Investigation on multicomponent solubility in eutectic (LiCl-KCl) chloride salts using combinatorial approach

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ABSTRACT:

The molten salt based pyroprocessing technology is the key for treating used nuclear fuels to close nuclear fuel cycle, which was developed by Argonne National Laboratory couple of decades ago. Since then, pyroprocessing has been drawing extensive research effort to develop an engineering scale process and device, which requires fundamental understanding of the thermochemical properties of the molten salts system. Through this process, the waste fission products from the uranium and other heavy radioactive elements in the used fuel are removed by electrorefining, and the remaining uranium and actinides can be recycled. The fission products including alkaline and rare earth metals in the used fuel are dissolved as chlorides and accumulated in the used molten salts. The LiCl–KCl eutectic salts become a multicomponent molten salt system loaded with fission products and actinides from the spent fuel, which could lead to the formation of high melting ternary or higher order compounds with increasing liquidus temperature of the salt medium. As fission element chlorides, especially lanthanide chlorides' concentrations increase and reach certain limit, the operation of electrorefining must be moderated and finally shut down. Therefore, the fission product solubility in molten chlorides salts impacts the timing for salt treatment, recycle, and disposal.

However, most of the available experimental data are limited to the extremely dilute systems, which makes it very challenging to validate the simulation results and the predictive models. Moreover, the solubility data as a function of temperature and composition is lacking for complex multicomponent chloride salt systems, especially those involving multiple lanthanide and actinide elements *due to the laborious work* involved in the solubility measurements. Most available solubility data are for many binary and ternary chloride systems especially those that contain alkali and alkaline earth chlorides

The overall goal of this project is to improve fundamental understanding on multicomponent fission products behavior in **eutectic (LiCl-KCl) chloride salts** for pyroprocessing technology. Four-component chloride salt systems with two of the most representative fission products including $CeCl_3$, $LaCl_3$ and $NdCl_3$ in the eutectic LiCl-KCl salt will be investigated at the temperature range of 723 K to 823 K. The objectives of this proposal are to experimentally establish solubility data as a function of temperature and salt composition and to provide a comprehensive understanding of the solid-liquid equilibria in complex chloride systems by combining thermodynamic assessments of these chloride systems and experimental measurements.