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Thermodynamics and separation factor of uranium from lanthanum on gallium-indium eutectic alloy

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Actinides recycling by separation and transmutation are considered worldwide as one of the most promising strategies for more efficient use of the nuclear fuel as well as for nuclear waste minimization, thus contributing to make nuclear energy sustainable. With this purpose, two major fuel reprocessing technologies have been explored so far to separate the actinides from the fission products arising from nuclear energy production: hydrometallurgical and pyrometallurgical processes.

Fast neutron reactors allow more efficient use of uranium resources. In addition this type of reactors is capable of burning long lived actinides (including those accumulated in thermal reactors spent fuel) thus reducing radioactivity of nuclear wastes. Application of fast reactors results in considerable increase of fuel burn up and, if the reactor is operated as breeder, reproduces fissile materials. At present non-aqueous pyrochemical methods employing molten salts and liquid metals are developed for reprocessing spent nuclear fuels (SNF) of fast reactors. Inorganic melts have very high radiation stability and can be employed for organizing a short closed fuel cycle. Fissile elements and fission products dissolved in a salt melt can be separated employing selective extraction by liquid metals. Detailed information on the properties and behavior of all elements present in SNF in fused salts and liquid metals is required to design a feasible separation process and these include rare earth elements representing an important group of fission products.

Behaviour of lanthanum has so far been studied only in binary systems, Nd-Ga and Nd-In and the behaviour of uranium –only in binary systems, U-Ga [4] and U-In. There is no information on thermodynamic properties of lanthanum and on separation factor of uranium from lanthanum on metallic Ga-In alloys. In the present study the base thermodynamic properties of lanthanum and the separation factor of uranium from lanthanum was determined in Ga-In eutectic alloy.

For calculation of thermodynamic properties of lanthanum and the separation factor of uranium from lanthanum the galvanic cells (1) and (2) were used:

Mo(s) (La}| 3LiCl-2KCl, La(III)|| 3LiCl-2KCl | C(s), Cl2(g) (1)

La (alloy) | 3LiCl-2KCl, La(III)|| 3LiCl-2KCl | C(s), Cl2(g) (2)

Variation of the apparent standard redox potential of the couple La(III)/La as a function of the temperature is: $ELa(III)/La = -(3.731 \pm 0.004) + (8.3 \pm 0.6) \cdot 10 - 4T \pm 0.003 V(3)$

Variation of the apparent standard redox potential of the alloy Me(Ga-In) as a function of the temperature were fitted to the following equation using Software Origin Pro version 7.5.

 $ELa(III)/La(Ga-In) = -(2.906 \pm 0.003) + (5.72 \pm 0.06) \cdot 10-4T \pm 0.002 V(4)$

 $EU(III)/U(Ga-In) = -(2.508 \pm 0.006) + (3.82 \pm 0.11) \cdot 10-4T \pm 0.003 V(5)$

 ${\it The\ activity\ coefficient\ of\ the\ lanthanum\ in\ liquid\ gallium-indium\ alloy\ was\ determined\ by\ equation\ (6):}$

 $log\gamma La(Ga-In) = 3F/2.303RT \cdot (ELa(III)/La - ELa(III)/La(Ga-In))$ (6)

 $\log \gamma \text{La(Ga-In)} = 3.91 - 12496/\text{T} \pm 0.08 (7)$

The expression for calculation of the separation factor (θ) of the metals M1 (uranium) and M2 (lanthanum) on gallium-indium eutectic alloy can be written as:

 $ln\Theta = [(n-m)FE + mFE2 - nFE^{**}1]/RT (8)$

Using the temperature dependences of the apparent standard potentials of lanthanum (4) and uranium (5) in alloys, the following expression for separation factor of uranium and neodymium was obtained:

 $ln\Theta = -6.61 + 13859/T \pm 0.02$ (9)

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