Thermoelectric Power Generation Materials: Technology and Application Opportunities

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Thermoelectric power sources have consistently demonstrated their extraordinary reliability and longevity for deep space missions (67 missions to date, more than 30 years of life) as well as terrestrial applications where unattended operation in remote locations is required. The development of new, more efficient materials and devices is the key to improving existing space power technology and expanding the range of terrestrial applications. The NASA Jet Propulsion Laboratory is leading collaborative research and development on novel advanced bulk materials capable of long-term operation at temperatures up to 1,300 K at more than 20% conversion efficiency. The research areas include refractory rare earth compounds and bulk three-dimensional nanostructures that emulate results obtained on low dimensional superlattices through "force engineering" and "self-assembling" techniques. Recent experimental results will be highlighted, and progress in transitioning thermoelectric technology to a more flexible, lower-cost modular array configuration suitable for various application opportunities will be discussed.

THERMOELECTRIC POWER GENERATION

Thermoelectric (TE) power sources (Figure 1a) have consistently demonstrated their extraordinary reliability and longevity for deep space science and exploration missions as well as terrestrial applications where unattended operation in remote locations is required.[1–3] Radioisotope thermoelectric generators (RTGs) have been continuously operated in some cases for more than 30 years using high-temperature heat sources (up to 1,300 K). They are static thermal-to-electric energy conversion devices (Figure 1b) with a high degree of redundancy (arrays of hundreds of thermocouples), no electromagnetic interferences, no moving parts, no vibrations, and with well-documented "graceful degradation" characteristics. They are easily scalable from milliwatts to hundreds of watts. They are also tolerant of extreme environments (temperature, pressure, shock, and radiation). Small, rugged terrestrial generators based on combustion of fossil fuels have also been successfully developed and have demonstrated an operating life of hundreds of thermal cycles.

How would you…

…describe the overall significance of this paper?

This paper highlights materials and device performance requirements for enabling penetration of thermoelectric power generation technology in various large-scale waste heat recovery applications. An overview of recent advances in materials research and development is provided, together with a brief assessment of the maturity of high-temperature thermoelectric and converter technology.

…describe this work to a materials science and engineering professional with no experience in your technical specialty?

Recent advances in materials synthesis techniques, theoretical modeling and design engineering of complex structure compounds, nanostructured composite materials have led to significant improvements in the thermoelectric figure of merit of some high-temperature materials. Novel approaches to thermal-to-electric device and converter technology developments that make use of these new materials are briefly described. Selected potential large-scale waste heat recovery power applications are listed.

…describe this work to a layperson?

There are very significant opportunities for producing electricity from waste heat recovered from a variety of energy-intensive industrial processes and the exhaust of transportation vehicles. Thermoelectric technology has some unique advantages in terms of ease of integration for retrofitting existing industrial equipment, hybridizing with other power technologies as topping or bottoming cycles, unique scalability and modularity, and low maintenance requirements. Recent advances in thermoelectric materials and device technologies could enable cost-effective applications.

Thermoelectric generators (TEGs) are Carnot heat engines, with electrons performing as the working "fluid."[4] The maximum thermal-to-electric conversion efficiency of a TEG is the product of the Carnot efficiency and a materials factor dependent on the dimensionless thermoelectric figure-of-merit, ZT (Equation 1).

$$\eta_{max} = \frac{T_{hot} - T_{cold}}{T_{hot}} \frac{\sqrt{1+ZT} - 1}{\sqrt{1+ZT} + \frac{T_{cold}}{T_{hot}}} \qquad (1)$$

ZT is a combination of three material transport properties of the n-type and p-type couple elements and the absolute operating temperature (T). It is defined as:

$$ZT = \frac{S^2}{\rho\lambda} T \qquad (2)$$

where S is the Seebeck coefficient ($V \cdot K^{-1}$), ρ is the electrical resistivity ($\Omega \cdot m$), and λ is the thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$) of the thermoelectric material.

Practical thermoelectric materials were discovered in the early 1950s and consisted of heavily doped semiconductors with peak ZT values of only

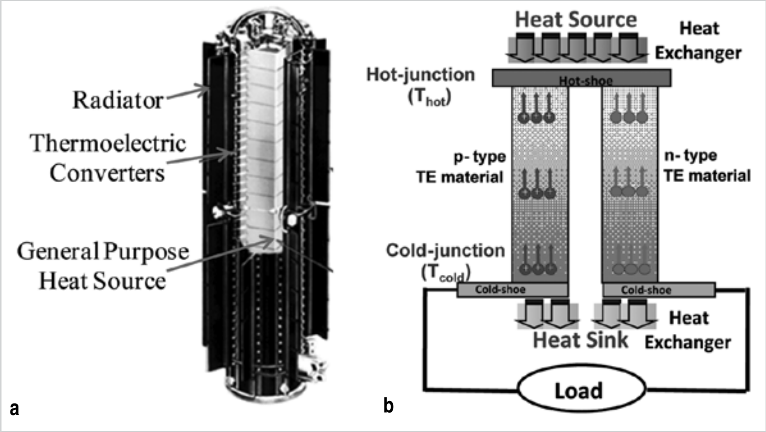

Figure 1. (a) The long-life thermoelectric power source (~300 W of electrical power) used on deep space science exploration missions and (b) a thermoelectric couple operating across a temperature differential and generating electrical power.

about 1, even though ZT has no known theoretical limit. To maximize the Carnot efficiency, thermoelectric power systems need to operate across as large a temperature differential as possible. Radioisotope thermoelectric generators typically operate across ΔT values as large as 700 K, leading to lower effective average values of ZT, ranging from 0.55 to 0.75. This means that state-of-practice TE materials only convert 10% to 15% of the available Carnot efficiency, and that the overall TE power system conversion efficiency ranges from 6.5% for space power sources down to about 3% for portable terrestrial generators.

Thermoelectrics is one of various technologies being considered for producing electricity from waste heat energy. To achieve performance levels comparable to internal combustion engines and dynamic power converter technologies would require effective average ZT values approaching 2 to 4 at the system level depending on that system's thermal and electrical losses (Figure 2). As a result, the key to improving existing space power technology and expanding the range and scale of terrestrial generators, for both civilian and military applications, lies for the most part in the development of new, much more efficient materials and devices.

The key challenges and barriers to the successful development and implementation of integrated TE waste heat recovery technology include: Improving TE materials conversion efficiency in the temperature range of interest; developing rugged, reliable high-temperature TE multi-couple module technology; optimizing heat transfer to/from the TE modules; surviving the operational thermal cycling, vibration stress, and strain environment; minimizing TE generator system weight for automobile applications; ensuring acceptance of this revolutionary technology; energy payback time; and return on investment.

LARGE-SCALE THERMOELECTRIC POWER-GENERATION OPPORTUNITIES

Recoverable Industrial Waste Heat

Extremely large amounts of waste heat energy are generated through inefficiencies of power-generating plants and manufacturing industries. Typically, end-to-end electrical production loses 67% of its available energy, mostly as waste heat with some small fraction coming from distribution losses.[5] Manufacturing industries overall reject about 33% of their energy as waste heat directly to the atmosphere or to thermal management systems because of their inability to recycle the excess energy.[6] In the U.S. manufacturing sector alone, more than 3,000 TWh of waste heat energy is lost each year, an amount equivalent to more than 1.72 billion barrels of oil. The "grade" of industrial manufacturing waste heat varies significantly from one industrial application to another in terms of its temperature, composition, and accessibility. The various temperature ranges are illustrated in Figure 3.

In addition to considerations for practical integration and economical viability of waste heat recovery to electrical power systems, the quality of the waste heat stream in terms of composition, corrosiveness, and mass flow is a key parameter. Many of the industrial processes listed in Figure 3 produce only low-grade waste heat in the form of flue gases in the temperature range of 400 K to 450 K. This is due to the mixing with ambient air in order to minimize costs. Because of the small temperature differential and low conversion efficiency, thermoelectric waste heat recovery systems would have to be extremely inexpensive to implement in order to justify producing significant amounts of electric power.

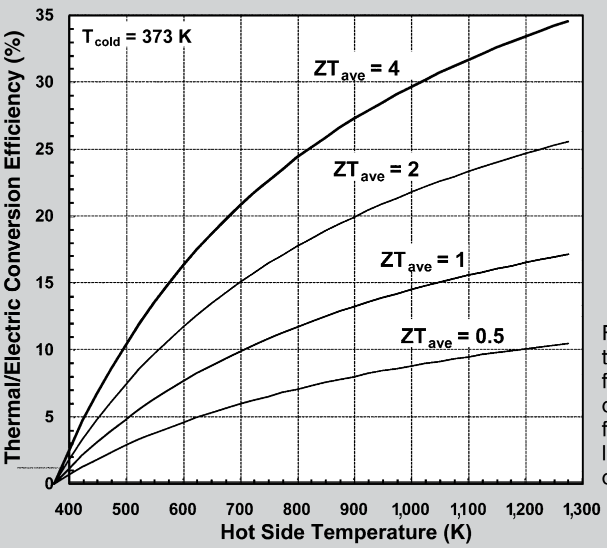

Figure 2. Thermal-to-electric energy conversion efficiency as a function of operating temperature differential and dimensionless thermoelectric figure of merit (ZT).

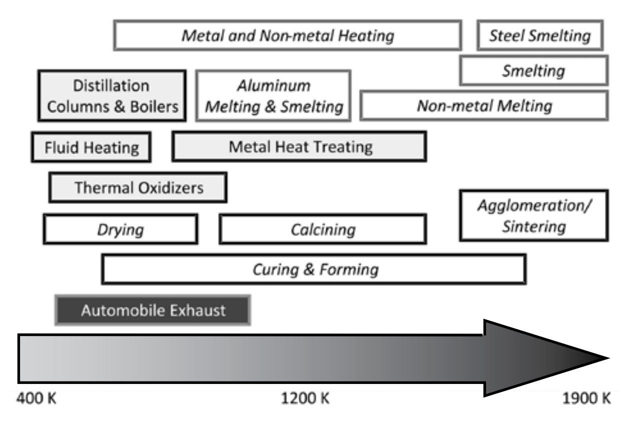

Figure 3. The temperature ranges of waste heat for various industrial processes.

Higher-grade waste heat, as much as 600 TWh per year, is considered to be a much more attractive opportunity for effective waste heat recovery.[6] Aluminum, glass, metal casting, non-metal melting, ceramic sintering, and steel manufacturing all have process furnaces discharging high-temperature waste heat combustion gases and melt pool gases (such as aluminum melting at 1,025 K and glass melting near 1,700 K). Whenever there is limited opportunity to reuse the waste heat and difficulties in effectively transporting that heat to separate energy conversion systems, integrated thermoelectric waste heat recovery systems are potentially attractive bottoming cycle co-generation systems. Recent U.S. studies[6] on near-term waste heat recovery applications determined that between 0.9 TWh and 2.8 TWh of electricity might be produced each year for materials with average ZT values ranging from 1 to 2.

Automotive Exhaust Waste Heat

The automobile industry has also recently developed a strong interest in a waste exhaust heat (Figure 4a) recovery power source operating in the 400 K to 1,000 K temperature range to supplement or replace the alternator and thus decrease fuel consumption.[7–11] Current estimates of available waste thermal power range from 20 kW to 400 kW for light-duty vehicle systems, depending on engine size and operation. Current waste energy assessments indicate that the energy equivalent of 46 billion gallons of gasoline annually is wasted down the exhaust pipe of the 200 million light-duty vehicles in the United States alone.

Recovering part of the waste thermal energy to produce electricity would allow for improving fuel efficiency, help meet increasing electrical loads, and reduce regulated and greenhouse gas emissions. A number of studies have been completed to date and there is ongoing work to develop kW-class TEGs capable of operating up to the 900–1,000 K temperature range and survive thousands of thermal cycles. Several full-scale prototypes have been successfully built using Bi_2Te_3 alloys, but the overall system efficiency remains low due to operating temperature limitations (Bi_2Te_3 alloys cannot operate for extended periods of time beyond 525 K) and heat-transfer inefficiencies. Near-term efforts aim at achieving a 10% improvement in the efficiency of modern internal combustion engines through a combination of new high-temperature thermoelectric materials and efficient heat exchanger technology (Figure 4b).

Solar Thermal

There has long been interest in solar thermal power systems where the solar energy is concentrated and turned into a high-temperature heat source, and then converted to electricity by using a dynamic or static Carnot engine. Large-scale power plants relying on dynamic converters (such as turbines operating on the Rankine cycle) have been built in recent years, with thermal-to-electric conversion efficiencies in the 20% to 30% range for moderate heat source temperatures around 800 K. From Figure 2, for solid-state thermoelectric systems to be performance competitive would require average ZT values near 4 or a combination of a higher heat source temperature and lower ZT (ZT ~2 and heat source ~1,200 K). While it appears unlikely that thermoelectrics can challenge such established technologies in the near future, interest has

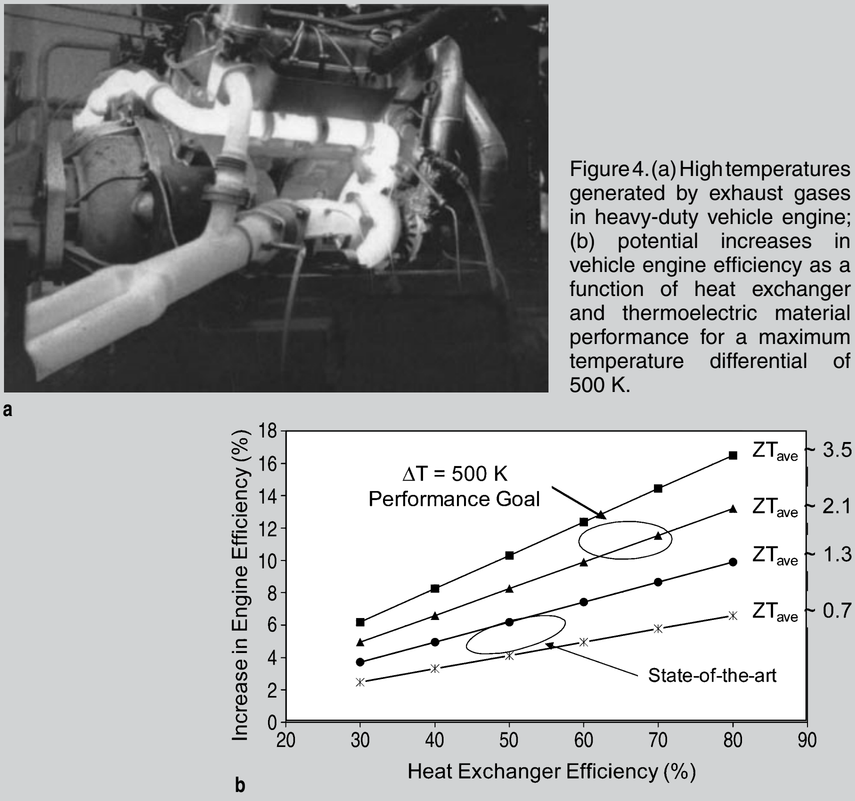

Figure 4. (a) High temperatures generated by exhaust gases in heavy-duty vehicle engine; (b) potential increases in vehicle engine efficiency as a function of heat exchanger and thermoelectric material performance for a maximum temperature differential of 500 K.

been growing for hybrid solar power systems based on all solid-state photovoltaic (PV) and thermoelectric converters.[13,14] The basic concept[13] consists of concentrating and splitting the solar energy spectrum into a low wavelength portion directed at PV cells and a high wavelength portion directed generating high grade heat for thermoelectric modules. Photovoltaic cells efficiently convert the ultraviolet and visible light and removing the infrared portion of the spectrum helps maximize their conversion efficiency by maintaining low operating temperatures. This is especially important for highly concentrated solar systems (up to 1,000×) where, in spite of improved conversion efficiencies, thermal management is a significant challenge. In an attempt to achieve cost-effective large-scale power generation, hybrid system design trades address various technology options. These include, in particular, lower-efficiency (~15–20%) single-junction versus high-efficiency (~30–35%) triple-junction PV cells, low concentration versus high concentration solar concentrators and collectors and their impact on the heat source temperature and the requirements for cooling of the PV cells and waste heat management.

DEVELOPING EFFICIENT HIGH-TEMPERATURE THERMOELECTRIC MATERIALS

Since the 1950s, the main approach to developing advanced thermoelectric materials was focused on identifying, characterizing, and optimizing bulk degenerate semiconductors. After the initial discoveries of such materials as Bi_2Te_3, PbTe, Bi-Sb, and Si-Ge alloys, which are still today's state-of-practice materials, experimental research efforts were unable to achieve peak ZT values significantly larger than 1.0 at any temperature.[14] The best results were obtained for p-type TAGS materials (GeTe-$AgSbTe_2$ compositions) with peak ZT values around 1.2. A significant portion of these efforts was devoted to optimizing thermal and electrical transport by controlling the type and amount of doping impurities as well as forming solid solutions. Solid solutions, such as $Bi_{2-x}Sb_xTe_{3-y}Se_y$ and $Si_{1-x}Ge_x$, have been used to tune the band gap for maximizing ZT values at a given temperature, but mostly to reduce the lattice thermal conductivity through point defect scattering of the phonons. However, introducing point defects typically also led to degradation in charge carrier mobility values, thus severely limiting overall enhancement in ZT.

It has been understood for some time that decoupling charge carrier and phonon scattering mechanisms to enable independent optimization of the electrical (Seebeck coefficient, carrier mobility, and electrical resistivity) and thermal (lattice thermal conductivity) transport properties are required to achieve much larger ZT values.[15] In the 1980s and early 1990s, a number of studies on bulk materials evaluated potential routes to lower thermal conductivity with limited impact on electrical properties, using "self-assembling" and "force-engineering" approaches to material synthesis. Self-assembling approaches were used to synthesize from the melt a number of layered complex chalcogenide materials that used building blocks from compounds such as Bi_2Te_3, PbTe, and GeTe.[16] Force-engineered approaches focused in particular on reducing the lattice thermal conductivity of Si-Ge alloys prepared through powder metallurgy processes, using techniques such as reducing grain size down to a few micrometers,[17] and introducing defects by neutron radiation[18] or dispersions of ultrafine inert insulating particulates.[19] While these studies advanced the state-of-the-art in materials syntheses and provided highly valuable scientific insight in understanding thermoelectric transport, improvements in ZT values were only marginal. In most cases this was due to the fact that decreases in thermal conductivity values were nearly matched by equivalent reductions in the charge carrier mobility values (and thus increases in electrical resistivity values).

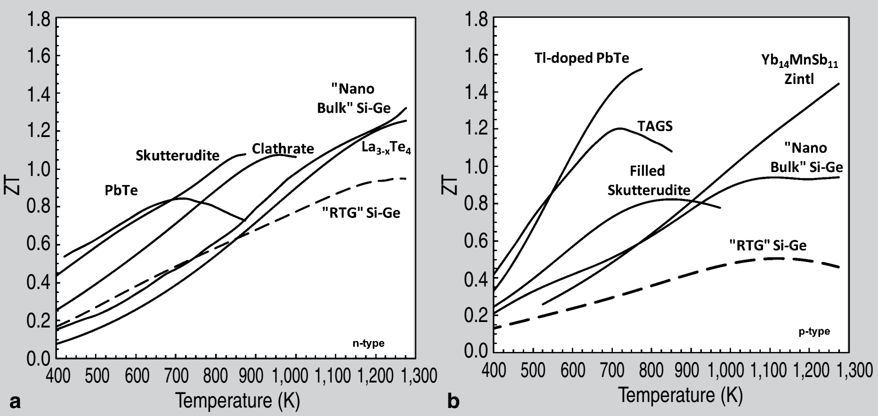

Figure 5. ZT values as a function of temperature for (a) n-type and (b) p-type bulk thermoelectric materials characterized at JPL.

In the mid-1990s, building on improved control in the synthesis of complex materials and structures, two different approaches were actively investigated for developing the next generation of new thermoelectric materials: one using new families of advanced bulk thermoelectric materials, and the other using low dimensional materials systems. The advanced bulk materials focused on complex structure compounds (see for example Reference 20) such as filled skutterudites ($CeFe_4Sb_{12}$), clathrates ($Ba_8Ga_{16}Ge_{30}$), Chevrels ($Cu_xMo_6Se_8$), Zintls ($Yb_{14}MnSb_{11}$), and rare earth tellurides ($La_{3-x}Te_4$). These materials typically exhibit very low lattice thermal conductivity values, and a great deal of effort has been spent on optimizing their electrical properties by controlling stoichiometry, introducing atomic substitutions and doping impurities. The best ZT values observed at the NASA Jet Propulsion Laboratory (JPL) are in the range of 1.0 to 1.5 at various temperatures, as reported in Figure 5. A very recent work on p-type Tl-doped PbTe has experimentally demonstrated a ZT improvement mechanism linked to the distortion of the electronic density of states.[21]

Several studies on low-dimensional, high-quality thin film structures, such as superlattices based on Bi_2Te_3 and PbTe materials, have shown that a significant increase in ZT could be achieved. While some of these structures exhibited enhancements in their electrical properties, most of the gains were attributed to scattering of phonons at interfaces leading to large reductions in lattice thermal conductivity values.[22,23] Such low-dimensional thin film structures are, however, ill suited to the demands of high-temperature power generation applications that require long term thermal and mechanical stability as well as efficient coupling with existing heat sources. Compared to bulk materials, cross-plane thin film devices would require very high heat flux densities to generate significant temperature differentials, while in-plane thin film devices are extremely sensitive to thermal shunts from supporting substrates and insulation packaging. In addition, it could prove to be a very significant challenge to scale up material synthesis and device fabrication to meet the potential needs of terrestrial waste heat recovery applications.

A more recent strategy consists of replicating the nanoscale features responsible for enhanced ZT values in bulk materials using advanced synthesis techniques. Several groups have reported a significant increase in ZT in some bulk materials, such as silver antimony lead telluride and its alloys, attributed mostly to the formation of "self-assembling" nanoclusters inside a host matrix.[24,25] To address the need for long-term material stability at temperatures as high as 1,300 K, a nanostructured bulk and composite materials "force-engineered" approach focused on $Si_{1-x}Ge_x$ compositions and other electronic semiconductors has been proposed.[26,27] This approach aims at generating a very large interface density, such as that formed in a superlattice, into dense bulk materials prepared from nanoparticles. Results to date have succeeded in improving significantly ZT values over that of state-of-practice "RTG" $Si_{0.8}Ge_{0.2}$ alloys, in particular for p-type materials, as shown in Figure 5. Of particular concern to high-temperature power generation applications is the thermal stability of such nanostructures. While there is ample experimental evidence that for some material systems long-term thermal stability (i.e., grain growth and interdiffusion) may be a very difficult challenge, preliminary short-term studies have shown nano bulk $Si_{1-x}Ge_x$ to be highly stable. After ageing the samples for more than three months at 1,275 K, results show that the thermal conductivity remained much lower than that of "RTG" $Si_{0.8}Ge_{0.2}$ material, with any variation within the experimental error limits. The ability to scale material synthesis in similar fashion to state-of-the-art materials produced through more traditional powder metallurgy techniques, combined with likely long-term thermal stability, are critical enabling requirements. While results will undoubtedly vary from one material class to another, it is clear that nano bulk $Si_{1-x}Ge_x$ materials are suitable candidates for high-temperature thermoelectric applications. This work has now expanded to III–V compounds, skutterudites, and Bi_2Te_3 materials.

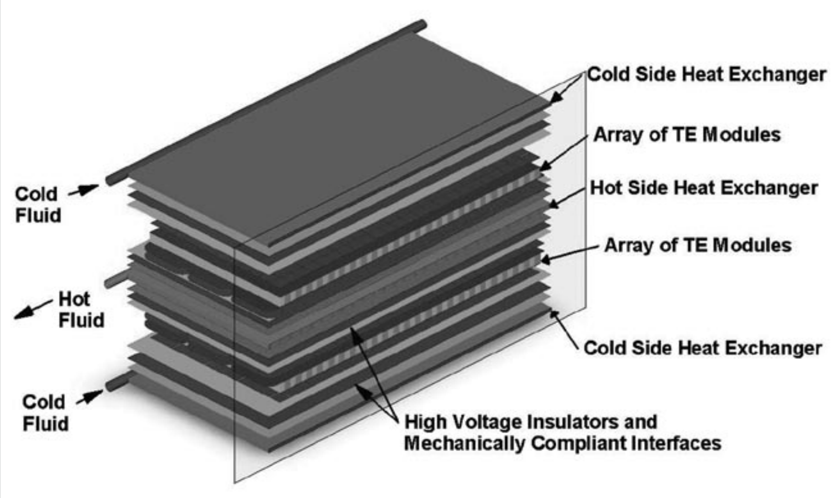

Figure 6. The conceptual design of a 1.5 kW spacecraft thermoelectric generator assembly. The cut-away shows the thermoelectric couple arrays conductively coupled to a central hot liquid metal heat exchanger and two cold side liquid metal heat exchangers located on both external sides.

THERMOELECTRIC GENERATOR TECHNOLOGY DEVELOPMENT

To fully reap the benefits of high ZT thermoelectric materials, device- and system-level losses must be kept to a minimum.

Heat Exchangers

For a given temperature differential, the waste heat flux density is inversely proportional to the thickness of the thermoelectric elements. Scaling thermoelectric converter devices down to very small dimensions means operating with very large heat fluxes, in the hundreds of W/cm^2. Typical heat flux densities available are usually more in the 1–40 W/cm^{2}range, which means that it is quite difficult to minimize thermal losses for thermoelectric elements much smaller than 5 mm to 10 mm in thickness. Adding to the challenge of high-temperature heat transfer is the fact that most waste heat gas streams have components that contaminate, degrade, and corrode the surfaces of heat exchangers. For waste heat recovery applications, it is preferable when possible to design cold-side heat exchangers to operate with a liquid as the heat exchange medium. Liquids have thermal conductivities and capacities an order of magnitude higher than that of gases, which allows the cold-side heat exchanger to use proven technologies and operate as low in temperature as possible.

Thermoelectric Converter

To illustrate the process of developing efficient high-temperature thermoelectric power systems, a conceptual TE power converter system design effort was conducted for a 100 kW-class space power system consisting of an array of generator units (shown in Figure 6). Each generator unit was comprised of arrays of thermoelectric multicouple

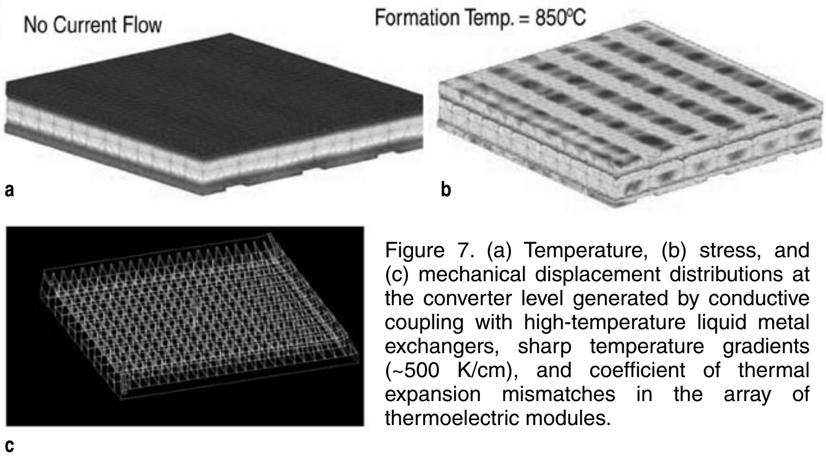

Figure 7. (a) Temperature, (b) stress, and (c) mechanical displacement distributions at the converter level generated by conductive coupling with high-temperature liquid metal exchangers, sharp temperature gradients (~500 K/cm), and coefficient of thermal expansion mismatches in the array of thermoelectric modules.

modules. This segmented thermoelectric multicouple converter (STMC) was designed for long-term steady-state operation at high temperatures (1,275 K to 725 K) and used counter-flow liquid metal heat exchangers to transfer about 30 W/cm^2 of heat between the hot-side and cold-side liquid metal loops.[28,29]

The proposed system was to experience a few thermal cycles due to start-up, shut-down, and safe modes of the heat source. While various candidate thermoelectric materials were considered at the time of the study, all configurations shared the same types of challenges in terms of practical integration of the thermoelectric multi-couple modules with the heat exchangers and survival under both transient and long-term steady-state operation. Thermal expansion mismatches within the TE modules, between the TE modules and the heat exchangers, and “bowing” of the TE modules subjected to sharp temperature gradients are all very relevant challenges to the successful development of waste heat TE power systems (). Automotive waste heat recovery systems additionally require the ability to survive thousands of thermal cycles during the vehicle lifetime.

The STMC design effort focused on developing rugged “conductively coupled” design approaches to ensure that the converter survives fabrication and assembly stresses as well as minimize stress under steady-state operation to enable long life. A solution consisted of redistributing thermally induced stresses by selecting optimal materials combinations and element geometries combined with simplifying and streamlining fabrication steps. To that effect a rapid STMC configuration evaluation tool was developed using an elastic model of the stack of material components that allows comparing trends and identifying key risk areas.[29] This was followed by a detailed thermal stress and loads analysis model at the full converter level that included an optimization for limiting parasitic thermal losses to less than 10%.

The detailed converter design work led to the development of first-generation subscale STMC modules composed of four or eight couples of new thermoelectric materials (skutterudites) capable of operating up to 975 K. The objective was to determine the best bonding methods and process conditions for preparing multicouple layer-to-layer bonds, but also to attempt to minimize the number of high-temperature assembly steps on the most fragile components, possibly the high voltage insulators and the thermoelectric elements. In addition, the fabrication approach addressed one of the key issues for high-power thermoelectric converters, namely scaling up of the various fabrication and assembly processes. A number of key innovations (Figure 8) were developed during the course of this program that concluded in 2005 due to budgetary constraints. The main features of the easily customizable and scalable aerogel-filled STMC module design and interconnection schemes are shown in Figure 8. Key innovations include: functionally grading the multicouple stack of materials to minimize stresses due to coefficient of thermal expansion mismatches; pre-fabrication of five discrete component subassemblies using separate optimized processes; alignment and indexing of all the component subassemblies using vaporizable egg crates; one-step bonding of the component subassemblies by using low-temperature intermetallic diffusion bonding processes amenable to long-term operation at significantly higher temperatures; and backfilling and thermal packaging of the assem-

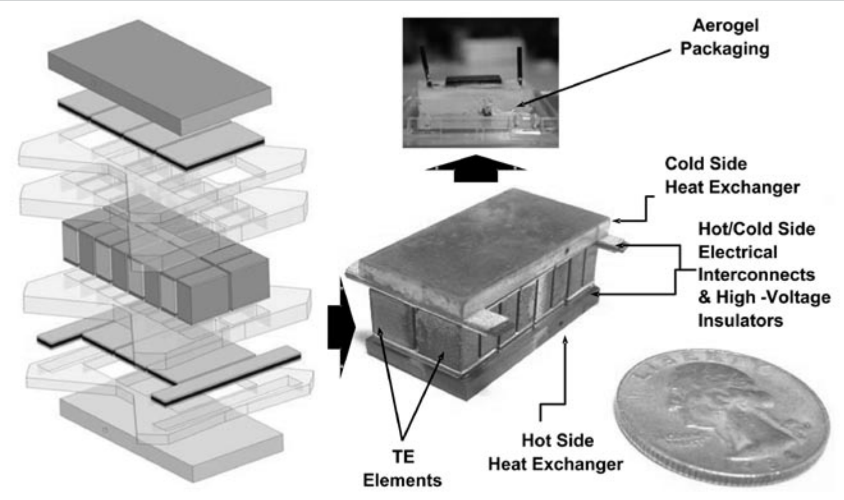

Figure 8. Eight-couple thermoelectric module using skutterudite materials with two parallel strings of four couples in series each. The egg-crate plates used for alignment and indexing of the component have vaporized, leaving empty spaces between the thermoelectric legs which are subsequently backfilled with thermal insulation (aerogel cast to fill the network of TE legs).

bled STMC module using specially designed refractory castable electrically and thermally insulating aerogel. The lightweight, nanoporous aerogel was also used as a barrier to potential sublimation products from the thermoelectric elements. The five sub-assemblies consisted of the cold side and hot side high-voltage insulator/electrical interconnects, the metallized TE elements, and the cold and hot side heat exchanger headers.

About ten STMC devices were built in successive iterations. The thermoelectric legs were prepared by dicing fairly large "pucks" of skutterudite materials (40 mm in diameter) which included thick metallizations. Other components such as the high-voltage insulators with electrical interconnects were either procured from commercial vendors or custom-fabricated at JPL. Measurements on the last set of thermoelectric modules demonstrated that they were able to operate across a large temperature differential (~350 K) and under high heat flux conditions (~25 W/cm^2) for extended periods of time (up to 1,500 hours). The parasitic thermal losses were less than 10%, and the internal device resistances were within 5% of initial predictions, leading to about 5% conversion efficiency. Remarkably, the module components did survive the few thermal cycles they were subjected to, from room temperature up to 1,000 K. Long-term tests pointed to the fact that significant room for optimization remains, but the design and fabrication approaches are compatible with a number of thermoelectric materials capable of operating in the 800 K to 1,300 K range.

CONCLUSION

There are very significant opportunities for producing electricity from waste heat recovered from a variety of energy-intensive industrial processes and the exhaust of transportation vehicles. Thermoelectric technology has some unique advantages in terms of ease of integration for retrofitting existing industrial equipment, hybridizing with other power technologies as topping or bottoming cycles, unique scalability and modularity, and low maintenance requirements. However, thermoelectrics are often seen as a low performance, immature, and costly technology for application to the megawatt-class power generation systems. Most recent studies point to the fact that the ZT values of state-of-practice materials only lead to thermoelectric generators of marginal performance. Improvements by a factor of 2 at least are required to truly enable penetration of TE technology on a large commercial scale. Additional considerations for thermoelectric materials include the cost of the constituent elements and their environmental suitability. In the last 15 years, significant progress in materials synthesis and processing coupled with better understanding of how to engineer superior thermoelectric materials has brought some very promising results. Building on prior technology development, efforts to integrate some of these new materials into new thermoelectric devices and systems have been initiated. Because of its fairly low power level (~1 kW), moderate temperature range, and potential large scale use, automobile thermoelectric waste heat recovery constitutes a unique opportunity to design, develop, validate and demonstrate technologies that are critical to a widespread application of thermoelectric generators.

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