Evaluation of Chemical Speciation to Enable Online Monitoring of Molten Salt Reactors

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Abstract:

Nonproliferation monitoring of next-generation molten salt reactor (MSR) designs requires strict monitoring of fuel salt compositions to ensure safe and secure operation. This project explores online monitoring capabilities for uranium (III) and other lanthanide chloride salts in the KCl-LiCl eutectic via experimental and computational approaches. The experimental portion of this project involved construction of a molten salt furnace and spectrophotometric monitoring system for the eutectic salt. The techniques used to determine the speciation will include Raman spectroscopy, UV-Vis spectroscopy, and electrochemical approaches. The furnace was designed to study the composition of molten salts over a range of temperatures from 650 - 1125 K. Classical molecular dynamics (CMD) simulations of uranium (III) chloride salt have been used to understand structural and kinetic properties of the salt such as coordination number, ion pair distributions, network formation, and diffusion coefficients. A nonpolarizable Born-Mayer-Huggins pair potential with damped dispersion interactions has been employed with the molecular dynamics software, GROMACS. The implementation of this force field marks the start of extending interactions to the UCl_3-LiCl-KCl eutectic and other f-element chloride salts. The CMD trajectories are provided as starting coordinates for *Ab Initio* Molecular Dynamics (AIMD) simulations. The AIMD simulations, done using CP2K, employed the Scalar-relativistic norm-conserving Goedecker–Teter–Hutter medium-core pseudopotentials for uranium and chlorine with 14 $([Xe\ 4f^{14}\ 5d^{10}]\ 6s^2\ 6p^6\ 7s^2\ 5f^3\ 6d^1)$ and 7 $([He]\ 2s^2\ 2p^5)$ valence electrons, respectively, and the molecularly optimized double-zeta basis set (MOLOPT-DZVP-GTH). The IR/Raman spectrum will be calculated from Wannier centers in the trajectory. Collection of Raman spectra on lanthanide solid samples has been initiated. The nonpolarizable CMD force field has been validated against prior experimental and computational results for system coordination, self-diffusion coefficients, and ion pair distributions.