

LIQUID-LIQUID EXTRACTION OF Am(III), Pu(IV), AND U(VI) USING MALONAMIDE AND TRIBUTYL PHOSPHATE IN ROOM TEMPERATURE IONIC LIQUIDS AS DILUENT

by

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The extraction behavior of americium, plutonium, and uranium from nitric or hydrochloric acidic medium by a solution of malonamide or tributyl phosphate in trioctylmethylammonium chloride (Aliquat 336) or trihexyl(tetradecyl)phosphonium chloride (Cyphos 101) ionic liquids were studied. The extraction percentage of these actinides was measured as a function of a concentration of nitric or hydrochloric acid and a set combination of a room temperature ionic liquid and an extractant. The pure room temperature ionic liquids could extract the plutonium and uranium to various degrees, with the combinations with extractants affecting the extraction percentages further. The 100 % extraction efficiency of plutonium was achieved across all concentrations of HCl with the combinations of Cyphos 101 and malonamide. 100 % extraction efficiency of uranium was achieved by Cyphos 101 and malonamide from distilled water. Americium was not extracted by any combination under any conditions.

Key words: room-temperature ionic liquid, americium, plutonium, uranium, malonamide, tributyl phosphate, extraction percentage

INTRODUCTION

With the focus on limiting carbon emissions in electricity production and phasing out fossil fuels, currently, nuclear energy remains one of the only two forms that can continuously produce needed power. While it has the lowest carbon footprint of the electricity sources, nuclear waste, and its handling and storing remains one of the detractors from nuclear energy. Therefore, reprocessing of spent nuclear fuel is of great interest, with the PUREX method already producing mixed oxide (MOX) fuel. Even though new processes and procedures for extracting plutonium and uranium from the spent fuel are being continuously developed [1], namely extraction chromatography and discoveries of various extraction materials, liquid-liquid extraction (LLE) remains one of the main methods for separation. However, traditional LLE with organic molecular solvents, while still being irreplaceable in some applications, is being reconsidered where possible to limit the amount of the solvents and to replace it with less volatile substances. The search for a *greener* solvent coincides with increased interest in room temperature ionic liquids (RTIL), a long-known but relatively recently further studied new category of molten salts. The RTIL includes one or more alkyl side chains connected to one of

its ions and one of the ions also needs to be large with a low degree of symmetry for the RTIL to remain liquid. The most common cations nowadays include imidazolium, pyrrolidinium, piperidinium, ammonium, and phosphonium cations. Anions can range from simple ones like chloride or nitrate to (fluorosulfonyl) imides. The perceived greenness of RTIL is however still discussed [2]. They are especially challenged by toxicity research studies, as while RTIL may not evaporate easily, they can still enter the environment. Due to the ionic structure, a wide range of anions and cations can be paired together to achieve the desired capabilities. It is estimated that there are up to 1018 possible combinations. The RTIL is being studied for a wide range of applications including extraction and separation, biomass utilization, lubricants or support to membranes, pharmaceutical salts, synthetic reactions, and others.

Numerous combinations of RTIL and extractants were investigated for the extraction of actinides by several researchers. The currently most studied RTIL are based on imidazolium cations in combination with various neutral extractants. The Pu(IV), Am(III), and uranyl ion UO_2^{2+} were all successfully extracted by N, N-Diisobutyl-2-[octyl (phenyl) phosphoryl] acetamide (CMPO) or tributyl phosphate (TBP) in $[C_4mim][PF_6]$ [3]. Extraction of rare earth metals by imidazolium $[C_nmim]$ RTIL with added organophosphorus, diglycolamide, ketone, or

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phosphine oxide extractants (HDEHP, TODGA, HPMBP, and Cyanex respectively) was studied by Kubota and Goto [4] and Gujar *et al.* [5]. Chen *et al.* [6] and Nakashima *et al.* [7] studied a combination of CMPO extractants in [bmim] RTIL. Extraction of U(VI) TBP in [bmim][Tf₂N] was studied by Giridhar *et al.* [8]. Vasudeva Rao *et al.* [9] studied the extraction of U(VI) by [bmim][PF₆] and [bmim][Tf₂N] with TBP. Rout *et al.* [10] studied the separation of Eu(III) from Am(III) using [omim][Tf₂N] with D2EHPA and HDEHDGA extractants. Extraction of Am(III) by CMPO-functionalized pillar[5]arenes (POP5A) in [C₈ mim][Tf₂N] was reported by Sengupta *et al.* [11]. Chen *et al.* [12] studied the extraction of U(VI) with phosphine oxide functionalized POP5A in [C₈ mim][Tf₂N]. Extraction of uranyl ion by TOPO and its mixture with D2EHPA in [C_n mim][Tf₂N] was studied by Mohapatra *et al.* [13]. Shen *et al.* [14] studied the extraction of uranyl ions with diglycolamide extractant in [C_n mim][Tf₂N]. Extraction of U(VI) and Pu(IV) using tri-alkylamines in [C_n mim][Tf₂N] was reported by Ansari *et al.* [15]. Rama *et al.* [16] reported the separation of Pu(IV) using 2-hydroxy acetamide in [C₄ mim][Tf₂N]. Panja *et al.* [17] also investigated the extraction behavior of selected actinides using TODGA in [C_n mim][Tf₂N].

Besides imidazolium RTIL, other types are also used for extractions. Bell and Ikeda [18] tested ammonium-based RTIL in combination with TBP to extract U(VI) from a nitric acid medium. Rout *et al.* [19] reported extraction of Pu(IV) by trioctylmethylammonium bis(trifluoromethanesulfonyl) imide [N₁₈₈₈][Tf₂N] RTIL containing monoamide N,N-dihexyloctanamide (DHOA).

In some cases, RTIL can extract the metal without an additional extractant, such as in the case of extraction of Ce(IV) by pure [C₈ mim] PF₆ [6]. Pure [N₁₈₈₈]⁺-based RTIL was successfully investigated for heavy rare earth metal separation by Kubota and Goto [4].

The most common mechanism of extraction of ions in various valence states with the addition of neutral organic extractants (TBP, CMPO, malonamide, or diglycol) is cation exchange. The presence of an extractant is necessary since extraction of cations by pure imidazolium RTIL is negligible [20]. Cation exchange is also preferred with shorter alkyl chains of the cationic part of RTIL and more hydrophobic anions. The cation exchange by its mechanism leads to a loss of RTIL into an aqueous phase, limiting the sustainability of the process.

The extraction of neutral ion pairs is usually achieved in higher concentrations of mineral acids, mostly HNO₃, where the acidic anion neutralizes the metallic cation. Extraction of neutral or ion pair species is also supported by hydrophobic cations of RTIL.

The extraction of anionic species is reported for combinations of imidazolium RTIL with various extractants (BenzoDODA, DMDOHEMA [21], DMDDBMA [22]) from higher, usually 3M* or 4M, concentrations of acids. Some of the anionic species of metals can be extracted by

pure RTIL [22]. While trivalent actinides require the presence of an extractant as pure RTIL achieves negligible extraction, tetra- and hexavalent actinides have been reported to be extracted by pure RTIL from higher concentrations of acids [20].

In general, the extraction techniques can differ based on the applied extractant, length of the alkyl chains of RTIL's cation, concentration of the acid in the aqueous phase, or any combination of the three.

Thus, based on the information provided by Rout *et al.* [23], trioctylmethylammonium chloride (Aliquat 336) and trihexyl(tetradecyl)phosphonium chloride (Cyphos 101) RTIL were selected for study of their capability to extract uranium, plutonium and americium. The reasoning was partially due to the stated assumption of them not exhibiting cation exchange, and partially because no paper was found at the time that studied these RTIL in greater detail. At the time of the beginning of the study, only the work by Rout *et al.* [19] was found that nevertheless used different extracting agents, and the structure of the RTIL is also significantly different due to using bis(trifluoromethanesulfonyl)imide instead of chloride. Since then, Rout and Ramanathan published a paper in 2022 about the use of Cyphos nitrate for extracting plutonium [24].

As the general assumption about the extraction of metals by RTIL is that pure RTIL rarely achieves significant values, extracting agents were selected to be dissolved in RTIL to study possible synergic effects. Based on the research, malonamide, and tributyl phosphate [22] [8] were chosen as the initial extractants.

EXPERIMENTAL SECTION

Materials

All the chemicals used were of analytical grade and were used as received, methyltrioctylammonium chloride (mixture of C8-C10 with C8 predominating, Aliquat 336, labeled AL) was obtained from Sigma-Aldrich (USA), trihexyltetradecylphosphonium chloride (Cyphos 101, labeled CY) from Sigma-Aldrich (USA) and Abcr (Germany), malonamide (labeled MA) from Lach-Ner, (Czech Republic), and tributyl phosphate (labeled TBP) from Fluka (USA). Nitric and hydrochloric acids were procured from Penta, Inc. (Czech Republic). Radiochemical isotope tracers ²³²U, ²³⁸Pu, and ²⁴¹Am were obtained from AEA Technology, UK, and QSA Amersham International. The ²³²U tracer was prepared by removing its ²²⁸Th daughter using ion exchange resin BIORAD AG 1-X8 (200-400 mesh).

Procedures

The steps to determine the extraction percentages for each combination of RTIL, aqueous phase, and radionuclide were as: dissolving the extractant in

* M stands for mol L⁻¹

the ionic liquid supported by an ultrasound bath, adding the aquatic phase into the separation tubes, adding the radionuclide, adding the equivalent of 1 mL of the ionic liquid with the extractant, mixing of phases supported by a rotator, separation of the phases supported by a centrifuge, transferring an aliquot of the organic phase into scintillation cocktail, and liquid scintillation counting. Each sample was prepared twice for the statistical evaluation.

Based on the research the initial concentration of extractants was selected as 0.1 mol dm^{-3} . The appropriate amount of malonamide was weighed and added to the weight equivalent of 20 mL of ionic liquid that was used for one batch of tests. The mixture was placed into the Sonorex ultrasonic bath (Bandelin electronic GmbH & Co. KG, Germany) for 90 minutes at $25 \text{ }^\circ\text{C}$. The length of time was determined experimentally to ensure complete dissolution of the extractant. In the case of TBP, the appropriate volume was measured, and the mixture was placed into the ultrasound bath for 90 minutes to ensure the same conditions as the ionic liquid.

The 1 mL of the aqueous phase was added into 5 mL ClikLok MCT tubes (Simpot Scientific Inc., Canada). The aqueous phase was formed by distilled water, 0.01M, 0.1M, and 1-5M nitric acid, or 0.1M, 1M, 4M, and 7 M hydrochloric acid.

An aqueous sample of a radionuclide was added to each tube with the aqueous phase. The activities of the samples were 80 Bq for ^{241}Am , 104.6 Bq for ^{238}Pu , and 60.6 Bq for ^{232}U .

Due to the high viscosity of the used ionic liquids, adding a specific volume introduced significant errors and was not feasible. Therefore we weighed an equivalent of 1 mL with an error margin of $\pm 0.005 \text{ g}$ (a single droplet from a 1 mL pipette) into the vials with the aqueous phase and radionuclide.

The tubes were placed at the Rotator Drive STR4 (Stuart Scientific, UK) to support the mixing of phases. The rotator was set at 40 rpm* for 2 hours.

The tubes were then settled for 30 minutes and placed into the MPE-340 centrifuge (Mechanika precyzyjna, Poland) for 1 minute at 626 RCF** to support the phase separation.

Similarly to the measuring of 1 mL of RTIL, measuring the precise volume to be added into the AquaLight (Hidex, Finland) scintillation cocktail was impossible, therefore the equivalent of 500 μL was weighted into the scintillation vial.

Vials with the samples were inserted into the Hidex 300 SL Scintillation Counter with PLI set at 0. The whole set of samples was left for 10 minutes in darkness before measurements. Each vial was measured 3 times for 300 seconds.

The activity of the samples was determined based on the measured counts per minute (cpm). The

cpm of each sample was averaged, then both samples of the same combination were averaged, and standard deviation σ was determined as

$$\sigma = \sqrt{\frac{\sum (X - \mu)^2}{N}} \quad (1)$$

The extraction percentage %E was calculated as a ratio of the calculated activity from the samples and the original activity of the tracer radionuclide, multiplied by 100

$$\%E = \frac{A_{\text{sample}}}{A_{\text{tracer}}} 100 [\%] \quad (2)$$

Background radiation was measured and averaged for each combination of RTIL, extractants, and concentrations of acids. The initial results from scintillation counting for each sample were reduced by the value of the respective background.

Quenching for all combinations of RTIL, extractants, and concentrations of acids was measured and included in the calculations of the activity of the sample.

Due to various levels of water absorption by RTIL based on the concentration of acids in the aqueous phase, the remaining aqueous phase was measured to determine the total volume of the organic phase from which a portion was taken for scintillation counting. This ratio was included in the final calculation of the sample activity.

RESULTS AND DISCUSSIONS

The extraction percentages from distilled water as the aqueous phase are included in both extractions from HCl and HNO_3 as a "0" M concentration of an acid in all figures. For better visibility at lower concentrations, a fixed interval for all values on the x-axis was used.

The initial intent to include extraction percentages of the extractants in traditional volatile organic solvents proved to be impossible to achieve as malonamide could not be dissolved in any of the used solvents that can produce a phase boundary with water (see tab. 1, relative polarities taken from [25]). However, the fact that malonamide can be dissolved in RTIL and the combination can enhance the extraction properties of pure RTIL is one of the benefits of RTIL implementation.

Since no studies were available for AL or CY, we could not compare the observed aqueous phase absorption to any existing data. Rout and Ramanathan [24] stated the strong hydrophobic character of Cyphos nitrate, Rout *et al.* [19] did not mention any hydrophobic or hydrophilic properties of $[N_{1888}][Tf_2N]$. Performed testing shows a significant aqueous phase absorption by RTIL at low nitric acid concentrations up to 2M, with AL absorbing up to 17 % of the aqueous phase and CY absorbing up to 11 %. Besides, AL forms a third phase with distilled water and 0.01M

*rpm stands for revolutions per minute

**RCF stands for relative centrifugal force

Table 1. Overview of the capability of selected solvents to dissolve MA and maintain phase boundary with water based on their relative polarity

Solvent	Relative polarity	Dissolves MA	Water miscible
<i>n</i> -dodecane	0.000	–	–
<i>n</i> -heptane	0.012	–	–
xylene	0.074	–	–
toluene	0.099	–	–
benzene	0.111	–	–
diethyl ether	0.117	–	–
chloroform	0.259	–	–
pyridine	0.302	–	–
1,2-dichloroethane	0.327	–	–
benzotrile	0.333	+	forming emulsion
N, N'-dimethylpropyleneurea	0.352	+	+
N, N-dimethylformamide	0.386	+	+
dimethylsulphoxide	0.444	+	+
formamide	0.799	+	+

nitric acid. For comparison, the amount of absorbed aqueous phase in $[C_n \text{ mim}][[Tf_2N]$ or $[bmim][[Tf_2N]$ RTIL is reported to be increasing with the concentration of nitric acid [8, 22]

The extraction of ^{241}Am proved ineffective by all tested combinations, the overall extraction efficiency is below 5 % and only in several instances reaches 10 %. The expected reason is that unsubstituted MA and TBP do not form complexes with ^{241}Am that can be extracted by the used RTIL. The extraction efficiency for ^{232}U is almost identical regardless of the presence or absence of an extractant, with the only significant distinction of extraction with AL + MA from distilled water or low concentrations of nitric acid. The extraction efficiencies for ^{238}Pu depend on the combination of RTIL, extractant, and aqueous phase.

Extraction of U

All but two combinations can extract uranium with an efficiency greater than 25 % and exceeding 50 % for most. There is a strong similarity between the extraction percentages of pure RTIL and RTIL with extractants, with several significant distinctions.

Extraction of U using AL

Extraction percentages for pure AL and AL + TBP are almost identical, especially in extractions from HCl aqueous phase, fig. 1. Extraction by AL + MA follows the same trend from 0.1–7M HCl solutions, the only significant outlier is the extraction using AL + MA from distilled water, where the extraction efficiency is $92 \pm 2\%$ indicating a different extraction method by MA. Spjuth *et al.* [26] does not include values for extraction from the distilled water and considers only HNO_3 conditions. However, it states that extraction at concentrations of

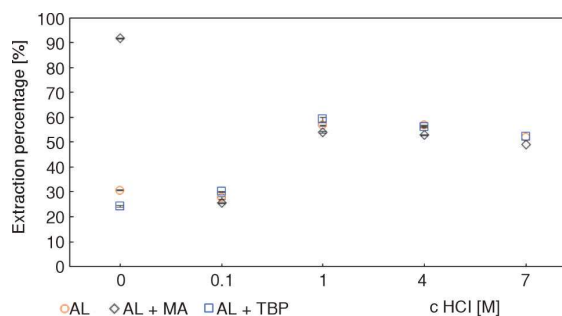


Figure 1. Extraction percentage of U from distilled water and 0.1-7 M HCl by AL, AL + MA, AL + TBP

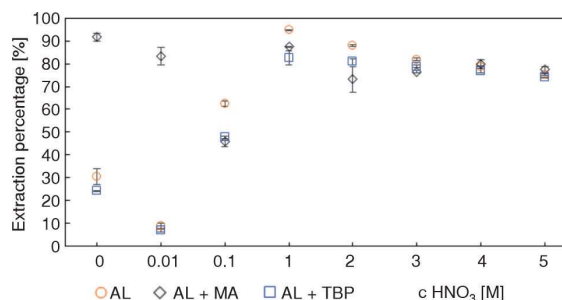


Figure 2. Extraction percentage of U from distilled water and 0.01-5 M HNO₃ by AL, AL + MA, AL + TBP

$\text{HNO}_3 < 1 \text{ M}$ follows the co-ordination of neutral nitrate complexes. This would indicate that chloride complexes are not extracted by MA and a different complex is formed in the distilled water even in the presence of chloride anions from the AL. Otherwise, the extraction percentage is 25-30 % in distilled water and 0.1M HCl, then increasing to 55-60 % from 1M HCl and slowly decreasing to the range of 50 % extraction efficiency from 7M HCl. Therefore, the presence of any of the tested extractants does not change the extraction efficiency of the pure AL from 0.1–7M HCl solutions, with the extraction percentage reaching a maximum of 1M HCl.

Extractions from HNO_3 show similarities to extractions from HCl, namely low efficiencies for pure AL and AL + TBP from distilled water or at low concentrations, reaching maximum from 1M HNO_3 and then decreasing. However, the extraction percentages are significantly higher than in HCl conditions reaching 82-95 %, with pure AL having the highest percentage, and the minimum from 5M HNO_3 is still over 73 %, fig. 2. Combination of AL + MA maintains high efficiency of $83 \pm 4\%$ even from 0.01M HNO_3 , suggesting extraction of neutral nitrate complexes by MA, whereas the extraction percentages of both pure AL and AL + TBP drop significantly to 7-8 % from 0.01M HNO_3 , even though TBP should form neutral nitrate complex as well, before increasing to 45-47 % for AL with extractants and $62 \pm 1\%$ for pure AL from 0.1M HNO_3 . From 0.1M HNO_3 , all variants show similar extraction percentages, with pure AL having slightly higher values than the variants with extractants till to 3M HNO_3 . The

drop of the efficiency for AL + MA at 0.1M HNO₃ to the level of AL + TBP suggests a change in the extraction mechanism to one that is suppressing the extraction properties of the pure AL. A possible explanation could be a cocreation of non-neutral complexes with the extractants that cannot be extracted by the pure AL.

Extraction of U using CY

Extraction of U by CY or CY + extractant follows a similar trend for both HCl and HNO₃, while extractions from HNO₃ have higher efficiency. The extraction percentages are like those for pure CY and CY + extractants, especially in extractions from HCl, fig. 3, where the efficiencies are nearly identical. The maximum efficiency of 91 ± 2% for pure CY and 99 ± 3% for CY + MA is achieved from distilled water. The efficiency then drops to the 64 % range at 0.1M HCl and gradually decreases to 5 the 4 % range.

In extractions from HNO₃, the highest efficiency for any combination is from distilled water, where CY + MA and CY + TBP reach over 98 % and pure CY has an extraction percentage of 91 ± 2%. The efficiency then decreases gradually to the 70-85 % range with CY + TBP combination maintaining higher efficiencies, fig. 4.

Extraction of Pu

Extraction of Pu is much less consistent compared to U. The values range from 0 to 100 % and there is some outlier value breaking the trend for most com-

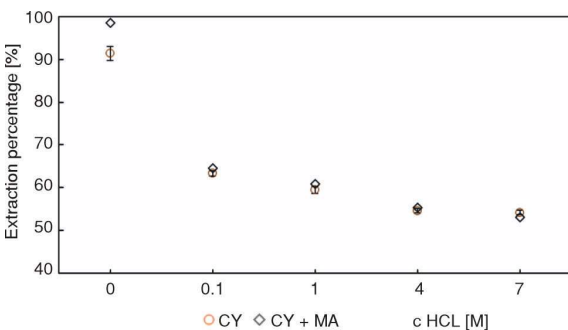


Figure 3. Extraction percentage of U from distilled water and 0.1-7 M HCl by CY, CY + MA

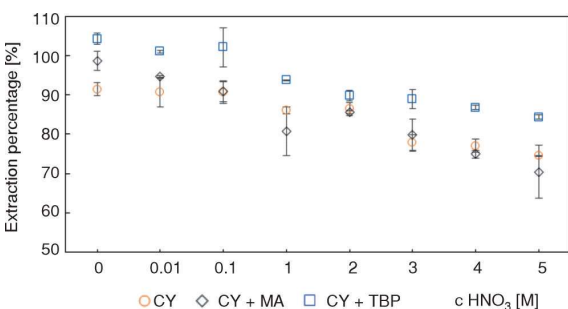


Figure 4. Extraction percentage of U from distilled water and 0.1-7 M HNO₃ by CY, CY + MA, CY + TBP

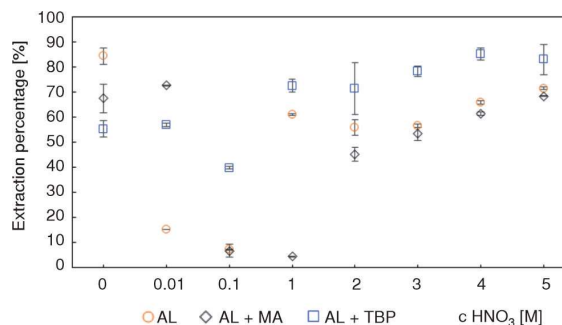


Figure 5. Extraction of Pu from distilled water and 0.01-5 M HNO₃ by AL, AL + MA, AL + TBP

binations. Adding the extractants to the RTIL has a noticeable impact in either increasing or decreasing the extraction percentage compared to the pure RTIL.

Extraction of Pu using AL

Pure AL extractions from HNO₃, fig. 5, reach their maximum extraction percentage of 85 ± 4% from distilled water, then decreases to 7 ± 1% from 0.1M HNO₃ before rising to 61 ± 1% at 1 M and then trending from 56% to 71% from concentrations 2M-5M. The addition of MA provides a slightly smaller efficiency than distilled water at 68 ± 6% increasing the extraction percentage to 73 ± 1% from 0.01M HNO₃ compared to 15 ± 1% of pure AL. This increase indicates the formation of neutral nitrate complexes by MA and Pu extracted by AL. The efficiency drop for AL + MA at 0.1M is comparable to the pure AL, however, the extraction percentage remains low at 5 ± 1% even from 1M HNO₃. From 2M-5M, the efficiency follows a similar increasing trend with slightly smaller values of 45-68%. While the addition of TBP decreases the initial extraction percentages from distilled water and 0.01M HNO₃ to 55 ± 4%, the efficiency drops only to 40 ± 1% at 0.01M compared to 7 ± 1% and 7 ± 3% for pure AL and AL + MA, respectively. The efficiency increases to 73 ± 3% at 1 M and then trends up to 85% from 4-5M HNO₃. The drop for AL + MA from 0.1M and 1M HNO₃ would indicate the formation of a different species than the neutral nitrate complex by MA and Pu, however, TBP achieves extractions over 40% and 70% from 0.1M and 1M HNO₃, respectively, and the extracted species should be similar neutral nitrate complex. Moreover, if a formed species is not extracted by the extractant, it should be extracted by the RTIL itself, therefore reaching at least the efficiency of pure RTIL, which is not the case. This means that the presence of MA decreases the extraction capability of the RTIL. The similar extraction percentages of pure AL and AL + MA above 2M HNO₃ suggest that the predominant mechanism is the anion exchange of the pure AL. The increasing extraction efficiency of TBP with the increasing concentration of HNO₃ was reported by Vasudeva Rao [27] with the expected extracted species

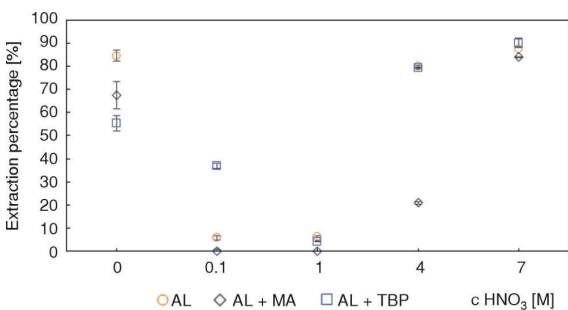


Figure 6. Extraction of Pu from distilled water and 0.1-7 M HCl by AL, AL + MA, AL + TBP

being the neutral nitrate complex. The TBP was used without any additional solvent. However, the reported neutral nitrate complexes at high concentrations of HNO₃ should not be extracted by AL as anion exchange should be the main extraction technique at that acidity range.

The extraction percentages from HCl, fig. 6, are initially similar to those from HNO₃, with pure AL and AL + MA dropping to sub 10% efficiency from 0.1M HCl and AL + TBP dropping to 37 ± 2 % efficiency, indicating that TBP is forming different complexes than MA. However, efficiencies for all three variants are less than 10 % at 1M, with extractions with the AL + MA combination being barely measurable. The efficiencies increase for pure AL and AL + TBP to 80 % range at 4 M, while for AL + MA the efficiency increases only to 21 ± 1 %. The extraction percentages of all three variants are within 80-90 % from 7M HCl. Higher percentages from concentrations of HCl > 4 M than from HNO₃ suggest greater affinity of the AL to the formed anionic chloride complexes than nitrate complexes. The decrease of efficiency in the presence of MA from 0.1-4M HCl below the efficiency of the pure AL can indicate an undesirable interaction of MA with AL or the formation of non-extractable complexes under those conditions.

Extraction of Pu using CY

The extraction of Pu by pure CY or CY + extractants from HNO₃ shows minimal differences in the extraction percentages among the three variants, fig. 7. All three variants maintain efficiency around 100 % from distilled water and 0.01-0.1M HNO₃. The efficiencies drop to 55-65 % at 1M. From 2M, CY + MA drops further to 57 ± 2 % indicating at least partial formation of non-extractable complexes, whereas pure CY increases to 80 ± 2 % and CY + TBP increases to 70 ± 3 %. For concentrations 3M and above, all three variants plateau within 83-91 %.

The most significant difference between pure CY and CY + MA was achieved in extractions of Pu from HCl, fig. 8. While pure CY produces similar extraction percentages to pure AL with maximum efficiency of around 100 % with extractions from distilled water, decrease to sub 10 % efficiency from 0.1M and

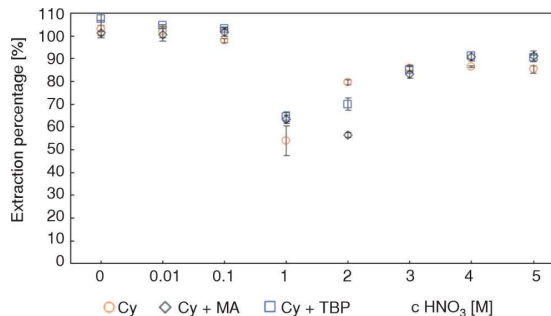


Figure 7. Extraction of Pu from distilled water and 0.01-5 M HNO₃ by CY, CY + MA, CY + TBP

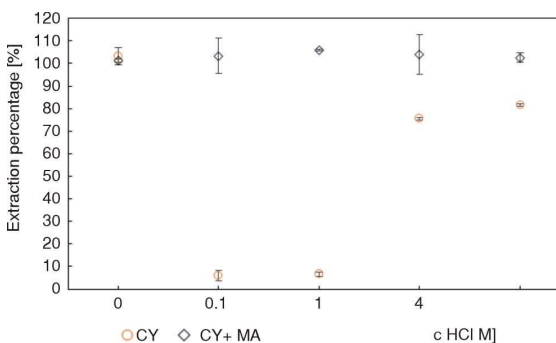


Figure 8. Extraction of Pu from distilled water and 0.1-7 M HCl by CY, CY + MA

1M HCl and then plateau within 75-80 % from 4M-7M, the combination of CY + MA maintains the extraction percentage of 100 % from all concentrations of HCl.

CONCLUSIONS

Whereas americium cannot be extracted by any combination of Cyphos 101 or Aliquat 336 and malonamide or tributyl phosphate in significant amounts, plutonium, and uranium can be extracted with extraction percentages above 80 % with several combinations. Moreover, some combinations can successfully provide separation of the two actinides, namely AL + MA from 1M HNO₃ where the extraction efficiency for plutonium is below 5 % and for uranium over 85 %. On the other hand, plutonium can be extracted with an efficiency of over 80 % with pure AL from distilled water, whereas the efficiency for uranium is below 10 %. The plutonium can be extracted with an efficiency reaching 100 % with CY + MA from HCl of any concentration. The uranium can be extracted with an efficiency reaching 100 % with AL + TBP from 0.1M HNO₃. Extraction percentages of most combinations from distilled water exceeded 50 %, except for extracting uranium with AL and AL + TBP. The extraction mechanism is expected to be anion exchange for acid concentrations above 3M and the formation of neutral complexes below 3M. However, significant irregularities in the extraction percentages, especially with MA as an extracting agent

would require further studies in this area to determine the exact extracted species. While plutonium can be extracted from higher concentrations of acids to limit the passing of the aqueous phase into the RTIL, extraction of uranium decreases with the increasing acid concentrations, making an aqueous phase transfer into RTIL a prevailing issue. Chloride variants of the used RTIL proved advantageous in higher concentrations of acids, as during the anion exchange no irreplaceable part is lost, compared to the loss of cation from imidazolium RTIL during their extraction processes.

AUTHORS' CONTRIBUTIONS

Conceptualization: J. Janda, L. Kanka; analysis and writing: L. Kanka; supervision: J. Janda.

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**ЕКСТРАКЦИЈА ТЕЧНОСТ-ТЕЧНОСТ Am(III), Pu(IV) И U(VI) КОРИШЋЕЊЕМ
МАЛОНАМИДА И ТРИБУТИЛ ФОСФАТА У ЈОНСКИМ ТЕЧНОСТИМА
НА СОБНОЈ ТЕМПЕРАТУРИ КАО РАЗБЛАЖИВАЧУ**

Испитивано је понашање екстракције америцијума, плутонијума и уранијума из азотне или хлороводоничне киселине јонским растворима малонамида или трибутил фосфата у триоктилметиламонијум хлориду (Aliquat 336) или трихексил(тетрадецил)фосфонијум хлориду (Сурphos101). Процент екстракције ових актинида мерен је као функција концентрације азотне или хлороводоничне киселине и постављене комбинације јонске течности на собној температури и екстрактанта. Чисте јонске течности на собној температури могу екстраховати плутонијум и уранијум у различитим степенима, у комбинацијама са екстрактантима који даље утичу на проценте екстракције. Стопостотна ефикасност екстракције плутонијума постигнута је у свим концентрацијама хлороводоничне киселине са комбинацијама Сурphos 101 и малонамида. Такође, стопостотна ефикасност екстракције уранијума постигнута је помоћу Сурphos 101 и малонамида из дестиловане воде. Америцијум није екстрахован било каквом комбинацијом ни под којим условима.

Кључне речи: јонска течност на собној температури, америцијум, плутонијум, уранијум, малонамид, трибутил фосфат, проценат екстракције
