



Spectroscopic Signatures of UCl_3 in LiCl/KCl eutectic via AIMD simulations

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Abstract:

Characterization of chloride molten salts with high f-element loading remains limited in literature despite relevance to nonproliferation and online monitoring of molten salt reactors and pyroprocessing separation schemes. Coupled spectroscopic and computational investigations provide certain benefits, as experimental information can validate simulation, while simulations can provide atomistic resolution. *Ab initio* molecular dynamic (AIMD) simulations utilize density functional theory (DFT) based wavefunctions to model UCl_3 in a LiCl-KCl eutectic over increasing uranium concentration (16-80% BW) to determine changes in local coordination environment through applied graph theory. As previously seen in other eutectics, the uranium atom forms a UCl_6 octahedral complex and under low uranium concentrations these complexes are generally isolated from one another. As the concentration of uranium increases, chloride atoms begin bridging octahedral complexes via corner (one chloride), edge (two chlorides), and face (three chlorides) sharing geometries. The Raman and IR spectroscopy of the systems were calculated to determine the relationship between local coordination environment and spectroscopic signatures to determine if there was distinguishing chemical signatures that may be used to identify molten salt composition.