OXIDATION OF AMERICIUM IN NITRIC MEDIA: A MECHANISTICAL STUDY

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Abstract

One of the alternatives selected by CEA for the partitioning of minor actinides from solutions containing fission products is the selective extraction of oxidised americium. This is the SESAME process (Selective Extraction and Separation of Americium by Means of Electrolysis) under development in the ATALANTE facility. This paper presents the study of the mechanism of americium oxidation in nitric media. Oxidised americium is produced though the use of heteropolyanionic ligands such as phosphotungstate or silicotungstate and electrogenerated Ag²⁺.

The first part of this work shows the existence of the lacunary heteropolynion (LHPA) complex with americium (stoechiometry 1:1) whose β_1 and k_2 constants have been estimated. Those results enable us to determine the conditional redox potential of Am(IV)/Am(III) pair in the presence of LHPA ligands and to explain the different behaviour of the two heteropolyanions during electrolysis. The second part of this study concerns the Am(IV)/Am(V) transition. Raman spectroscopy studies would prove that the only oxidable species is the 1:1 Am(IV) complex. According to the previous results, a modelisation of americium electroxidation has then been performed and is in good agreement with experimental results.

Eventually the Am(VI)/Am(V) step was investigated to improve a selective analysis of americium oxidation states. A method has been developed to follow the americium concentration during extraction process.