Paper ID: 40605
student

MATERIALS AND STRUCTURES SYMPOSIUM (C2)
Interactive Presentations (IP)

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SYNTHESIS AND CHARACTERIZATION OF PR(3-X)TE(4) AND ND(3-X)TE(4) FOR HIGH TEMPERATURE THERMOELECTRIC APPLICATIONS

Abstract

Radioisotope thermoelectric generators (RTGs) have been a vital electrical energy generation technology for many NASA space missions such as Voyager, Cassini, New Horizons, and Curiosity. RTGs are employed when mission environments are not optimal for photovoltaics, such as deep space probes where the solar flux is limited or on missions to the moon or Mars where day and night cycles or dust can disrupt photovoltaic operation. Furthermore RTGs have demonstrated long term reliability, in some cases having been continuously operated for over three decades. The dimensionless figure of merit, denoted ZT, is a measure of the conversion efficiencies of the thermoelectric materials in the RTGs and is defined as ZT=$(\sigma S^2/\kappa)$T where is electrical conductivity, S is the Seebeck coefficient, is thermal conductivity, and T is temperature. One of the current limitations to RTGs is that the heritage thermoelectric materials, SiGe and PbTe, have ZT values of approximately 1 at 1275K, which corresponds to system level efficiencies of 6.5%. Therefore increasing the material efficiencies in the RTGs is vital for increasing scientific payload or reducing nuclear fuel costs for future missions.

Lanthanum telluride ($La_{3-x}Te_4$) has recently emerged as a high efficiency thermoelectric material. The performance of the $La_{3-x}Te_4$ system stems from a complex thorium phosphide (Th_3P_4) crystal structure resulting in an inherently low lattice thermal conductivity. Additionally, the La^{3+} vacancies directly control the carrier concentration of the material, resulting in tunable electronic properties. With an optimized stoichiometry, the ZT of this system can attain values of 1.2 at 1275K. Computational modelling indicates that the conduction band states are dominated by the La d-orbitals. Introduction of states near the Fermi level would have a significant impact on the thermoelectric properties. Praseodymium and neodymium tellurides ($Pr_{3-x}Te_4$ and $Nd_{3-x}Te_4$) are $La_{3-x}Te_4$ analogues and density functional theory (DFT) calculations indicate both have a sharp peak in the conduction band near Fermi level from the f-orbitals in these materials. We utilized mechanochemical approach to synthesize $Pr_{3-x}Te_4$ and $Nd_{3-x}Te_4$ powders with various vacancy concentrations. The powders were compacted using spark plasma sintering (SPS) and the compacts were characterized using X-ray diffraction, scanning electron microscopy, and electron microprobe analysis. The temperature dependent electrical resistivity, Seebeck coefficient, and thermal conductivity will be presented. Preliminary data indicates that there is a large increase in the Seebeck coefficient from the f-orbitals, resulting in increased ZT values of 1.7 for $Pr_{3-x}Te_4$ and 1.8 for $Nd_{3-x}Te_4$ at optimized vacancy concentrations.