AMERICIUM SEPARATION FROM OTHER MINORACTINIDES FOR ITS RETURN TO THE FUEL CYCLE. THERMODYNAMIC SIMULATION

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In the currently implemented "Breakthrough" program it is proposed to solve the problem of americium produced in reactors by including it in regenerated nitride fuel. In order to close the cycle on americium in this way, it must be completely extracted from the spent nuclear fuel and, especially, purified from curium [1].

In the molten LiCl-KCl eutectic, the Am^{2+}/Am and Cm^{3+}/Cm potentials are so close that it is impossible to say with certainty which of them is more negative and which is more positive. This issue is discussed in detail in [2]. Due to such closeness of potentials, the possibilities of their electrochemical separation are doubtful. The available isolated data on the potential shift when using liquid metal cathodes indicate a rather identical shift of both potentials.

The aim of this work is to find approaches to separating americium from other actinides and plutonium based on the fact that in molten salts the most stable ionic form of americium is Am^{2+}. While for most fissile materials and fission products the formation of divalent ions is not typical.

Various options for separating americium from other minoractinides and plutonium in molten LiCl-KCl eutectic have been studied using thermodynamic modeling methods. They can be divided into two groups. The first group is selective precipitation of americium, and the second group is precipitation of the remaining elements, in which americium remains in soluble form.

Figure 1 shows, as an example, the results of one of the calculations. Initial data. The system consisted of: LiCl – 30; KCl – 20; $PuCl_3$ – 1.5; $AmCl_2$ – 1; $CmCl_3$ – 0.5; Ar – 1.6 kmol. This melt was titrated with lithium oxide, 0…3 kmol Li_2O.

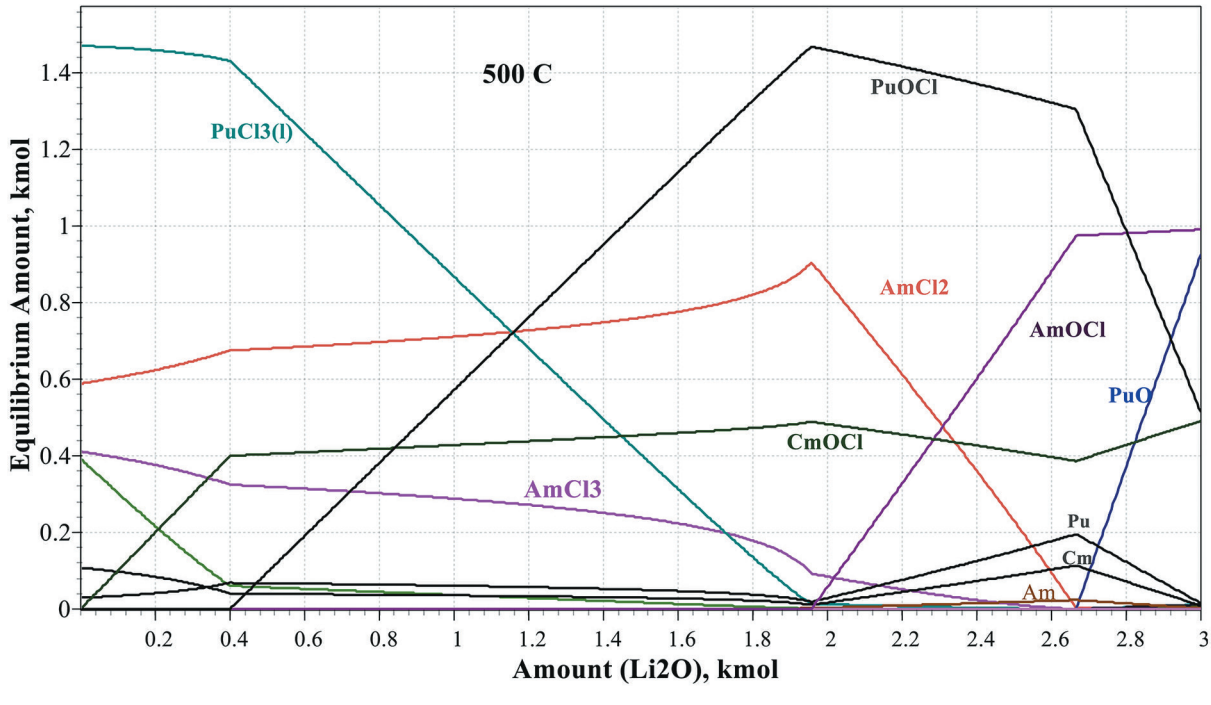

Fig. 1. Equilibrium composition of the mixture LiCl – 30; KCl – 20; $PuCl_3$ – 1.5; $AmCl_2$ – 1; $CmCl_3$ – 0.5; Ar – 1.6; Li_2O 0…3 kmol at 500°C

As can be seen from the figure, when 1.95 kmol Li_2O is added to the system, almost all americium remains in a dissolved state, 90%$AmCl_2$ + 10%$AmCl_3$. Plutonium and curium precipitate as oxychlorides, PuOCl and CmOCl.

If the melt is separated from the sediment, americium can be precipitated as nitride, which can be returned to the fuel cycle:

$$AmCl_2 + Li_3N = AmN + 2LiCl + Li \qquad \Delta G = -330 \text{ kJ/mol}$$

$$AmCl_3 + Li_3N = AmN + 3LiCl \qquad \Delta G = -417 \text{ kJ/mol at } 500°C$$

Conclusions. Based on thermodynamic calculations, approaches to separating americium from plutonium and other minoractinides in the environment of molten LiCl-KCl eutectic are proposed.

References

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