



## Investigation of Species Aggregation in $\text{UCl}_3$ - $\text{LiCl}$ - $\text{KCl}$ Molten Salts by Classical Molecular Dynamics

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### Abstract:

Electrorefining of uranium containing fuels may occur via on-line and post-processing methods using chloride molten salts. These high-temperature salts contain a variety of radioactive fission products which complicate both the chemistry and logistics of experimental investigations in such systems. Classical molecular dynamics simulations offer the ability to evaluate nanoscale aggregation tendencies and thermophysical properties of molten salts. This work employs a polarizable Born-Mayer-Huggins potential using CP2K to evaluate the coordination and aggregation behavior of  $\text{U}^{3+}$  in the presence of fission products such as  $\text{Sc}^{3+}$ ,  $\text{Y}^{3+}$ ,  $\text{La}^{3+}$ , and  $\text{Tb}^{3+}$ .

At low trivalent metal concentrations in the  $\text{LiCl}$ - $\text{KCl}$  eutectic, results show a muted influence of fission products on  $\text{U}^{3+}$  aggregation highlighted by increased trivalent metal coordination with  $\text{K}^+$  via shared  $\text{Cl}^-$  in comparison with  $\text{Li}^+$  solvent and  $\text{M}^{3+}$  solute cations. In contrast, higher metal loading concentrations demonstrate a larger fraction of anions being shared between trivalent cations. The magnitude of induced chloride dipoles was also calculated for each system. This data reveals distinct chloride dipole magnitudes in each pure  $\text{MCl}_3$  system; however, lower concentrations of trivalent cations accompany signal dilution of their coordinated chlorides due to sharing of the anions with solvent cations. A comparison of induced chloride dipole magnitudes and the coordination numbers of the  $\text{MCl}_x^{3-x}$  complexes reveals that complex anions of different coordination numbers do not adopt unique anion dipole magnitudes. Findings of this work suggest that fission products will struggle to compete with potassium cations in the solvent for uranium coordination at low solute concentrations; in addition, the magnitude of the induced anion dipole within a complex ion coincides with changes in the cation species and concentration, but not the coordination number of the complex.