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optimized flowsheets for heterogeneous recycling of advanced MOX fuel in GEN IV reactors

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Summary

The Deliverable Report, D6.3, Optimized flowsheets for heterogeneous recycling of advanced MOX fuel in GEN IV reactors, compiles optimization studies towards the development of separation processes for the heterogeneous recycling of actinides performed within the GENIORS project. ? i-SANEX process with PTD ? ALSEP process ? i-SANEX process with SO3-Ph-BTP

Approval

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INTRODUCTION

The Deliverable Report, *D6.3, Optimized flowsheets for heterogeneous recycling of advanced MOX fuel in GEN IV reactors,* compiles optimization studies towards the development of separation processes for the heterogeneous recycling of actinides performed within the GENIORS project.

- i-SANEX process with PTD
- ALSEP process
- i-SANEX process with SO_3-Ph-BTP

I-SANEX PROCESS WITH PTD

A counter-current i-SANEX process was demonstrated using PyTri-Diol (PTD, Figure 1) for the An(III) selective stripping. The solvent was the same as tested in the first i-SANEX process, 0.2 mol/L TODGA (Figure 1) + 5%$_{vol.}$ 1-octanol in TPH.[1] The CHON compliant PTD was shown to be a promising replacement for the non-CHON compliant SO_3-Ph-BTP.[2-4] CDTA was used to mask Zr and Pd extraction in the feed.[5] Chemical conditions for the An(III) stripping section were derived from single stage data.[3] Optimised conditions for An(III) stripping were 0.08 mol/L PTD in 0.45 mol/L HNO_3. Flow-sheet calculation were conducted using the SX Process software.[6]

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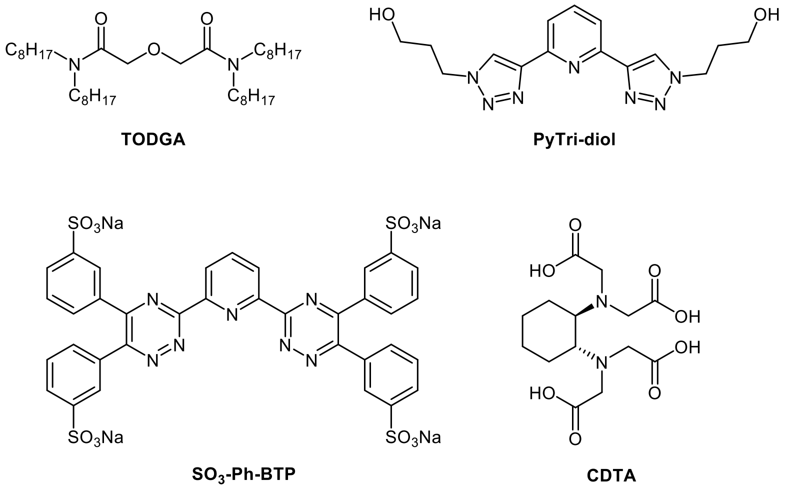

Figure 1. Chemical structures of TODGA, PyTri-diol, SO_3-Ph-BTP, and CDTA.

A 16-stage flow-sheet was calculated for the An(III) selective stripping and Ln(III) re-extraction sections. Due to the limited number of available contactors in our laboratory, only the An(III) selective stripping and Ln(III) re-extraction sections were tested. As the extraction and scrubbing sections of the PTD i-SANEX test were identical to the previous i-SANEX test[1] and did not involve the use of the new hydrophilic stripping agents, it was decided to prepare the loaded organic phase by a series of batch contacts. Analyses of the loaded organic phase showed a composition well comparable to the original i-SANEX test. Similarly, the Ln(III) stripping was not tested, as it was demonstrated successfully in the previous i-SANEX test.[1] Figure 2 shows the series of batch contacts for the preparation of the loaded organic phase and the flowsheet of the i-SANEX test.

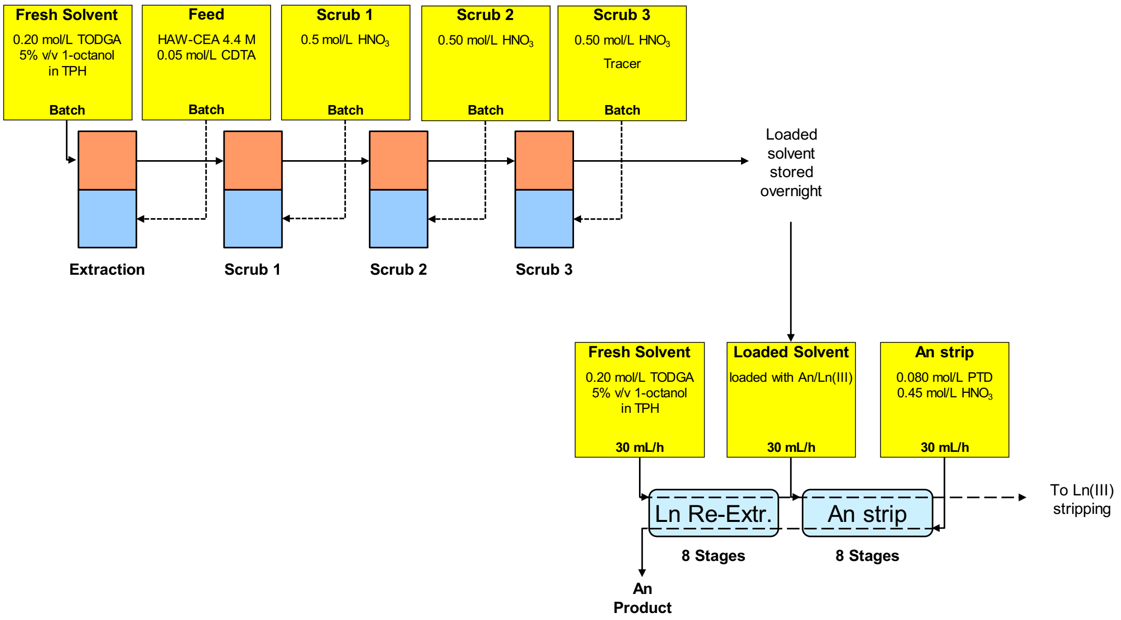

Figure 2. Flowsheet of the PTD i-SANEX test. Scrub 3 solution contains ^{241}Am(III) (5.6 MBq/L), ^{244}Cm(III) (5.6 MBq/L), ^{152}Eu(III) (10 MBq/L).

Figure 3 shows Am and Eu stage profiles of the PTD i-SANEX demonstration test. Very good An(III)/Ln(III) selectivity were obtained, and the demonstration test is believed to successfully show that PTD is a good candidate to replace SO_3-Ph-BTP in the i-SANEX process. Probably PTD can also be used as a replacement in other processes, e. g. the EURO-GANEX process. A full paper with full evaluation of the analytical data obtained from the centrifugal contactor test is in preparation and will soon be submitted for publication.

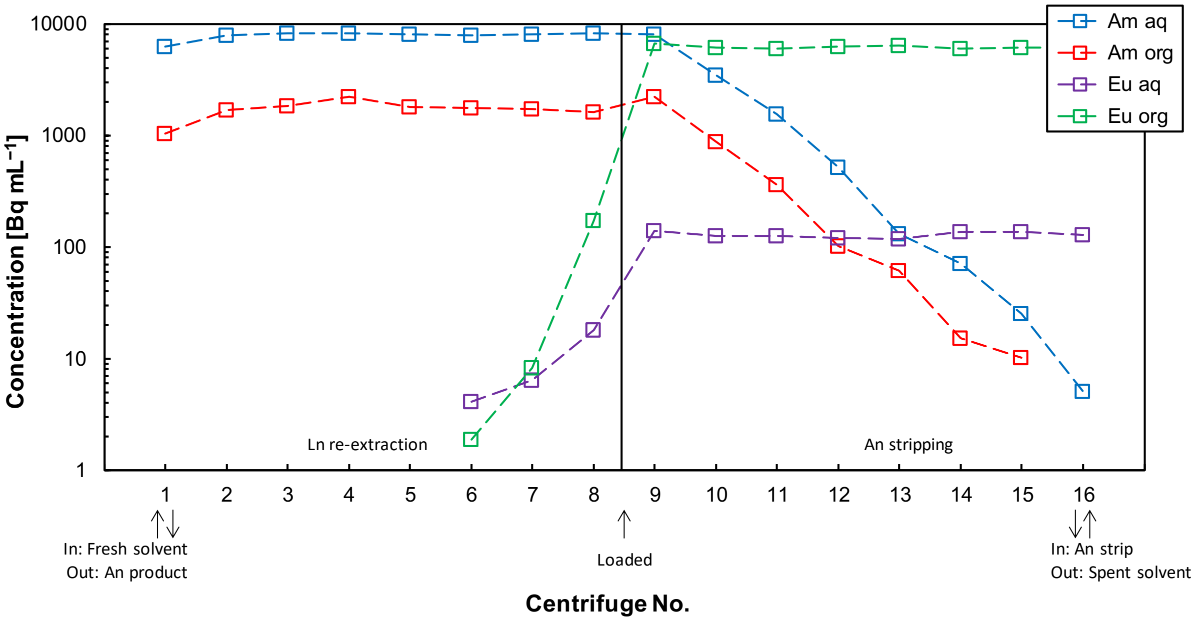

Figure 3. Am and Eu stage profiles of the PTD i-SANEX demonstration test (data from gamma spectroscopy).

ALSEP PROCESS

Within the GENIORS collaboration with the US DOE, an Actinide Lanthanide Separation Process (ALSEP) was demonstrated on the laboratory scale using the centrifugal contactors installed in the JUELICH laboratories. The results were published in Appl. Sci.[7] The abstract from this paper is cited below:

"An Actinide Lanthanide Separation Process (ALSEP) for the separation of trivalent actinides (An(III)) from simulated raffinate solution was successfully demonstrated using a 32-stage 1 cm annular centrifugal contactor setup. The ALSEP solvent was composed of a mixture of 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester (HEH[EHP]) and N,N,N',N'-tetra-(2-ethylhexyl)-diglycolamide (T2EHDGA) in n-dodecane. Flowsheet calculations and evaluation of the results were done using the Argonne's Model for Universal Solvent Extraction (AMUSE) code using single stage distribution data. The co-extraction of Zr(IV) and Pd(II) was prevented using CDTA (trans-1,2-diaminocyclohexane-N,N,N',N'-tetraacetic acid) as a masking agent in the feed. For the scrubbing of co-extracted Mo, citrate-buffered acetohydroxamic acid was used. The separation of An(III) from the trivalent lanthanides (Ln(III)) was achieved using citrate-buffered diethylene-triamine-N,N,N',N'',N''-pentaacetic acid (DTPA), and Ln(III) were efficiently back extracted using N,N,N',N'-tetraethyl-diglycolamide (TEDGA). A clean An(III)

product was obtained with a recovery of 95% americium and curium. The Ln(III) were efficiently stripped, but the Ln(III) product contained 5% of the co-stripped An(III). The carryover of Am and Cm into the Ln(III) product is attributed to too few actinide stripping stages, which was constrained by the number of centrifugal contactors available. Improved separation would be achieved by increasing the number of An strip stages. The heavier lanthanides (Pr, Nd, Sm, Eu, Gd) and yttrium were mainly routed to the Ln product, while the lighter lanthanides (La, Ce) were mostly routed to the raffinate."

HETEROLEPTIC COMPLEX FORMATION IN THE SO_3-Ph-BTP/TODGA SYSTEM

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EXTRACTABLE HETEROLEPTIC COMPLEXES OF Am(III) AND Eu(III) WITH A TRIPODAL DIGLYCOLAMIDE EXTRACTANT AND SULPHONATED BIS-TRIAZINYL PYRIDINE AS A STRIPPING AGENT. CRITERION INDICATING THE FORMATION OF SUCH COMPLEXES IN SOLVENT EXTRACTION SYSTEMS

Solvent extraction-based separation Am(III) and Cm(III) from the lanthanide fission products in the i-SANEX processes is achieved by selective stripping of the actinides from the loaded organic phase with TODGA (Figure 1) extractant. Such particular two-phase systems contain two different ligands, a lipophilic extractant and a hydrophilic stripping agent, which compete for a given metal ion to form its lipophilic or hydrophilic complexes. The stripping agent is the anion of (2,6-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)pyridine (SO_3-Ph-BTP^{4-}, Figure 1) which forms strong hydrophilic complexes with Am(III) and Cm(III).[8-9] However, unexpected results on the Am(III) behaviour in a similar system allowed us to formulate a hypothesis that lipophilic heteroleptic complexes of Am^{3+} ions with both TODGA and SO_3-Ph-BTP^{4-} ligands may form in this system, and their extraction to the organic phase could reduce the efficiency of the Am(III) stripping.[10] Assuming this hypothesis being correct, we intended to look for a way to improve the stripping step by preventing formation of the heteroleptic complexes.

Both ligands used in the process are tridentate with the donor atoms either O- or N- for TODGA and SO_3-Ph-BTP, respectively. Therefore, trivalent cations of minor actinides (Am^{3+}, Cm^{3+}) and middle lanthanides, e. g. Eu^{3+} (all of the coordination numbers, CN, close to nine) form with these ligands coordinatively saturated 1:3 complexes. The complexes with lipophilic TODGA are well extracted to the organic phase. Also, hypothetical anionic heteroleptic complexes, $[Am(TODGA)_2(SO_3\text{-Ph-BTP})]^-$, were expected to be extracted as ion pairs with the protonated lipophilic ligand $TODGA{\cdot}H^+$. However, for entropic reasons, heteroleptic complexes should not form in a similar system with a nonadentate extractant.

Our research was undertaken with such an extractant, T-DGA, consisting of three interconnected DGA molecules (Figure 4), which was considered to be nonadentate, fully meeting the coordination requirements of the M^{3+} ions studied.[11] However, further studies have shown that not only the 1:1 but also 1:2 complexes of Am^{3+} [12] and Eu^{3+} [13] with T-DGA are extracted to the organic phase. It has

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been suggested that this is due to steric constraints in the T-DGA molecule, preventing the use of all three tridentate pendant arms of the same ligand to coordinate the metal ion.[13] The same effect, leading to the formation of extractable heteroleptic complexes of $Eu^{3+}$and Am^{3+} ions with T-DGA and SO_3-Ph-BTP^{4-} ligands, has been observed in our present work (see below). On the other hand, our further study of the TODGA / SO_3-Ph-BTP system proved that the suggested heteroleptic complexes do not practically form and do not affect the extraction equilibrium of Cm^{3+} the analogue of Am^{3+}. [14] However, we decided to continue our investigation of the T-DGA / SO_3-Ph-BTP system in order to compare solvent extraction of Am^{3+} and Eu^{3+} in two systems containing the same stripping agent (SO_3-Ph-BTP) and chemically similar extractants (TODGA and T-DGA), but differing in the fact that heteroleptic complexes form in one system and not in the other. The diluent used was 1-octanol which in our opinion plays a special role in the extraction of T-DGA complexes. Inner-sphere solvation by three octanol molecules of the M^{3+} ions (of CN =9) already hexacoordinated by one T-DGA ligand, makes the complexes extractable to the organic phase. The tridentate SO_3-Ph-BTP^{4-} anion easily replaces the three 1-octanol molecules in the inner-sphere of the metal to form extractable heteroleptic complexes. In the present work we hoped to find and formulate a simple criterion allowing to determine whether there are heteroleptic complexes in a given solvent extraction system or not.

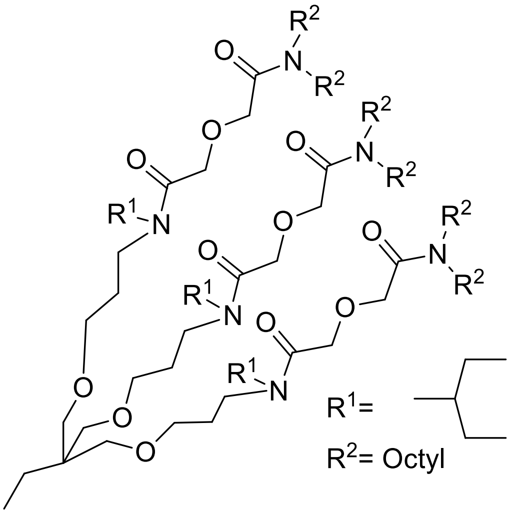

Figure 4. Structural formula of T-DGA.

The solvent extraction experiments were carried out in two-phase systems consisting of varying concentrations of T-DGA in 1-octanol (from ca. 0.01 to ca. 0.07 mol/L) and varying concentration of $Na_4(SO_3$-Ph-BTP) (from $1.2 \cdot 10^{-5}$ to 0.1 mol/L) in an aqueous solution of 0.3 mol/L HNO_3 + 0.7 mol/L $NaNO_3$. Distribution ratios of the M(III) metals studied, $D = C_{M,org}/C_{M,aq}$, where $C_{M,}$ is the molar concentration of M in the organic or aqueous phase at equilibrium, were radiometrically determined at 25°C using ^{241}Am and ^{152}Eu radiotracers (with carriers), separately. D_0 values denote the distribution ratios determined in the systems containing no SO_3-Ph-BTP. The concentration of the deprotonated ligand SO_3-Ph-BTP^{4-}, $[L^{4-}]$, in the 0.3 mol/L HNO_3 aqueous solutions containing the metal ions at trace concentrations, was calculated from a simplified [4] equation: $[L^{4-}] = [L](1 + K_{H,1}[H^+])^{-1}$, where $K_{H,1}$ is the

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first protonation constant of SO_3-Ph-BTP^{4-}, equal to $\log K_{H,1} = 0.5$.[10] With these values it is shown that $\log[L^{4-}] = \log[L] - 0.29$.

Figure 5 shows the log *D* values of both metals studied, plotted as a function of log [L], with the organic phase being approximately 0.07 mol/L T-DGA in 1-octanol. Unfortunately, due to a mistake in the preparation of the stock solution of T-DGA in 1-octanol (that's why the word "approximately" was used above), this and several other results obtained should be considered "preliminary" and they need to be repeated.

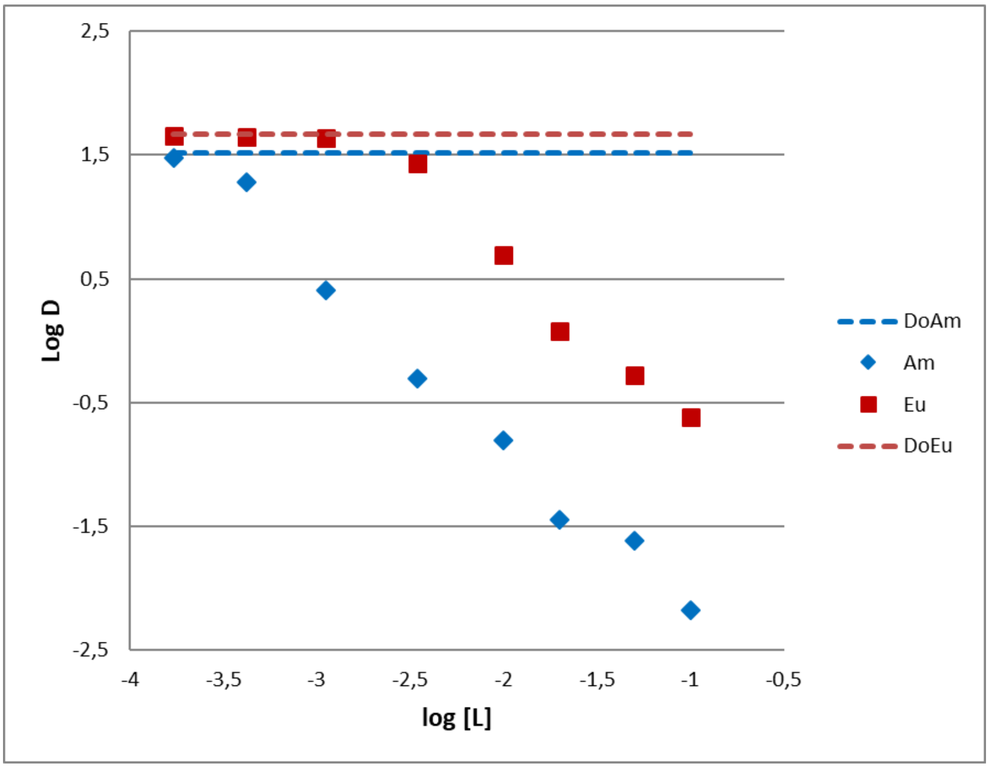

Figure 5. Log-log dependences of the distribution ratios, *D*, of Am(III) (blue diamonds) and Eu(III) (red squares) on the total molar concentration of SO_3-Ph-BTP, [L], in the aqueous phase (0.3 mol/L HNO_3 + 0.7 mol/L $NaNO_3$). Organic phase, ≈ 0.07 mol/L T-DGA in 1-octanol. *T* = 25 °C.

The basic equation used to determine the stoichiometries and stability constants ($\beta_{L,i}$) of a series of i consecutive ML_i complexes formed in the aqueous phase containing a hydrophilic stripping agent L and equilibrated with an organic phase containing a lipophilic extractant is given by Eq. (1).[15] In the present case, the stripping agent is the anionic ligand SO_3-Ph-BTP^{4-} dissolved in the 1 mol/L $HNO_3/NaNO_3$ solution, but the complexation of the Am^{3+} and Eu^{3+} ions by nitrates is neglected.[10]

$$\sum_{i=1}^{k} \beta_{L,i}[L^{4-}]^{i} = \left(\frac{D_0}{D} - 1\right) \tag{1}$$

The symbols i, *D*, D_0, $\beta_{L,i}$ and $[L^{4-}]$ have been defined above. After limiting our interest to only one region of L concentration where a given complex (1:1, 1:2, etc.) predominates, we arrive at:

$$\log\left(\frac{D_0}{D}-1\right)=\mathrm{i}\log[\mathrm{L}^{4-}]+\log\beta_{\mathrm{L,i}} \qquad (2)$$

where i = 1, 2, etc.

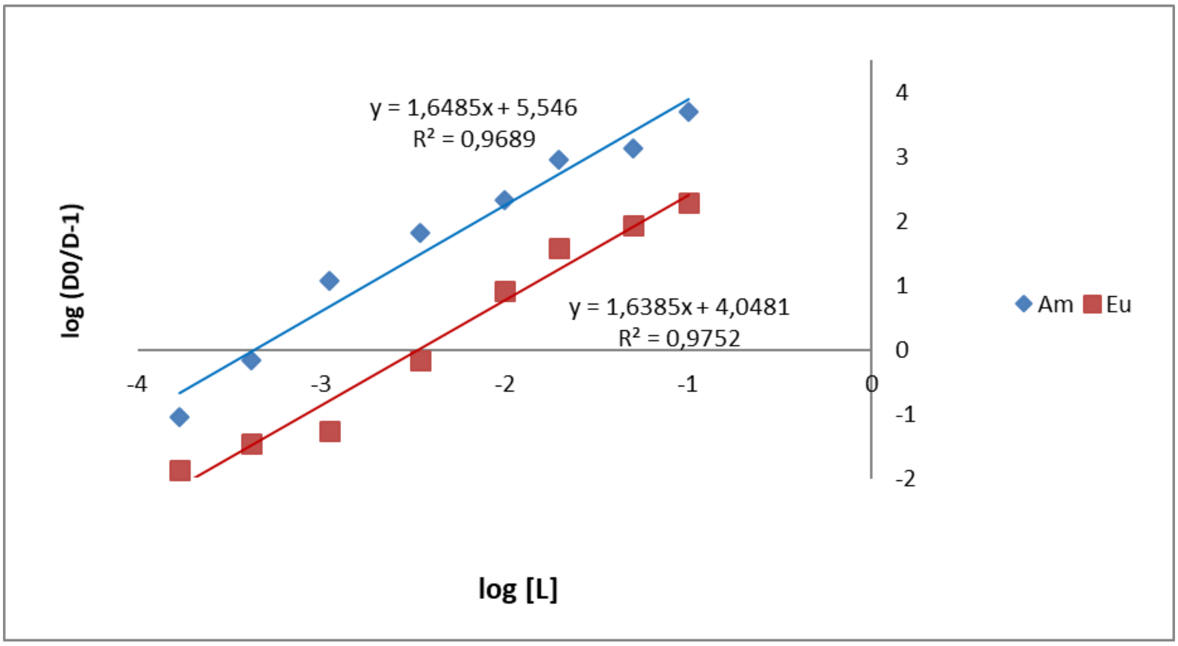

Figure 6. Log (D_0/D - 1) as a function of log [L^{4-}] for Am(III) (diamonds) and Eu(III) (squares). Organic phase, ≈ 0.07 mol/L T-DGA in 1-octanol. T = 25 °C.

Figure 6 shows the plots corresponding to Eq. (2). It can be seen that roughly straight-line relationships (as expected) were obtained for both metals, but their non-integer slopes (ca. 1.6 each) strongly differ from the expected integer values equal to 1 and 2 obtained in the earlier work for the TODGA / SO_3-Ph-BTP system.[10] The inconsistency of the expected slope values and those obtained in the present work can easily be interpreted by the fact that equations (1) and (2) describe a simple solvent extraction-stripping process when only homoleptic complexes of a given metal appear in each liquid phase: the complexes with the extractant in the organic and the complexes with the stripping agent in the aqueous phase. Therefore, the present case of non-integer slopes that is inconsistent with the above equations should be interpreted as the evidence of the appearance in the organic phase of additional heteroleptic metal complexes which are not considered in the model of a simple extraction process.

The existence of only one straight line for each metal in Figure 6 may be due to limiting the experiments to the higher [L] values. In fact, similar studies carried out at in a broader range of [L] values, since log [L] = –5.0 (at 0.04 mol/L T-DGA in 1-octanol), show distinct two straight line dependences for each metal, the slope of each line being also a non-integer value (Figure 7). A similar picture has also been observed for the case of 0.02 mol/L T-DGA in 1-octanol.

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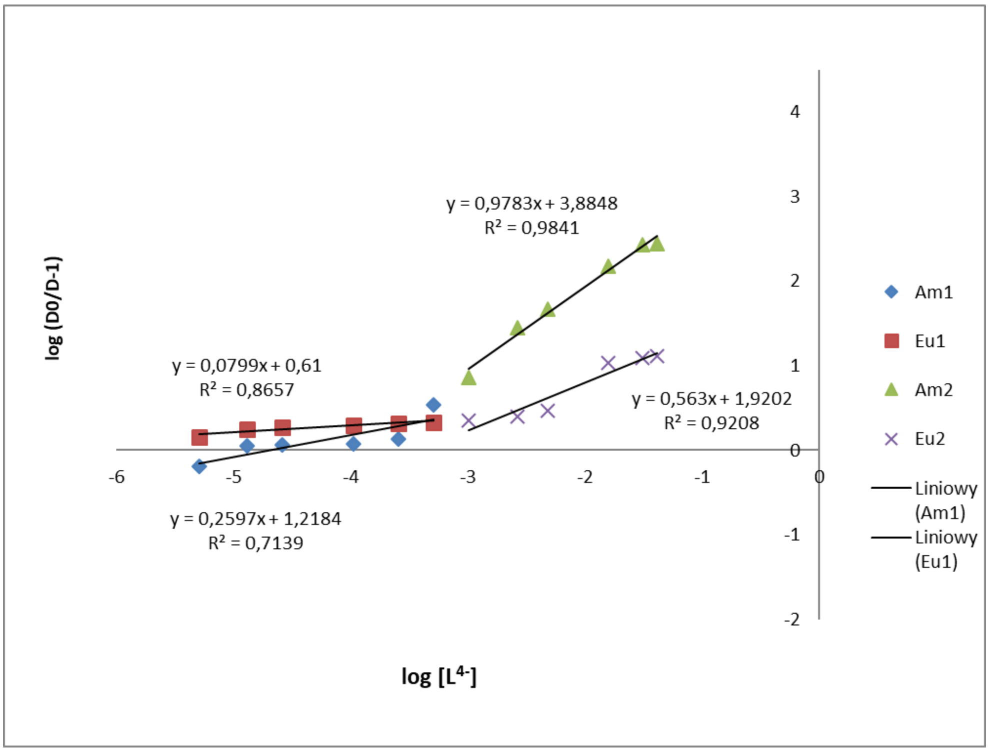

Figure 7. Log (D_0/D - 1) as a function of log [L^{4-}] for Am(III) (diamonds + triangles) and Eu(III) (squares + X symbols). Organic phase, 0.04 mol/L T-DGA in 1-octanol. T = 25 °C.

We expected that a correction of the simple model of the solvent extraction-stripping process, in particular taking into account the presence in the organic phase of heteroleptic complexes of a given metal as well when deriving the equation analogous to Eq. (1), would result in correcting the slopes of the straight lines in Figure 6 and Figure 7 to the integer values.

Such correction was done for the case shown in Figure 6. We have derived the following new relationship which considers the additional extraction of the heteroleptic complex:

$$\log\left(\frac{D_0+\beta_{\mathrm{MEL}}[\mathrm{H}^+][\mathrm{E}][\mathrm{L}^{4-}]}{D}-1\right)=\mathrm{i}\log[\mathrm{L}^{4-}]+\log\beta_{\mathrm{L,i}} \qquad (3)$$

where [E] is the molar concentration of the extractant E (in this case T-DGA) in the organic phase, β_{MEL} – the stability constant of the hypothetical heteroleptic complex. The other symbols are defined above.

In the case of a metal ion with CN=9 (Am^{3+}, Eu^{3+}), the formation of only one, 1:1:1, heteroleptic complex with hexadentate (T-DGA) and tridentate (SO_3-Ph-BTP^{4-}) ligands is possible. This heteroleptic anionic complex [M(T-DGA)(SO_3-Ph-BTP)]$^-$ is probably extracted either as the protonated neutral molecule or as an ionic pair with the protonated extractant, T-DGA.H^+. Therefore, the acidity of the aqueous phase should be kept constant within the experiments so as not to disturb the measured values of D. Equation (3) is valid at trace concentrations of the metal ions, [M^{3+}] << [L^{4-}]. Moreover, it is valid in such ranges of the [L^{4-}] values where i becomes equal to the defined integer value, *i.e.* i=1, i=2, *etc.*

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In the present work we examined the correctness of equation (3) and tried to find such a value of β_{MEL}, at which the slope, i, takes on a given integer value. Eq. (3) contains three experimental parameters: two of them ($[H^+]$ and [E]) constant and one, $[L^{4-}]$, variable (calculated from the total concentration of hydrophilic ligand SO_3-Ph-BTP, $[L]_{tot}$, using $K_{H,1}$ = 3.16 L/mol [10]). Together with the two experimentally measured parameters, D_0 and D, and a guessed β_{MEL} value, they allow to calculate the numerical value of A_L equal to the left side of Eq. (3), corresponding to the given value of $[L^{4-}]$ for the entire range of the $[L^{4-}]$ values tested. Further plotting of the calculated A_L values as a function of the corresponding $\log[L^{4-}]$ ones results in a straight-line dependence with a slope j. The procedure, performed for Am and Eu with the experimental data shown in Figure 6, was repeated with new guessed β_{MEL} values until the slope i for Am became equal to 2.00. That has been attained at $\beta_{MEL} = 8.57 \cdot 10^5$ (Figure 8). However, due to the uncertainty in the T-DGA concentration (see above), this β_{MEL} value is approximate and will be determined again. Extrapolation of the straight line to $\log[L^{4-}] = 0$ allows to determine the value of the stability constant of the 1:2 complex, $[Am(SO_3\text{-Ph-BTP})_2]^{5-}$, in the aqueous phase, uncorrected for Am(III) complexation by nitrate anions: $\log \beta_{L,2} = 7.69$. The corrected (by adding 0.51 ± 0.05[10]) value, $\log \beta'_{L,2} = 8.2$, is greater than the value of 7.67 ± 0.06 determined in the TODGA system.[10]

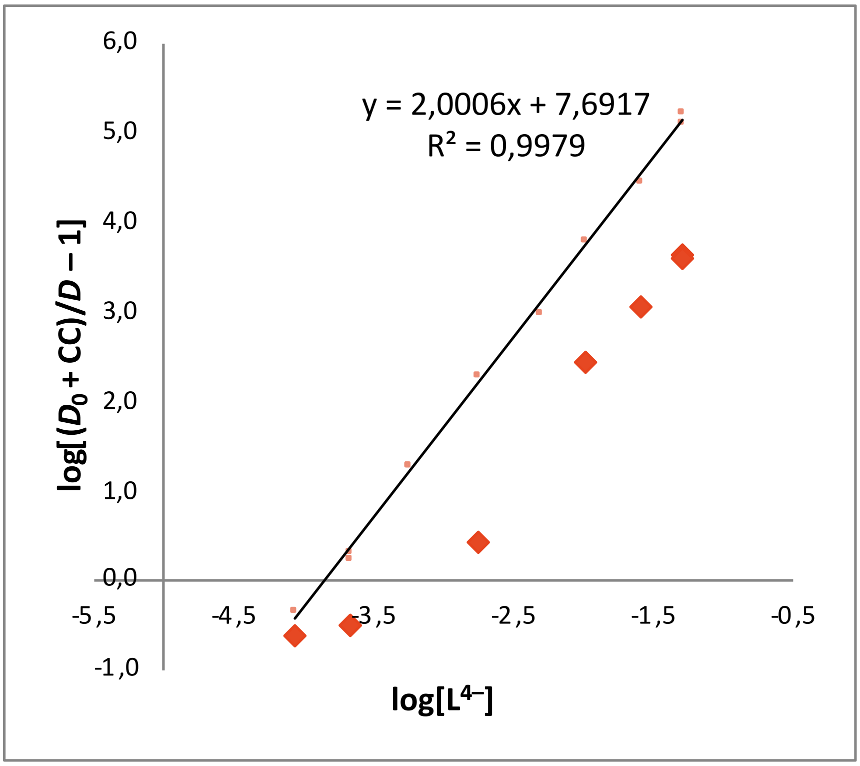

Figure 8. Plots of log $[(D_0 + CC)/D - 1]$ vs. log $[L^{4-}]$ for Am(III) (small orange squares) and Eu(III) (red diamonds), stripped from the organic phase with the SO_3-Ph-BTP^{4-} ligand (L), where the correcting factor, CC = $\beta_{MEL}[H^+][E][L^{4-}]$

The experimental data for Eu^{3+} (Figure 8) appear insufficiently coherent to obtain a similar fit, but their likely similar slope shows that the β_{MEL} value for the heteroleptic Eu complex should be close to $8 \cdot 10^5$. The lower slope of the curve for Eu(III) at the lowest SO_3-Ph-BTP concentrations studied is probably due to the 1:1 Eu(III) – SO_3-Ph-BTP^{4-} complex prevailing at $\log[L^{4-}] < -4$. Further studies should, therefore, be extended to the range $-5.5 < \log[L^{4-}] < -4.5$, like in the case of 0.04 mol/L T-DGA (Figure 7). The experiment at 0.04 mol/L T-DGA should also be repeated because of too large scatter of the experimental points.

Nevertheless, the data presented above make possible to formulate a new experimental criterion that indicates whether the extractable heteroleptic complexes of a metal cation with a lipophilic extractant and a hydrophilic stripping agent are really formed and extracted to the organic phase of a given solvent extraction system containing the two ligands that compete for the metal cation:

If, in the areas of stripping agent concentrations used in the experiment, equations (1) and (2) show linear relationships with the slopes equal to small integers, the extraction of the metal into the organic phase is described by a simple model that predicts the extraction of only its homoleptic complex(es) with the extractant. In contrast, if the slopes are significantly different from the integers, a simple model does not correctly describe the extraction process, and then equation (3) must be applied, describing the extraction into the organic phase of heteroleptic complexes with both extractant and stripping agent.

CONCLUSIONS

A criterion has been formulated to indicate the formation of heteroleptic complexes in solvent extraction systems containing two different ligands competing for the extracted metal ions: a lipophilic extractant and a hydrophilic stripping agent. Such heteroleptic complexes are readily formed in a solvent extraction system containing the T-DGA and SO_3-Ph-BTP ligands. Further work is in progress.

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