Thermodynamic database development for the NaCl-$MgCl_2$-UCl_3-UCl_4 system

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Recent interest in Molten Salt Reactor (MSR) triggers a wide range of thermochemistry research for the fluoride and chloride salt systems. In particular, the chloride system of the NaCl-$MgCl_2$-UCl_3-UCl_4 will be the key salt system for chloride based MSR process. For the investigation on the stable operation condition and possible safety risk of MSR, the phase diagram information and thermodynamic properties such as vapor pressure of U containing gas species, heat capacity of liquid and solid salts and melting enthalpy depending on the chemical composition of salt are essential.

Since the experimental determination of such key thermochemical information is costly and time-consuming, thermodynamic database based on CALculation of PHAse Diagram (CALPHAD) type assessment is widely accepted in nuclear research in these days for providing such information. As a part of a long-term research project to construct the comprehensive thermodynamic database of multicomponent chloride salt system for MSR, the thermodynamic optimization of the NaCl-$MgCl_2$-UCl_3-UCl_4 system was conducted based on the critical review of all the available thermodynamic and phase diagram data. The Gibbs energy of liquid phase was optimized by using Modified-Quasichemical model (MQM) with appropriate coordination number among cations as analyzing the structural investigation of molten salts by using molecular dynamics simulations. The thermodynamic parameters were optimized to reproduce the experimental data of phase diagram and thermodynamic properties by using Calphad optimizer.

The optimized thermodynamic database can be used to calculate the distribution of U^{3+} and U^{4+} in molten salt depending on oxygen partial pressure. The calculation results can be used to estimate the possible oxidation trend of molten salts in MSR operation condition.