HIGH-ENTROPY Li-Na-K FLUORIDE-CHLORIDE MELTS FOR MOLTEN SALT REACTORS

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This report presents a theoretical study that evaluates thermodynamic and thermophysical properties of high-entropy molten salt mixtures composed of lithium, sodium, and potassium fluorides and chlorides. These liquid mixtures of six alkali halide components have enhanced thermodynamic stability as a result of the high configurational entropy as expected [1]. The equimolar solution, $Li_{1/6}Na_{1/6}K_{1/6}F_{1/4}Cl_{1/4}$, maximizes configurational entropy with the value of $\frac{1}{2}R\ln 24 \approx 1.59R = 13.21$ J/mol·K approaches the high-entropy threshold of $R\ln 5$ (13.38 J/mol·K). Such entropy value significantly exceeds that of FLiNaK (6.9 J/mol·K). So they can be promising coolants for molten salt reactors (MSRs) due to improved thermodynamic stability [2]. Two eutectic compositions with reduced lithium content, derived from the phase diagram (melting points 606 °C and 630 °C), are also analyzed [3]. For example, composition on the phase diagram (point B) in which the proportion is LiF (10%) LiCl (10%) NaF (20%) NaCl (20%) KF(20%) KCl (20%) has high entropy as well: $\frac{\Delta S_{\text{conf}}}{R} = \frac{11}{10}\ln 2 + \frac{1}{2}\ln 5 \approx 1.57$.

Density, heat capacity, thermal conductivity, and viscosity are calculated from liquidus to 1000 °C using empirical superposition-extrapolation method, thermodynamic perturbation theory (TPT), as well as molecular dynamics with machine learning interatomic potentials (MLIP).

The diversity of high-entropy coolants offers significant flexibility for tailoring compositions to specific MSR needs, such as fast-spectrum actinide burning or thermal-spectrum efficiency. Through such efforts, high-entropy molten salts could enable MSRs to achieve a closed nuclear fuel cycle, reduce long-lived radioactive waste, and deliver sustainable, carbon-free energy, advancing the goals of Generation IV nuclear technology.

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