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INVESTIGATION OF CO-SN-TE PHASE SPACE FOR ADVANCEMENT OF SKUTTERUDITE MATERIALS FOR RADIOISOTOPE THERMOELECTRIC GENERATORS

Abstract

Radioisotope Thermoelectric Generators (RTG) power systems have been utilized for NASA missions for over 50 years, providing power to missions as diverse as Voyager and Mars Science Laboratory. Within an RTG, a plutonium-238 core radiates heat to thermoelectric generators (TEGs), which are composed of numerous thermocouples that dictate the overall efficiency of the system. The next generation of RTGs, the enhanced-multi mission radioisotope thermoelectric generator (e-MMRTG), aims to increase efficiency of the system via advancement of materials for the thermocouples. Such new materials could lead to increased performance of the e-MMRTG, leading to longer missions and/or a reduction in plutonium load, and ultimately cost. The thermocouple system is composed of one n-type leg and one p-type leg. The system operates at a hot-side and a cold-side which allows the Seebeck effect to induce a voltage via a temperate gradient. This generates electricity via the diffusive flow of carriers to provide the RTG with power. Investigation of advanced materials for the couples include various chemical flavors of the skutterudite structure; a crystal structure that can be described by a network of anion rings surrounding a transition metal species. This network builds a large interstitial void within the ring framework. The most basic skutterudite: $CoSb_3$, is a cubic structure that shows promise as a thermoelectric (TE) material because it has a high intrinsic carrier mobility, and therefore a high electrical conductivity. However, the material lacks outstanding thermoelectric performance since it has high intrinsic thermal conductivity. Introducing a mixed-anion split to the framework by changing the chemistry to $CoSn_{1.5}Te_{1.5}$ leads to increased disorder of the structure, since it changes from cubic ordering to rhombohedral ordering. This decreases the thermal conductivity as a direct consequence. Optimization of $CoSn_{1.5}Te_{1.5}$ could lead to better TE materials for use in e-MMRTGs; however, the $CoSn_{1.5}Te_{1.5}$ phase space on the Co-Sn-Te ternary diagram needs to be explored to understand the dopability of this system. We investigate the $CoSn_{1.5}Te_{1.5}$ phase region by synthesizing various off-stoichiometry compositions of $CoSn_{1.5}Te_{1.5}$ via traditional bulk synthesis to understand the phase boundary. Utilization of powder x-ray diffraction allows us to define the Alkamade triangles surrounding the skutterudite phase region by investigation of the phase separation that occurs. The ternary diagram formed by this study gives new insight to whether or not this material can sustain large dopant concentrations necessary to optimize the thermal and electrical properties to enhance the next generation of RTGs.