INFLUENCE OF MASS AND CHARGE DISORDER ON THE PHONON THERMAL CONDUCTIVITY OF SOME HIGH ENTROPY CERAMICS BY MOLECULAR DYNAMICS SIMULATION

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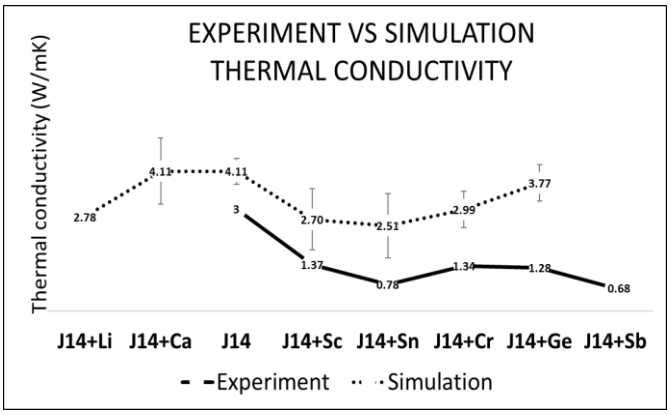
We are exploring how the interplay between mass and charge disorder affects the thermal conductivity of high entropy ceramics that have potential use as ultra-high temperature materials and their oxides. Recent experiments by our team, for example, have shown that the thermal conductivity of the entropy stabilized oxide $(Mg_{0.1}Co_{0.1}Ni_{0.1}Cu_{0.1}Zn_{0.1})O_{0.5}$, termed J14, is reduced by the addition of a sixth cation Sc, Sn, Cr, Ge or Sb in an equi-molar proportion. Classical phonon transport theory cannot account for this reduction based on mass scattering alone. Therefore we have been using molecular dynamics simulations to gain a better insight of the combined effects of disorder in mass and in electrostatic interactions on phonon-mediated thermal conductivity for these systems.



Our initial oxide simulations model short-range interatomic interactions with an exponential-6 pair sum, and model long-range electrostatics via atom-centered point charges. The pair sums and an initial set of point charges were fit to the bulk properties of MgO. To model the entropic materials, the same pair sum is used, but with the electrostatic charges are fit to Bader charges from Density Functional Theory calculations. Despite the relative simplicity of this molecular model, it is able to capture many of the variations in thermal conductivity seen experimentally (see figure). Our simulations suggest that taken together, both changes in the average mass and in the electrostatic interactions within these materials can account for much of the reduction in thermal conductivity, with further scattering resulting from disorder in ion charges. Using this model, we are also evaluating whether the Bridgman equation for thermal conductivity, which has been used for ionic liquids where scattering lengths are of the order of inter-atomic distances, can be used to estimate thermal conductivity for these materials from their compressibility, density and heat capacity.

Similar calculations will be discussed for related ceramics, including entropic diborides, carbides and nitrides, all of which are being studied experimentally by our research team.

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