

Investigations into Mechanism of High Entropy Telluride Formation

G. Dey, S. Soliman, R. Schaak

High entropy tellurides (HETs), in the nanoscale, have recently gained interest owing to their superior and exotic thermoelectric and superconductivity properties compared to their constituent counterparts. HETs have been traditionally accessible only via rapid heating at very high temperatures followed by rapid quenching which traps the metastable entropy stabilized phase. HETs can be synthesized by solution-based colloidal techniques, at significantly lower temperatures, where the driving force of formation is a complex interplay among entropy, enthalpy, and chemical reactivities. At present, little is known about the reaction pathways and synergistic reactivities in colloidal synthesis that allow the formation of HETs, hence we need rational design rules to selectively target these materials. Here, we present a generalizable solution based synthetic method for the formation of HETs and investigate their formation mechanism. $(\text{NiPdPtRhIr})\text{Te}_2$ was synthesized directly as well as by a post-synthetic modification technique where the corresponding alloy was converted into the telluride. However, $(\text{SnPdPtRhIr})\text{Te}_2$ could only be synthesized directly and not from the alloy. Temporal investigation of these materials indicated that these HETs form rapidly, and the product outcome is determined by the dominant stable crystal structure of the end members. However, the metastable phases can be kinetically trapped by crystal structure engineering to override the thermodynamic product. This mechanism and reaction pathway thereby provides guidelines to access unconventional materials trapped in crystal phases that haven't been accessed or studied before. A third reaction pathway was uncovered where a diffusion-based pathway can be leveraged to utilize bulk Te as a reagent. Controlling the diffusion and reactivity rates of the metals is crucial in this case to form homogeneously mixed products. Overall, this work provides the design guidelines to selectively target novel HETs for thermoelectric and superconductivity applications.