Experimental and theoretical investigation of Cr$_{1-x}$Sc$_x$N solid solutions for thermoelectrics

The ScN- and CrN-based transition-metal nitrides have recently emerged as a novel and unexpected class of materials for thermoelectrics. These materials constitute well-defined model systems for investigating mixing thermodynamics, phase stability, and band structure aiming for property tailoring. Here, we demonstrate an approach to tailor their thermoelectric properties by solid solutions. The trends in mixing thermodynamics and densities-of-states (DOS) of rocksalt-Cr$_{1-x}$Sc$_x$N solid solutions (0 ≤ x ≤ 1) are investigated by first-principles calculations, and Cr$_{1-x}$Sc$_x$N thin films are synthesized by magnetron sputtering. Pure CrN exhibits a high power factor, 1.7 × 10$^{-3}$ Wm$^{-1}$K$^{-2}$ at 720 K, enabled by a high electron concentration thermally activated from N vacancies. Disordered rocksalt-Cr$_{1-x}$Sc$_x$N solid solutions are thermodynamically stable, and calculated DOS suggest the possibility for power-factor improvement by Sc3d orbital delocalization on Cr3d electrons giving decreasing electrical resistivity, while localized Cr3d orbitals with a large DOS slope may yield an improved Seebeck coefficient. Sc-rich solid solutions show a large improvement in power factor compared to pure ScN, and all films have power factors above that expected from the rule-of-mixture. These results corroborate the theoretical predictions and enable tailoring and understanding of structure-transport-property correlations of Cr$_{1-x}$Sc$_x$N.