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Thermodynamic simulation of the oxidation processes at the reprocessing of spent nuclear fuel in the LiCl-KCl melt

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To date spent nuclear fuel (SNF) reprocessing is a promising field of study. More than 370 thousand tons of SNF have been accumulated in the world and 10-12 thousand tons are added to this amount annually. In Russian Federation, ˜25000 tons of SNF were accumulated according to the data of 2018. Pyrochemical technology of SNF processing, which is supposed to substitute aqueous technologies, is currently developed in several countries.

The aim of the present work is the thermodynamic modeling of the processes that take place at the pyrochemical nitride SNF processing.
Spent nuclear fuel is a complex multicomponent system, which is difficult to study because of the composition complexity and high radioactivity. Methods of thermodynamic modeling are indispensable in the process of evaluation of the SNF properties and development of the SNF treating processes.

In the framework of the present work:
- the material composition of the nitride SNF was calculated. The obtained results are in good agreement with the literature data;
- the oxidation processes of UN and some fission products by cadmium and lead chlorides in the molten LiCl-KCl eutectic were modeled. In particular, the formation of the deposit containing UNCl and non-stoichiometric uranium nitrides is explained. The optimal chlorination temperature is suggested;
-the oxidation process of metallic uranium and some U-noble metals alloys are modeled;
- the amount of missing thermodynamic data is evaluated. In particular, the thermodynamics of nitrides of several rare-earth metals as well as americium and curium nitrides was evaluated.

The modeling was performed using the software HSC Chemistry 9.9.

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