the American Association for the Advancement of Sci-

ence (2012) and has earned two R&D 100 awards, has 20 issued US patents, as well as several awards for technical excellence and innovations from US government agencies and the energy conversion professional community. His email address is ramavenkatasubramanian@jhuapl.edu.



Jonathan M. Pierce, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Jonathan M. Pierce is a senior research engineer in APL's Research and Exploratory Development Department. He has a BS in ceramic engineering from Alfred

University and an MS in materials science and engineering from North Carolina State University. Jon has more than 15 years of experience with metalorganic chemical vapor deposition and epitaxial growth of semiconductor materials for thermoelectrics and wide bandgap devices. His email address is jonathan.pierce@jhuapl.edu.

Meiyong Himmtann, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Meiyong Himmtann is a research engineer in APL's Research and Exploratory Development Department. She has a BS in electrical engineering from Iowa State University. Meiyong has experience in semiconductor processing and fabrication, including photolithography, electroplating, and flip-chip bonding, as well as skills in the characterization of semiconductor materials and testing of thermoelectric devices. Her email address is meiyong.himmtann@jhuapl.edu.

Geza Dezsi, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Geza Dezsi is a senior research engineer in APL's Research and Exploratory Development Department. He has a BS in physics and an MS in mechanical engineering from North Carolina Central University. Geza has more than 15 years of experience with engineering design, 3-D computer-aided design (CAD) solid modeling, thermal engineering, and finite element analysis (FEA). He has extensive hands-on experience with semiconductor processing tools and component- and system-level packaging and integration as well as in-depth knowledge of

solid-state sensors, thermoelectric technologies, solar cells, light-emitting diodes, and electromagnetic generators. His email address is geza.dezsi@jhuapl.edu.



Yo-Rhin Rhim, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Yo-Rhin Rhim is a project manager and the acting assistant supervisor of the Materials Development and Integration Section in APL's Research and Exploratory Development Department. She has a BS, an MS, and a PhD in mechanical engineering, all from Johns Hopkins University. Her research activities involve theoretical and experimental studies of carbon materials, specifically on the topics of graphene synthesis, advanced carbon material composites, application of carbon nanomaterials, development of nano-textured surfaces for drag reduction and optical applications, and advancement for power and energy, specifically for thermal batteries and lithium-ion battery materials. Yo-Rhin also has experience with materials characterization and testing techniques involving chemical, microstructural, optical, thermal, and electrical analyses. Her email address is yo-rhin.rhim@jhuapl.edu.