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## The SACSESS Hydrometallurgy Domain — an Overview

Andreas Geist<sup>a\*</sup>, Robin Taylor<sup>b</sup>, Christian Ekberg<sup>c</sup>,  
Philippe Guilbaud<sup>d</sup>, Giuseppe Modolo<sup>e</sup>, Stéphane Bourq<sup>d</sup>

<sup>a</sup>Karlsruhe Institute of Technology (KIT), Institute for Nuclear Waste Disposal (INE), Karlsruhe, Germany

<sup>b</sup>National Nuclear Laboratory, Central Laboratory, Sellafield, Seascale, CA20 1PG, United Kingdom

<sup>c</sup>Chalmers University of Technology, Nuclear Chemistry/Industrial Materials Recycling, SE-412 96 Göteborg, Sweden

<sup>d</sup>CEA, Nuclear Energy Division, Marcoule, France

<sup>e</sup>Forschungszentrum Jülich GmbH (FZJ), Institut für Energie- und Klimaforschung (IEK-6), Jülich, Germany

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### Abstract

The EURATOM FP7 project SACSESS (Safety of Actinide Separation Processes) is in continuity of a long line of preceding EURATOM projects. SACSESS is organised along four domains, one of them related to the development of hydrometallurgical (i.e. solvent extraction based) actinide separations processes. Within this domain, the most promising processes developed in previous projects are further developed, improving their technology readiness level (TRL) towards the point at which safe industrial implementation will be achievable.

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### 1. Introduction

The potential benefits of recycling plutonium and minor actinides as nuclear fuel — rather than direct disposal of spent fuel without actinide recycle — are discussed and studied in the context of sustainable nuclear energy production.<sup>1,2</sup> The recycling of actinides requires their prior separation from the used nuclear fuel, i.e. from fission products. Solvent extraction is a suitable separation method.<sup>3</sup> While the separation of plutonium by the PUREX process is well established, separating minor actinides has not been performed on an industrial scale. Solvent extraction processes for separating minor actinides (mainly americium and curium) have been developed in Europe in a continuous series of EURATOM research programmes, initiated in the early 1990s by the late Charles Madic and the late Mike Hudson. In the course of these projects a plethora of new molecules for extracting actinide ions were synthesised and tested. The most suitable of them, together with selected molecules developed by scientists from outside these projects, were used for lab scale process development and testing. Many process flow-sheets were developed and tested on the lab scale. Several were selected as reference flow-sheets, i.e. versions of the so-called r-SANEX,<sup>4,5</sup> i-SANEX,<sup>6</sup> 1c-SANEX,<sup>7</sup> EURO-GANEX<sup>8,9</sup> and CHALMEX<sup>10,11,12</sup> processes. However, many aspects relevant to a safe implementation of such processes have never or only marginally been addressed. Consequently, a focus was needed on filling the identified knowledge gaps. This is the overall aim of the latest EURATOM project, SACSESS (Safety of Actinide Separation Processes)

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\* Corresponding author. Tel.: +49-721-608-26249.  
E-mail address: [andreas.geist@kit.edu](mailto:andreas.geist@kit.edu)

that ran from March 2013 to June 2016. In this way, SACSESS promotes a longer term ambition of raising the technology readiness level (TRL) of the hydrometallurgical processes selected.

Several baseline molecules — hydrophobic extracting agents and hydrophilic complexing agents — used in the above flow-sheets were selected and are studied regarding their behaviour under irradiation, their effect on downstream processes and waste management and the effect of degradation products on process performance. Alternative molecules were identified, that offered potential improvements in performance. Furthermore a multi-scale modelling approach is followed, ranging from the molecular scale via equilibrium and kinetics modelling to flow-sheet calculations.

Addressing a specific gap in options for safe actinide recycling, processes are developed extracting only americium directly from PUREX or COEX™ raffinate solutions but rejecting curium. Such processes have not been studied in earlier EURATOM projects.

Finally, a formal cooperation between the US Department of Energy (DOE) and SACSESS was agreed upon. This cooperation includes joint workshops and mutual access to internationally important facilities such as an irradiation loop and centrifugal contactor equipment.

This paper is an overview of the SACSESS hydrometallurgy domain. For more details please refer to further papers found in the proceedings of the 5<sup>th</sup> International ATALANTE Conference on Nuclear Chemistry for Sustainable Fuel Cycles, published in *Procedia Chemistry*.

## 2. The molecules studied

The hydrophobic extracting agents and hydrophilic complexing agents used in the reference flow-sheets are shown in Figure 1.

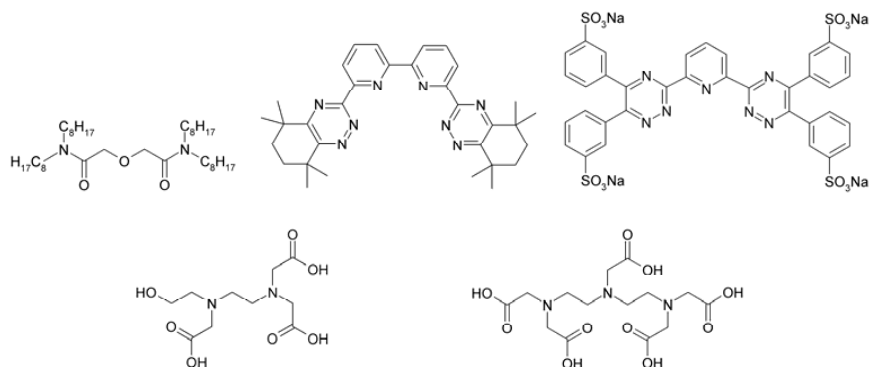


Fig. 1. SACSESS reference compounds: TODGA, CyMe<sub>4</sub>-BTBP, SO<sub>3</sub>-Ph-BTP (top, from left), HEDTA, DTPA (bottom, from left).

*N,N,N',N'*-tetra-*n*-octyl diglycolamide (TODGA) is used in i-SANEX,<sup>6</sup> EURO-GANEX<sup>8,9</sup> and Am-only extraction processes to co-extract actinides and lanthanides from high-acidity raffinate solutions, separating them from the non-lanthanide fission products. TODGA is furthermore added to r-SANEX<sup>5</sup> and 1c-SANEX<sup>7</sup> solvents to accelerate the rather slow extraction kinetics of CyMe<sub>4</sub>-BTBP.<sup>13</sup> As a possible alternative to TODGA, Me-TODGA (Figure 2) is studied. Being a weaker extracting agent, the unwanted co-extraction of strontium is less pronounced than with TODGA.<sup>14</sup>

6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-2,2'-bipyridine (CyMe<sub>4</sub>-BTBP) is the extracting agent used in r-SANEX,<sup>4,5</sup> 1c-SANEX<sup>7</sup> and CHALMEX<sup>10,11,12</sup> processes. It extracts actinides selectively over lanthanides and many other fission products. Bis-2,6-(5,6,7,8-tetrahydro-5,9,9-trimethyl-5,8-methano-1,2,4-benzotriazin-3-yl)pyridine (CA-BTP)<sup>15</sup> and 2,9-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-1,10-phenanthroline (CyMe<sub>4</sub>-BTPhen)<sup>16</sup> (Figure 2) are alternatives to CyMe<sub>4</sub>-BTBP, having faster extraction kinetics. However, issues with the scale-up of the syntheses have so far kept these compounds from superseding CyMe<sub>4</sub>-BTBP.

*N*-(2-hydroxyethyl)ethylenediaminetriacetic acid (HEDTA) and diethylenetriaminepentaacetic acid (DTPA) (Figure 1) are used to strip actinides selectively over lanthanides from an organic phase containing both actinides and lanthanides. This TALSPEAK-like chemistry is applied in processes such as the i-SANEX<sup>17</sup> and GANEX<sup>18</sup> processes developed at CEA.

2,6-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)pyridine (SO<sub>3</sub>-Ph-BTP,<sup>19</sup> Figure 1) was developed to overcome some of the drawbacks of HEDTA and DTPA, such as the narrow pH window they are effective in. It is used for actinide stripping in an i-SANEX process<sup>6</sup> and in the EURO-GANEX process.<sup>8,9</sup> 2,6-bis(1-(3-hydroxypropyl)-1H-1,2,3-triazol-4-yl)pyridine (PyTri-diol, Figure 2) can be used for such processes as well. Having the advantage of being a CHON compound, it avoids the secondary waste generation caused by the sulphur content of SO<sub>3</sub>-Ph-BTP.

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