

**High Efficiency Rare-Earth-Based Thermoelectric Materials for Space Power Generation Applications.** S. K. Bux<sup>1</sup>, J. M. Ma<sup>1,2</sup>, S. C. Clark<sup>1,2</sup>, T. Vo<sup>1</sup>, Alexandra Zevalkink<sup>1</sup>, Yufei Hu<sup>3</sup>, Jason Grebenkemper<sup>3</sup>, David Uhl<sup>1</sup>, Billy C.-Y. Li<sup>1</sup>, C.-K. Huang<sup>1</sup>, Susan Kauzlarich<sup>3</sup>, P. Von Allmen<sup>1</sup>, and J.-P. Fleurial<sup>1, 2</sup>, <sup>1</sup>Jet Propulsion Laboratory/California Institute of Technology, 4800 Oak Grove Drive, MS: 277-207, Pasadena, CA Sahbah.K.Bux@jpl.nasa.gov, <sup>2</sup>Department of Chemistry, University of California, Los Angeles, 607 Charles E. Young Drive, Los Angeles, CA 90095 <sup>3</sup>Department of Chemistry, University of California, Davis, One Shields Drive, Davis, CA 95616

**Abstract.** Radioisotope thermoelectric generators (RTGs) are solid state devices that rely upon a temperature gradient to generate power (with the heat supplied by the radioisotope). Due to their solid state nature, these generators have a high level of reliability and longevity as well as a high level of redundancy. As a result, RTGs have been a key enabling technology for many missions for the past 50 years. Although reliable and relatively low risk, the heritage system efficiency based upon either PbTe or SiGe has been limited to about 6.5%. As the power demands for missions increases, higher efficiency thermoelectric systems are necessary to support the larger scientific payload. The goal of the Advanced Thermoelectric Materials project in the Advanced ThermoElectric Converter (ATEC) program at the Jet Propulsion Laboratory (JPL) is to develop the next generation RTG through the development of TE materials that are capable of providing 20% conversion efficiency (~15% system level conversion efficiency).

Thermoelectric generators are effectively Carnot engines and the thermal-to-electric conversion efficiency,  $\eta$ , of a material is defined as the product of the Carnot efficiency and a materials conversion efficiency factor,  $f$  (equation 1), which is directly related to the dimensionless thermoelectric figure of merit,  $ZT$  (equation 2).

$$(1) \eta = \frac{\Delta T}{T_h} f(ZT), \quad f = \frac{\sqrt{1+ZT} - 1}{\sqrt{1+ZT} + \frac{T_c}{T_h}}$$

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

Where  $S$  is the Seebeck coefficient,  $\rho$  is the electrical resistivity and  $T$  is the absolute temperature and  $\lambda$  is the total thermal conductivity which is the sum of the electronic thermal conductivity  $\lambda_e$  (due to the charge carriers moving through a lattice) and the lattice thermal conductivity  $\lambda_L$  of the structure. The ideal thermoelectric material has the electronic properties of heavily doped semiconductor, with low metal like resistivity and high Seebeck coefficients, and the thermal

properties of glass. As a result of these seemingly paradoxical properties, the  $ZT$  of state-of-the-art (SOA) materials remained relatively stagnant at a  $ZT$  of 1 for both p- and n-type materials across the 300-1275 K temperature range until recently. Recent efforts to improve the efficiency of thermoelectric materials has relied upon several strategies: nanostructuring of traditional heavily-doped semiconductors to reduce the thermal conductivity, band engineering to enhance the Seebeck coefficient and using materials with complex crystal structures that possess intrinsically low lattice thermal conductivities

New classes of higher performance materials based upon rare-earth based complex structures such as n-type  $\text{La}_{3-x}\text{Te}_4$  and p-type  $\text{Yb}_{14}\text{MnSb}_{11}$  have been developed over the last few years with peak  $ZT$ s as high as 1.3 at 1275 K, a factor of 2 improvement over the heritage combination of p-type and n-type  $\text{Si}_{0.8}\text{Ge}_{0.2}$  alloys. The high  $ZT$  in these materials is attributed to inherently low glass-like lattice thermal conductivity brought by structural complexity and unique covalent bonding. These material systems often possess small band gaps in which the electronic properties of the materials can be tuned via structural modifications or by chemical substitutions. We will present an overview of recent research efforts and describe approaches for tuning the properties of these rare earth compounds through experimental chemical substitutions coupled with guidance from first principle electronic structure simulations achieve 20% conversion efficiencies.<sup>1</sup>

#### References

1. Bux, S. Fleurial, J.-P., & Caillat, T. in 11th Int. Energy Convers. Eng. Conf. (American Institute of Aeronautics and Astronautics, 2013). doi:doi:10.2514/6.2013-3927