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# Towards a new GANEX 2nd cycle process for the co-separation of TRU

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*Keywords:* GANEX process, TRU separation, TOGDA, DMDOHEMA, AHA, BTP

**Abstract**

We report first results on the development of a new system for the GANEX 2nd cycle: A solvent consisting of *N,N,N',N'*-tetra-*n*-octyl-diglycolamide (TODGA) + *N,N'*-dimethyl-*N,N'*-dioctyl-2-(2-hexyloxyethyl)malonamide (DMDOHEMA) in kerosene was developed which co-extracts TRU and lanthanides with high distribution ratios and allows for Pu(IV) loading up to 35 g/L. Also the behaviour of non-lanthanide fission products was studied. A substitute for oxalic acid (used as Zr masking agent in DIAMEX processes but not useful in the presence of Pu(IV)) had to be found: CDTA efficiently suppresses the extraction of Zr and also masks Pd which would otherwise be co-extracted. In the selective TRU strip section actinides are back-extracted into an aqueous solution containing a hydrophilic 2,6-bis(1,2,4-triazin-3-yl)pyridine (BTP) and acetohydroxamic acid (AHA) in nitric acid. Lanthanides are kept in the organic phase by a sufficiently high nitric acid concentration; no salting out or buffering agents are required.

## Introduction

Separation of long lived radionuclides from irradiated nuclear fuel through partitioning and transmutation (P&T) provides potential advantages over long term storage of once through spent fuel. Several hydrometallurgical processes are being considered in Europe and involve the PUREX process to remove U and Pu then routing the raffinate through a minor actinides and lanthanides co-extraction process (DIAMEX) [1] followed by An(III)/Ln(III) separation (SANEX) [2].

To avoid pure Pu product streams, alternative processes are being developed that are capable of handling all the transuranium elements (TRU = Np, Pu, Am, Cm) in one stream throughout the process. This requires a solvent capable of extracting all TRU and being able to cope with high Pu loading (up to 50 g/L, to cope with fast reactor fuels). Also, all TRU must be selectively back extracted from the loaded solvent in one section.

To this, the GANEX process (Group ActiNide EXtraction) was developed by the French CEA for the homogenous recycling of actinides [3, 4]. After the selective extraction of uranium(VI) from the dissolution solution in the 1st cycle [5], TRU are co-separated in the 2nd cycle [6]. Successful hot tests of the 1st and 2nd cycles were performed. The 2nd cycle solvent is composed of a neutral extracting agent (DMDOHEMA) and an acidic one (HDEHP, di(2-ethylhexyl)phosphoric acid), which may complicate the solvent regeneration. Also, HEDTA is used for stripping TRU, which works only in a narrow pH region, requiring a buffer.

In the framework of the European ACSEPT project [7] an alternative GANEX 2nd cycle is developed. First step was developing a solvent capable of co-extracting TRU at relevant concentrations. Next step was developing a system for co-stripping the TRU into one product solution. This paper reports some of the most important results resulting from a significant work programme shared between several labs involved in the ACSEPT project.

## Solvent formulation

Initially developed in Japan [8], the tridentate ligand TODGA *(N,N N',N'*-tetraoctyl diglycolamide) has shown promise for extracting actinides(III, IV, VI) and is found to have positive radiolytic and hydrolytic stability properties [9, 10]. In kerosene diluents, TODGA is prone to 3rd phase formation, although additives such as TBP [10], 1-octanol [11] and amines [12] can serve to suppress 3rd phase formation. Indeed, a number of active DIAMEX-style flowsheet trials have been performed on 0.2 mol/L TODGA and 0.5 mol/L TBP, successfully demonstrating the extraction of An(III) [13, 14].

Building on previous work on the TODGA/TBP system [15], which showed good heavy metal loading capacity with uranium, we have explored how solvent systems formulated around DGA extractants behave in

contact with high aqueous phase plutonium concentrations. Unfortunately, solvents composed of 0.2 mol/L TODGA + TPB and/or 1-octanol in kerosene had a limiting initial aqueous Pu(IV) concentration of no more than 5 g/L.

Several solvents based on diglycolamides, DMDOHEMA, HDEHP, DHOA *(N,N*-dihexyloctanamide), TBP were screened for Pu(IV) loading. Following extensive testing, a solvent composed of 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80 was selected; it can handle initial aqueous Pu(IV) concentrations of up to 40 g/L in 3 mol/L $HNO_3$.

### $HNO_3$ Extraction

Both TODGA and DMDOHEMA extract $HNO_3$. Knowledge of $HNO_3$ extraction is important for calculating a process flowsheet. Hence, this issue was studied to determine whether the amount of $HNO_3$ extracted by the TODGA + DMDOHEMA solvent is equal to the sum of $HNO_3$ extracted by TODGA and DMDOHEMA.

Experimental data (Figure 1) on the extraction of $HNO_3$ into 0.5 mol/L DMDOHEMA in Exxsol D80 or 0.2 mol/L TODGA in Exxsol D80 (blue triangles) was fit by polynomials (blue dashed lines). The sum of these polynomials (blue solid line) is compared to the extraction of $HNO_3$ into the TODGA + DMDOHEMA solvent (blue circles). A similar comparison was made with previous data for DMDOHEMA [16] and TODGA [11] (red symbols, red lines).

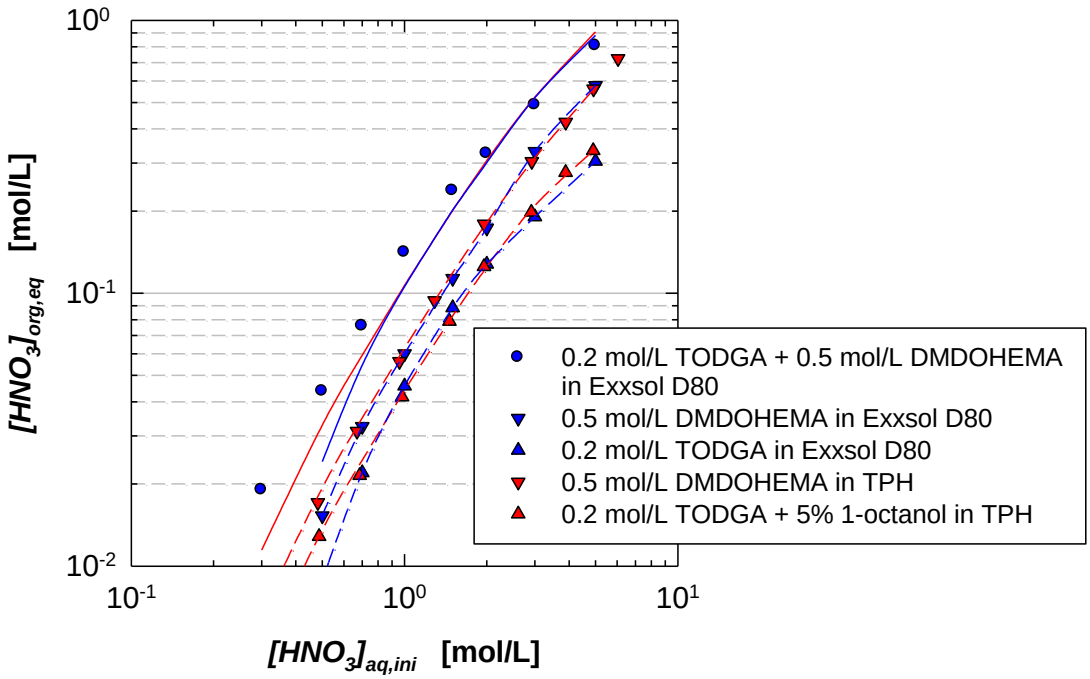


**Figure 1. $HNO_3$ extraction. Organic equilibrium $HNO_3$ concentration as a function of initial aqueous $HNO_3$ concentration.**
Organic phase, as indicated. Aqueous phase, $HNO_3$. *A/O* = 1; *T* = (293 ± 0.5) K.

For 2 mol/L – 5 mol/L $HNO_3$, $HNO_3$ extraction into the TODGA + DMDOHEMA solvent is in agreement with the sum of $HNO_3$ extraction by TODGA + DMDOHEMA. For lower $HNO_3$ concentrations this is not the case; the TODGA + DMDOHEMA solvent extracts more $HNO_3$ as to be expected from adding $HNO_3$ extraction by TODGA and DMDOHEMA. However, for lower aqueous $HNO_3$ concentrations the absolute deviations are only in the range of 20 mmol/L.

### TRU + Ln(III) co-extraction

Data for the extraction of Pu(IV), Am(III) and Ln(III) from $HNO_3$ are shown in Figure 2 as a function of $[HNO_3]$. Similar to TODGA alone, the TODGA + DMDOHEMA solvent co-extracts An(III) and Ln(III) with high distribution ratios. Lighter Ln(III) are less extracted than heavier ones. Pu(IV) extraction is similar to that of Am(III).

The extraction of Np was studied independently using Np(IV), Np(V), and Np(VI) in $HNO_3$ of varied concentration, see Figure 3. Np(IV) is well extracted with distribution ratios slightly lower than those for Pu(IV). Np(V) is poorly extracted; a maximum distribution ratio of 3 is achieved when extracting from 4 mol/L $HNO_3$. Finally, Np(VI) is well extracted, however with lower distribution ratios than Np(IV).

In summary, all TRU are well extracted into the TODGA + DMDOHEMA solvent at acidities relevant to a GANEX 2nd cycle process, i.e., 3–4 mol/L $HNO_3$.

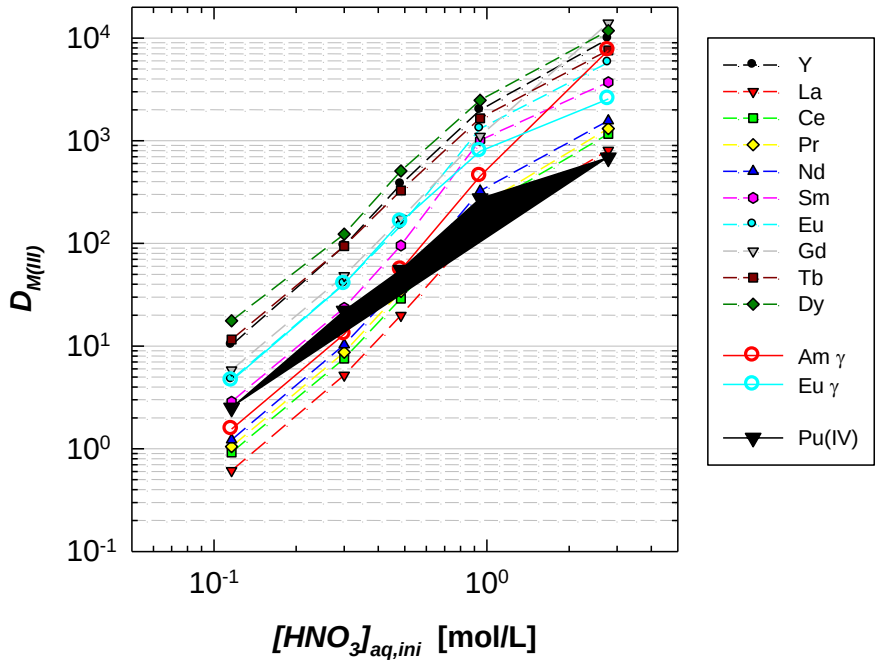


**Figure 2. Pu(IV) + An(III) + Ln(III) extraction, distribution ratios as a function of *[HNO₃]*.**
Organic phase, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80. Aqueous phase, $^{242}$Pu(IV) ($10^{-6}$ mol/L) + $^{241}$Am(III) + $^{152}$Eu(III) (1 kBq/mL each) + Ln(III) ($10^{-4}$ mol/L each) in $HNO_3$. *A/O* = 1; *T* = (293 ± 0.5) K.

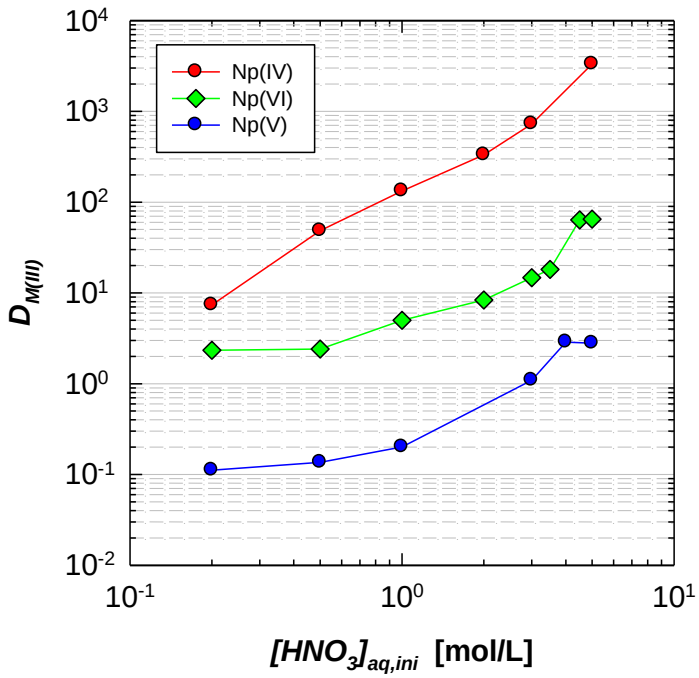


**Figure 3. Extraction of Np(IV), Np(V), and Np(VI), distribution ratios as a function of *[HNO₃]*.**
Organic phase, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80. Aqueous phase, $^{237}$Np(IV) or $^{237}$Np(V) or $^{237}$Np(VI) ($10^{-6}$ mol/L) in $HNO_3$. *A/O* = 1; *T* = (293 ± 0.5) K.

## Fission product extraction and masking

Although TODGA and DMDOHEMA reject most non-lanthanide fission products, they do extract Zr, Mo, and Pd. Hence, the extraction of all fission products into the TODGA + DMDOHEMA solvent was studied. Also, a substitute for oxalic acid (used as Zr(IV) masking agent in DIAMEX processes [17] but not usable in the presence of Pu(IV)) had to be found.

Following extensive screening tests [18], CDTA was found to efficiently suppress the extraction of Zr(IV) and Pd(II) while not influencing the extraction of Pu(IV), Am(III) and Ln(III). Distribution ratios for the extraction of

fission products from 3 mol/L $HNO_3$ without or with 0.05 mol/L CDTA are shown in Table 1. The effect of CDTA on the extraction of Zr and Mo from HNO3 of varied concentration is shown in Table 2.

**Table 1. Effect of CDTA on the extraction of fission and corrosion products.**
Organic phase, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80.
Aqueous phase, HAR elements in 3 mol/L $HNO_3$. *A/O* = 1; *T* = (295 ± 0.5) K.

| Element | Distribution ratios; *[CDTA]* = 0 | 0.05 mol/L |
|---|---|---|
| Ln | >100 | >100 |
| Zr | >100 | ≤0.01 |
| Pd | 4.20 | 0.07 |
| Mo | 1.93 | 2.10 |
| Sr | 1.43 | 2.25 |
| Se | 0.77 | 0.74 |
| Fe | 0.25 | 0.25 |
| Cd | 0.25 | 0.17 |
| Ru | 0.23 | 0.32 |
| Ba | 0.13 | 0.20 |
| Cr, Ni, Rb, Rh, Sn, Sb, Te, Cs | ≤0.02 | ≤0.02 |

**Table 2. Effect of CDTA on the extraction of Zr and Mo at varied *[$HNO_3$].***
Organic phase, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80.
Aqueous phase, HAR elements in $HNO_3$. *A/O* = 1; *T* = (295 ± 0.5) K.

| Element | Masking agent | Distribution ratios; *[$HNO_3$]* = 1 mol/L | 2 mol/L | 3 mol/L | 4 mol/L |
|---|---|---|---|---|---|
| Zr | – | 30 | 86 | 142 | 376 |
| | CDTA | ≤0.01 | ≤0.01 | ≤0.01 | 0.02 |
| Mo | – | 1.21 | 1.36 | 1.93 | 3.62 |
| | CDTA | 1.43 | 1.08 | 2.10 | 4.12 |

The behaviour of CDTA at nominal Pu(IV) concentrations (17 g/L) was also investigated. No precipitation was observed. CDTA suppresses the extraction of Zr(IV) and Pd(II) even in the presence of 17 g/L Pu(IV); results are shown in Table 3.

**Table 3. Effect of CDTA in the presence of nominal Pu(IV) concentration.**
Organic phase, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80. Aqueous phase,
3 mol/L $HNO_3$ + $^{239}$Pu (17 g/L) + $^{241}$Am(III) (0.1 g/L) + HAR elements + CDTA. *A/O* = 1; *T* = 293 K.

| Element | Distribution ratios; *[CDTA]* = 0.01 mol/L | 0.05 mol/L | 0.1 mol/L |
|---|---|---|---|
| $^{241}$Am(III) | 65 | 42 | 40 |
| $^{239}$Pu(IV) | 60 | 35 | 32 |
| Zr(IV) | 7.7 | 1.2 | 0.2 |
| Pd(II) | 0.5 | 0.05 | 0.02 |

## TRU stripping

The initially adopted strategy for the selective TRU back-extraction is using a solution containing a hydrophilic 2,6-bis(1,2,4-triazin-3-yl)pyridine (BTP) [19, 20] and acetohydroxamic acid (AHA) [21] in nitric acid. The former strips Am(III) and Cm(III), while the latter strips Pu(IV) and Np. Lanthanides are kept in the organic phase by a sufficiently high nitric acid concentration; no salting out or buffering agents are required.

*Np + Pu(IV) stripping with AHA*

When stripping an organic phase loaded with Pu(IV) from 0.5 mol/L $HNO_3$ into solutions of AHA in nitric acid, the following results are obtained: Stripping into a solution of 0.5 mol/L AHA in 0.2 mol/L $HNO_3$ yields a Pu(IV) distribution ratio of 0.55; when the acidity in the stripping solution is reduced to 0.1 mol/L $HNO_3$, the Pu(IV) distribution ratio is 0.14.

When stripping Np into a solution of 0.5 mol/L AHA in 0.2 mol/L $HNO_3$, a Np distribution ratio of approx. 0.4 is achieved. At reasonably low nitric acid concentration (ca. 0.1 mol/L), Np in all oxidation states is stripped with a distribution ratio of approx. 0.1.

*Am(III) + Cm(III) stripping with $SO_3$-Ph-BTP*

Tests analogous to those reported in [19, 20] show that $SO_3$-Ph-BTP can be used for stripping Am(III) + Cm(III) from the TODGA + DMDOHEMA solvent, while Ln(III) are kept in the aqueous phase by a sufficiently high $HNO_3$ concentration of approx. 0.5 mol/L. Am(III) and Cm(III) have distribution ratios which are practically identical. Results are not shown but Am(III) and Ln(III) distribution ratios for similar experimental conditions are presented below in Figure 4.

*Pu(IV) + Am(III) stripping with $SO_3$-Ph-BTP*

Hydrophobic BTP-type extracting agents extract Pu(IV) from $HNO_3$ with distribution ratios similar to those found for Am(III) [22]. This is why it was tested whether $SO_3$-Ph-BTP would also suppress the extraction of Pu(IV) thus acting as a stripping agent for Pu(IV).

Figure 4 shows data for the separation of Pu(IV) + Am(III) from Ln(III) at varied $HNO_3$ concentration. $SO_3$-Ph-BTP suppresses the extraction of both Pu(IV) and Am(III). Pu(IV) distribution ratios are higher than for Am(III) approx. by a factor of 2. The results indicate that an efficient separation of (Pu(IV) + Am(III)) from Ln(III) is viable for 0.3 mol/L < $[HNO_3]$ < 0.7 mol/L.

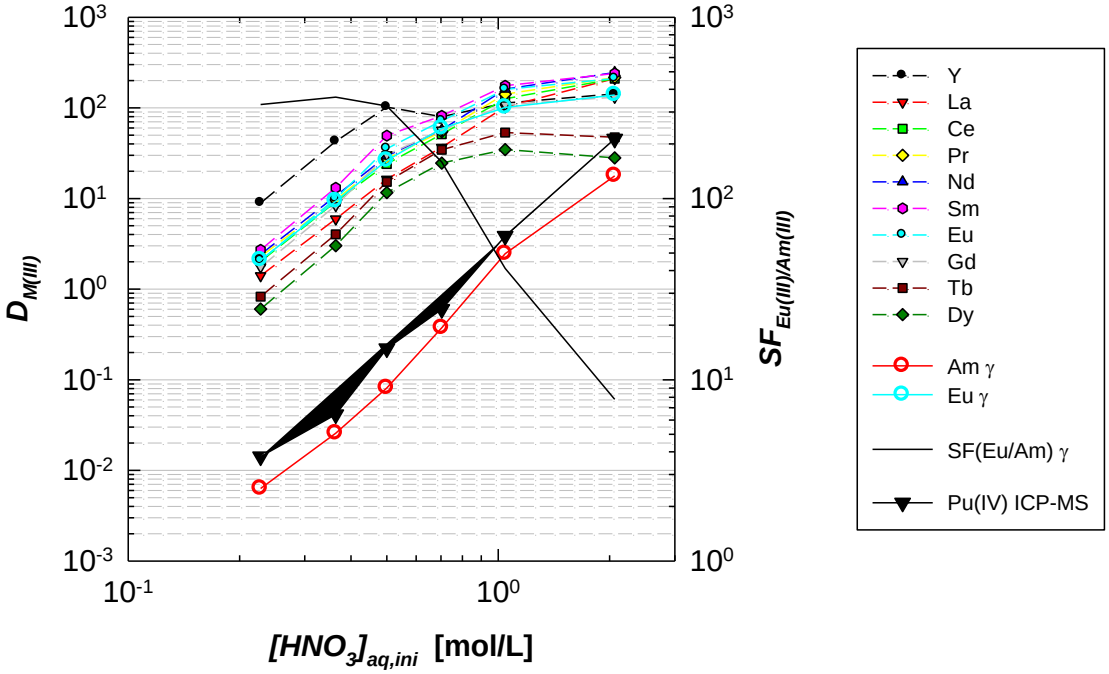


**Figure 4. Actinide stripping by $SO_3$-Ph-BTP, distribution ratios as a function of $[HNO_3]$.**
Organic, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80. Aqueous, $^{242}$Pu(IV) ($10^{-6}$ mol/L) + $^{241}$Am(III) + $^{152}$Eu(III) (1 kBq/mL each) + Ln(III) ($10^{-4}$ mol/L each) + 20 mmol/L $SO_3$-Ph-BTP in $HNO_3$. *A/O* = 1; *T* = (293 ± 0.5) K.

The influence of $SO_3$-Ph-BTP concentration on Pu(IV), Am(III) and Ln(III) distribution ratios is shown in Figure 5. With increasing $[SO_3\text{-}Ph\text{-}BTP]$, Pu(IV) and Am(III) distribution ratios are decreasing with a slope of approx. −2. Also, $SF_{Eu(III)/Am(III)}$ increases from 170 to 460 when increasing $[SO_3\text{-}Ph\text{-}BTP]$. $SF_{Pu(IV)/Am(III)}$ remains fairly constant at approx. 2.

*Pu(IV) + Am(III) stripping with $SO_3$-Ph-BTP + AHA*

Using a mixture of 20 mmol/L $SO_3$-Ph-BTP + AHA (0–3 mol/L) for suppressing actinides extraction is shown in Figure 6. Adding AHA has only a slight effect on Pu(IV) distribution ratios; obviously $SO_3$-Ph-BTP is a stronger complexing agent for Pu(IV) than is AHA. Ln(III) distribution ratios determined by ICP-MS show a slightly decreasing trend with increasing AHA concentration, which is not in agreement with the $^{152}$Eu gamma measurements. Thus the slight decrease of Pu(IV) distribution ratios (which were also determined by ICP-MS) may be due to analytical uncertainties.

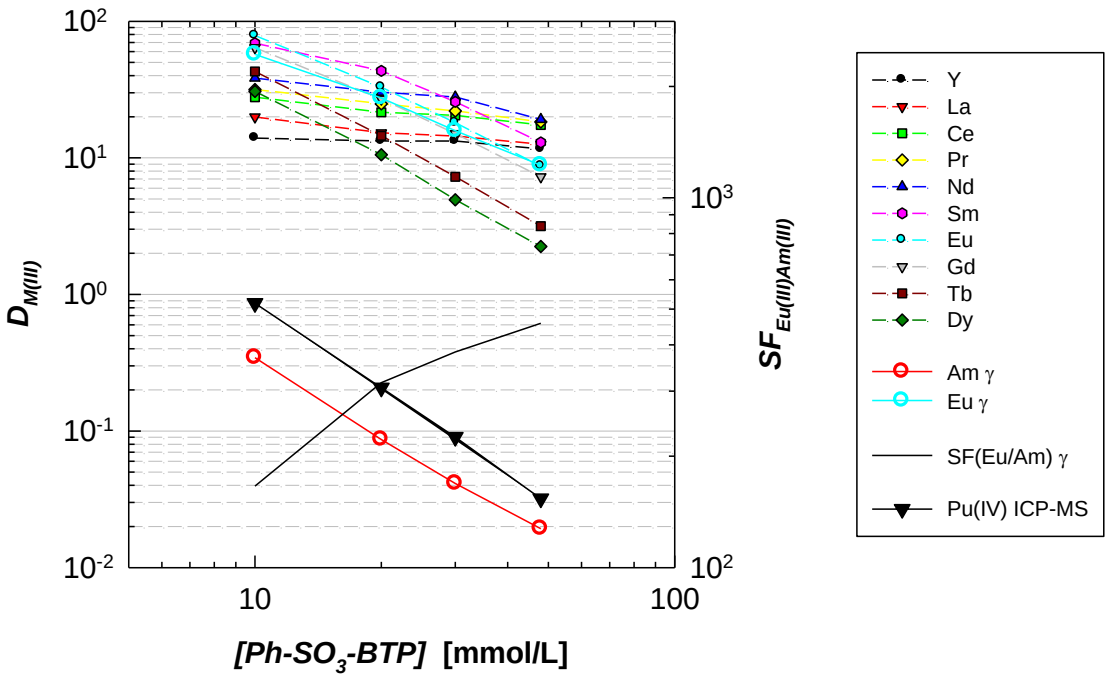


**Figure 5. Actinide stripping by $SO_3$-Ph-BTP, distribution ratios as a function of *[$SO_3$-Ph-BTP]*.**
Organic, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80. Aqueous, $^{242}$Pu(IV) ($10^{-6}$ mol/L) + $^{241}$Am(III) + $^{152}$Eu(III) (1 kBq/mL each) + Ln(III) ($10^{-4}$ mol/L each) + $SO_3$-Ph-BTP in 0.5 mol/L $HNO_3$. *A/O* = 1; *T* = (293 ± 0.5) K.

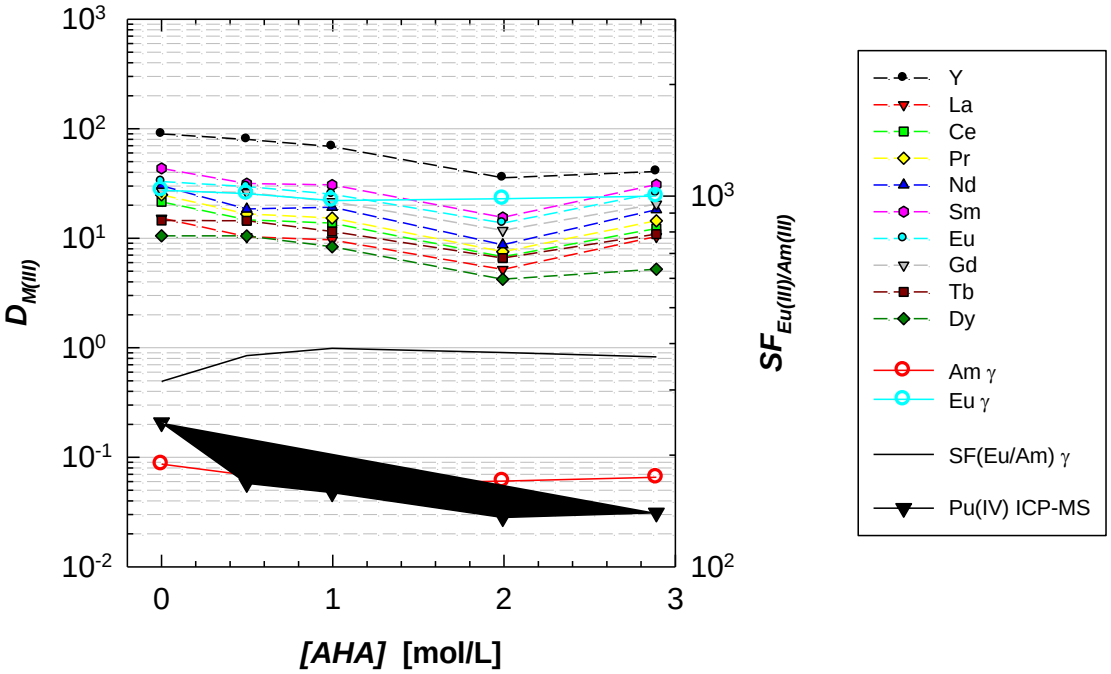


**Figure 6. Actinide stripping by $SO_3$-Ph-BTP + AHA, distribution ratios as a function of *[AHA]*.**
Organic, 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80. Aqueous, $^{242}$Pu(IV) ($10^{-6}$ mol/L) + $^{241}$Am(III) + $^{152}$Eu(III) (1 kBq/mL each) + Ln(III) + 20 mmol/L $SO_3$-Ph-BTP + AHA in 0.5 mol/L $HNO_3$. *T* = (293 ± 0.5) K.

## Conclusion and Outlook

In the framework of the European ACSEPT project a GANEX 2$^{nd}$ cycle process for the co-separation of all TRU is under development. First step was the successful development of a solvent capable of co-extracting all TRU in relevant concentrations, most notably up to several 10 g/L Pu(IV). A solvent consisting of 0.2 mol/L TODGA + 0.5 mol/L DMDOHEMA in Exxsol D80 was developed for this task, extracting up to 40 g/L Pu(IV) without precipitation or 3$^{rd}$ phase formation. The undesired co-extraction of some fission products such as Zr(IV) and Pd(II) could be suppresses by adding CDTA as a masking agent.

Since lanthanides(III) are co-extracted by this solvent, a selective TRU stripping system had to be developed. Initially it was planned to use a solution containing AHA (to strip Pu(IV) + Np) and $SO_3$-Ph-BTP

(to strip Am(III) and Cm(III)) in $HNO_3$ (to keep lanthanides(III) in the organic phase). Since AHA does not efficiently strip Pu(IV) + Np at $HNO_3$ concentrations sufficient to keep lanthanides(III) from also being stripped by $SO_3$-Ph-BTP, a different TRU stripping strategy is currently being developed:

$SO_3$-Ph-BTP at a sufficiently high concentration could be used to strip Pu(IV), Am(III) and Cm(III). Np stripping will most reasonably be achieved by reducing to Np(IV)using AHA. Np(IV) is expected to be stripped by $SO_3$-Ph-BTP similarly to Pu(IV).

These tests will lead to the development of a GANEX 2$^{nd}$ cycle flow-sheet first to be tested on a surrogate feed solution containing approx. 10 g/L Pu(IV). Final goal is a hot counter-current test to be performed with a genuine spent fuel feed solution.

**Acknowledgement**

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