Thermal conductivity reduction through increasing number of distinct components in entropy-stabilized oxides

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Reducing the thermal conductivity of crystalline solids while maintaining desired electrical or mechanical properties is a difficult challenge. Such materials hold promise for thermal barrier coatings and thermoelectric applications. Disordered alloys and solid solutions have been particularly promising for such applications, as they allow for reducing thermal conductivity through mass and strain scattering. Such reduction in thermal conductivity is well studied in binary systems like Silicon-Germanium alloys, but there remains a lack of understanding of such reduction for higher order systems. This is primarily due to the instability of such systems such that structural and chemical segregation prevents a uniform material on all length scales. To this end, using time-domain thermoreflectance, we study the thermal conductivity of a new class of materials called entropy-stabilized oxides (ESOs), which are random solid solutions comprised of metallic cations engineered into an oxide such that the cation sublattice consists of 5 to 6 distinct metallic components randomly distributed. In this case, the ESOs are 300 nm thin films consisting of a 5-component system, termed J14 ($Mg_xNi_xCu_xCo_xZn_xO$, x = 0.2) and a 6-component system J30 ($Mg_xNi_xCu_xCo_xZn_xSc_xO$, x = 0.167). For comparison, we also study a two-component $Zn_{0.4}Mg_{0.6}O$ 300 nm sample. We find that the thermal conductivity is greatly reduced with the introduction of more configurational disorder through the addition of distinct metallic cations. Fig. 1 shows the measured thermal conductivity data as a function of temperature, together with the analytical models for MgO, one of the constituents of these samples. The results show that the thermal conductivity can approach the MgO theoretical minimum limit to thermal conductivity at room temperature, suggesting that engineering structure through configurational entropy may be a promising way to reduce the thermal conductivity of crystalline materials.



Fig. 1: Thermal conductivity vs. temperature for of $Zn_{0.4}Mg_{0.6}O$, J14 (Mg_xNi_xCu_xCo_xZn_xO, x = 0.2) and J30 (Mg_xNi_xCu_xCo_xZn_xSc_xO, x = 0.167) together with analytical models for thermal conductivity of bulk MgO, 300 nm MgO, and the minimum limit to thermal conductivity for MgO.