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Standard thermodynamic functions and conditional electrochemical potentials of hypothetical crystalline U(II) and Pu(II) chlorides in LiCl-KCl melt

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Equilibrium electrochemical behavior of (U, Pu)N in LiCl–KCl eutectic melts is of great significance for computer modeling of high-temperature electrochemical reprocessing of fast reactors with nitride fuel and lead coolant (BREST) fuel [1]. We have carried out thermodynamic modeling of pyrochemical reprocessing of (U, Pu)N in LiCl-KCl eutectic melt by using software codes and databases HSC 7.1, OUTOTEC together with our own estimates of thermodynamic data. To estimate the thermodynamic characteristics of U, Pu, Am, as well as of numerous chemical analogs of fission products and to harmonize the latter with the databases in HSC 7.1, we used the well-known comparative physicochemical methods. In this work, we coupled comparative approach with system analysis of chemical properties of considered chemical elements. This allowed us to estimate also the thermodynamic functions for such hypothetical crystalline compounds as UCl2 and PuCl2. The Gibbs free energy of formation of these chlorides in standard conditions is estimated to be (-1196+/-15) and (-1129+/-15) kJ/mole respectively. By comparing the Gibbs free energies for lanthanides, actinides and transition element chlorides with the known conditional equilibrium electrochemical potentials (E*, V, vs. Cl02/Cl-) of the same compounds in LiCl-KCl eutectic melts at 772 K [2], we have estimated E* for the hypothetical M2+/M0 in chloride melt M=U, Pu to be -4.70 and -4.40 v respectively. The reliability and precision of the estimates for UCl2 and PuCl2, as well as the impossibility to detect these species in the chloride melts are discussed in the presentation.

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Reference

[1]. White Book of Nuclear Power. General editing by Prof. E.O. Adamov, Moscow, NIKIET (2001).

[2]. O.V. Skiba, V.A. Kiskii, Yu.P. Savochkin, S.K. Vavilov. Pyroelectrochemical processes in fuel cycle in fast reactors. Dimitrovgrad, NIIAR, 2012. 314 p. (in Russian).

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