CHEMICAL SEPARATION OF AMERICIUM AND CURIUM IN MOLTEN SALTS. THERMODYNAMIC MODELING

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Americium and curium are completely artificial elements. They are formed in reactors during the irradiation of nuclear fuel. They are highly radioactive, making their separation during spent nuclear fuel reprocessing a very complex problem, as they destroy almost everything—water, ion-exchange resins, and so on. Molten salts exhibit significant radiation resistance. Currently, the molten LiCl-KCl eutectic is most often used as a medium for reprocessing spent nuclear fuel.

In the molten LiCl-KCl eutectic, the most stable form of americium is $AmCl_2$, while curium is always trivalent - $CmCl_3$. Available literary data on their equilibrium potentials are collected in [1]. The potentials are so close (-2.87 ± 0.05 V vs. Cl_2/Cl-) that it is impossible to determine which is more positive and which more negative. The use of active cathodes shifts the deposition potentials of these metals by almost the same amount. Therefore, the feasibility of their electrochemical separation is questionable.

At the same time, the properties of di- and trivalent ions (Am^{2+} and Cm^{3+}) differ significantly. For example, if quartz wool is immersed in a melt of $(LiCl\text{-}KCl)_{eut.}$ + $AmCl_2$ + $CmCl_3$, americium will precipitate as a silicate, while curium will remain in the melt:

$AmCl_2 + 0.5SiO_2 = 0.333AmSiO_3 + 0.667AmCl_3 + 0.167Si$, ΔG = -86 kJ

$CmCl_3 + 1.25SiO_2 = 0.5Cm_2SiO_5 + 0.75SiCl_4(g)$, ΔG = +229 kJ

Instead of SiO_2, other oxides, TiO_2, SnO_2, can also be used:

$SnO_2 + 0.5AmCl_2 + 0.5Zn = 0.5AmSnO_3 + 0.5SnO + 0.5ZnCl_2$, ΔG = - 33.8 kJ

Another approach involves selectively precipitating curium with lithium nitride or oxide, while americium remains in the melt as $AmCl_2$ + $AmCl_3$. Other trivalent ions, U^{3+} and Pu^{3+}, are also precipitated along with curium.

$CmCl_3 + Li_3N = CmN\downarrow + 3LiCl(l)$, ΔG = -430 kJ

$CmCl_3 + 1.5Li_2O = 0.5Cm_2O_3\downarrow + 3LiCl(l)$, ΔG = -234 kJ (500 °C)

Thus, the work proposes a thermodynamic justification for several methods of separating americium and curium in LiCl-KCl melt using the difference in the chemical properties of Am^{2+} and Cm^{3+} ions.

1. Filatov A.A., Vlasov M.I., Potapov A.M., Zaikov Yu.P. Possible electrochemical imitators for $AmCl_2$ and $CmCl_3$ in the molten LiCl–KCl eutectic. Russian Metallurgy (Metally) (2023) № 2, pp. 244-247.