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Modeling and flowsheet design of an Am separation process using TODGA and H₄TPAEN

V. Vanel*^a, C. Marie^a, P. Kaufholz^b, M. Montuir^a, N. Boubals^a, A. Wilden^b, G. Modolo^b, A. Geist^c, C. Sorel^a

^a CEA Marcoule, Nuclear Energy Division, Radiochemistry & Processes Department, Bagnols-sur-Cèze, F-30207, France

^b Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research, Nuclear Waste Management and Reactor Safety, D-52428, Germany

^c Karlsruher Institut für Technologie (KIT), Institut für Nukleare Entsorgung (INE), Karlsruhe, Germany

Abstract

Recycling americium from spent fuels is an important consideration for the future nuclear fuel cycle, as americium is the main contributor to the long-term radiotoxicity and heat power of the final waste, after separation of uranium and plutonium using the PUREX process. The separation of americium alone from a PUREX raffinate can be achieved by co-extracting lanthanide (Ln(III)) and actinide (An(III)) cations into an organic phase containing the diglycolamide extractant TODGA, and then stripping Am(III) with selectivity towards Cm(III) and lanthanides. The water soluble ligand H₄TPAEN was tested to selectively strip Am from a loaded organic phase.

Based on experimental data obtained by Jülich, NNL and CEA laboratories since 2013, a phenomenological model has been developed to simulate the behavior of americium, curium and lanthanides during their extraction by TODGA and their complexation by H₄TPAEN (complex stoichiometry, extraction and complexation constants, kinetics). The model was gradually implemented in the PAREX code and helped to narrow down the best operating conditions. Thus, the following modifications of initial operating conditions were proposed:

- An increase in the concentration of TPAEN as much as the solubility limit allows.
- An improvement of the lanthanide scrubbing from the americium flow by adding nitrates to the aqueous phase.

A qualification of the model was begun by comparing on the one hand constants determined with the model to those measured experimentally, and on the other hand, simulation results and experimental data on new independent batch experiments.

* Corresponding author. Tel.: +33-4-66-39-78-66.
E-mail address: vincent.vanel@cea.fr

A first sensitivity analysis identified which parameter has the most dominant effect on the process. A flowsheet was proposed for a spiked test in centrifugal contactors performed with a simulated PUREX raffinate with trace amounts of Am and Cm. If the feasibility of the process is confirmed, the results of this test will be used to consolidate the model and to design a flowsheet for a test on a genuine PUREX raffinate. This work is the result of collaborations in the framework of the SACSESS European Project.

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1. Objectives of the EuroEXAm process for the Am stripping step

The EuroEXAm process aims at recovering Am alone from a PUREX raffinate. This process was developed within the SACSESS project in a 3-year collaboration between the CEA, Jülich and NNL. The TODGA extractant at 0.2 M + 5%vol. n-octanol in TPH is used to co-extract trivalent lanthanides and actinides from highly concentrated nitric acid solutions^[1]. Americium (III) is stripped selectively from Cm(III) and Ln(III) into an aqueous phase containing H₄TPAEN at around pH 1. The Am extraction step had already been developed and demonstrated by the Jülich team within the i-SANEX process. The loaded solvent is used as feed solution for the Am stripping step.

This article summarizes the work undertaken to design the flowsheet of the Am stripping step in preparation for demonstration tests at Jülich and ITU:

- Acquisition of batch experimental data for the model,
- Development of the model, flowsheet design and sensitivity analysis.

Nomenclature

TODGA	<i>N,N,N',N'</i> -tetraoctyldiglycoamide
H ₄ TPAEN	<i>N,N,N',N'</i> -tetrakis[(6-carboxypyridin-2-yl)methyl]-ethylenediamine

During the Am extraction step, significant amounts of curium and lanthanides are extracted with americium (more than 99%)^[1]. The challenge of the Am stripping step is to combine the following performances:

- An Am recovery rate as high as possible, typically over 99%.
- A decontamination factor between Am(III) and Cm(III) typically over 500, in order to limit the radiation protection necessary when producing AmO₂ pellets.
- A mass percentage of lanthanides in americium below 5%, as Ln are neutron-absorbing elements negatively impacting reactor operations. With the TODGA-TPAEN chemical system, the light lanthanides, especially lanthanum, are the least separated elements. A DF(Am/La) greater than 200 was suggested.

Therefore, a study in theoretical stages (without any kinetics effect) was performed to have a first estimation of the number of stages necessary to jointly achieve a high Am recovery rate and a sufficient purification from curium and lanthanides. The number of theoretical stages was evaluated depending on:

- The Am recovery rate.
- The separation factor between americium and contaminants (SF(La/Am) ~ 2-2.5, SF(Cm/Am) ~ 5).

To recover more than 99% of americium with a DF(Am/La) around 200, a minimum of between 20 and 30 stages is needed. However for demonstration tests, only 16 stages of centrifugal extractors are available both in Jülich and ITU. It was considered that tests should demonstrate the capacity of the process to decontaminate the Am flow. If this objective was reached, it would be possible to extrapolate the process to recover more americium with a greater number of stages. Thus, for demonstration tests, an Am recovery rate of 60-70% was targeted.

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