SKUTTERUDITES: HOW DO THEY FARE AGAINST STATE-OF-PRACTICE THERMOELECTRIC MATERIALS USED IN RADISOTOPE THERMOELECTRIC GENERATORS?

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Abstract: Of the various static energy conversion technologies considered for Radioisotope Power Systems for space applications, thermoelectric (TE) energy conversion has received the most interest. Radioisotope Thermoelectric Generators (RTGs) generate electrical power by converting the heat released from the nuclear decay of radioactive isotopes (typically plutonium-238) into electricity using a thermoelectric converter. RTGs have been successfully used to power a number of space missions including the Apollo lunar missions, the Viking Mars landers, Pioneer 10 and 11, and the Voyager, Ulysses, Galileo, and Cassini outer planet spacecrafts. These generators have demonstrated their reliability over extended periods of time (tens of years) and are compact, rugged, radiation resistant, scalable, and produce no noise, vibration or torque during operation. These properties have made RTGs suitable for autonomous missions in the extreme environment of the outer space and on planetary surface. Converter units use TE materials, which, when operating over a temperature gradient, produce a voltage called the Seebeck voltage. System conversion efficiency for state-of-practice RTGs is about 6%. The most widely used TE materials in RTGs, in order of increasing temperature, are: tellurides of Antimony, Germanium and Silver (TAGS); lead Telluride (PbTe) and lead Tin Telluride (PbSnTe); and Silicon Germanium (SiGe). All of these materials have been used in RTGs, which have been flown on space missions.

In the early 1990s, a systematic search for advanced thermoelectric materials resulted in the "rediscovery" [1] of a family of attractive semiconducting materials with the skutterudite crystal structure. Skutterudite is a cobalt arsenide with variable amounts of nickel and iron and that was first mined in Skutterdud, Norway in 1845 [2]. The skutterudite structures are of the form $(Co,Rh,Ir)(P,Sb,As)_3$ and are cubic with space group Im3. Unfilled, these materials contain voids into which low-coordination ions (usually rare earth elements) that can be filled in order to alter thermal conductivity by producing sources for lattice phonon scattering and decrease thermal conductivity. These filled compositions correspond to the chemical formula LM_4X_{12}, where L is a rare earth metal, M a transition metal and X a group V element or pnicogen. The ability of filling the voids with a variety of guest atoms result in many options for tuning the electrical and thermal properties of skutterudite materials. Those opportunities have been widely explored worldwide since the 1990s, especially for TE applications. ZT > 1 have been reproducibly achieved for both p- and n-type materials which have been considered for a variety of applications including automobile waste heat recovery and RTGs.

Many physical, mechanical, and TE performance requirements must be met before these materials can compete with state-of-practice (SOP) thermoelectric materials in practical applications. This paper will review the thermal-physical, mechanical, and transport properties of the most promising skutterudite compounds and compare them to SOP TE materials such as PbTe and SiGe. The data acquired to date on these materials shows that they fare well both in terms of TE performance and mechanical strength relative to SOP TE materials.

References:

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