Crystal growth of multicomponent high entropy rare-earth oxides and aluminates

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Compositionally complex (or high entropy) materials are a rapidly growing area. Since the initial report of high-entropy oxides in 2015 [1], many families of compounds containing high concentrations of multiple principal cations residing on one sublattice have been explored. Remarkable structural and functional properties for electronic, magnetic, and thermal applications have been discovered that do not exist in single-component counterparts. Despite the abundance of publications, most of the new compounds are typically synthesized in a polycrystalline form. Single crystals are needed to unlock their full potential and assess properties that cannot be replicated in polycrystalline samples.

In this presentation, we report melt crystal growth of multi-component rare earth (RE) compounds using the micro-pulling-down (m-PD) method: sesquioxides RE_2O_3, aluminates (perovskites $REAlO_3$, garnets $RE_3Al_5O_{12}$, and $RE_4Al_2O_9$), and double perovskites RE'RE"O_3. We also used the Czochralski method to demonstrate scalable growth of selected garnets. We investigated the effect of the size and type of rare earth elements on phase formation and compositional uniformity in the grown crystals. In our composition design, three-to-six rare earths were taken in equiatomic and non-equiatomic ratios to cover a wide range of the average cationic radii. We demonstrated that the correlation between phase formation and average cationic radius can be used a first step towards developing a predictive capability for complex compositions. We also investigated whether congruent melting can be achieved for multi-RE garnets with average cationic radii that are outside the congruency region for single-RE garnets. In addition to phase analysis at room temperature, we used high temperature X-ray and neutron diffraction experiments to investigate structural evolution upon cooling from the melt. We investigated elemental distribution in radial and axial directions using electron probe microanalysis, and morphology and composition of inclusions using scanning electron microscopy and elemental dispersive spectroscopy.

References

[1] Rost CM, et al. Entropy-stabilized oxides. Nat Commun. 2015;6:8485.