Chlorination route for recovery of actinides from actinide-aluminium alloys*

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Abstract

Pyrochemical processes based on electrochemical separation techniques in molten LiCl-KCl are being developed in ITU for spent nuclear fuel reprocessing. The metallic fuel is anodically dissolved to the melt and actinide ions are group-selectively electrochemically reduced on the cathode, forming solid actinide-aluminium (An-Al) alloys. A chlorination route is under development for recovery of actinides from the obtained alloys. The proposed process consists of three steps: i) vacuum distillation for removal of salt adhered on the electrode; ii) chlorination of remaining An-Al alloy by chlorine gas or HCl gas; iii) sublimation of formed $AlCl_3$ by heating under inert atmosphere. The present work summarises the main results obtained during development of the chlorination route.

Initially, a thermochemical study has been performed, showing thermodynamic feasibility of all three steps. On base of the conditions, proposed by this theoretical study, a systematic experimental series using pure UAl_3 alloy have been carried out to evaluate and optimise the chlorination step. The work has been focused on determination of optimal temperature and Cl_2/UAl_3 molar ratio, with aim to provide full alloy chlorination without formation of volatile UCl_5 and UCl_6. The tested temperatures have been 150, 160 and 170°C and the Cl_2/UAl_3 molar ratio has varied in a range 18-72. The results have shown highly efficient chlorination already at a temperature of 150°C with no uranium volatilisation at all studied Cl_2/UAl_3 molar ratios.

Demonstration experiments on the complete chlorination route have been carried out using U-Pu-Al alloys, which have been prepared by electrochemical deposition of U and Pu onto solid aluminium plates in molten LiCl-KCl. The salt distillation steps have been carried out under vacuum of $4{\cdot}10^{-2}$ mbar at a temperature of 800°C, the chlorination at a temperature of 150°C and molar ratio $Cl_2/UAl_3 = 36$, the sublimation of $AlCl_3$ at a temperature of 400°C under purified argon gas. The results are very promising, showing high efficiency of the salt distillation and indicating also high efficiency of chlorination and sublimation steps. At present, some analyses are ongoing; however the final evaluation of the demonstration experiments will be presented during the meeting.

* The full paper being unavailable at the time of publication, only the abstract is included.