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# Radiolytic behaviour of a TODGA based solvent under alpha irradiation

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**Abstract**
In this work the radiolytic stability of a TODGA based solvent has been investigated in alpha radiolysis measured by solvent extraction distribution ratios. Solutions of TODGA in alkane diluents have been subjected to $^{244}$Cm α-irradiation in the presence of nitric acid (0.5 M) and analysed using radiometric techniques to determine their rates of radiolytic degradation. It was shown that alpha radiolysis, similarly to external gamma irradiation, produces an exponential decrease of the free TODGA concentration of the organic solvent as the solvent is degraded with increasing absorbed alpha dose. The degradation constant, obtained from the exponential decrease, indicates that the free TODGA concentration is halved with every 1.0 MGy deposited to the solvent from internal alpha decay. This result could be useful information when dealing with high activity actinide solutions in TODGA based solvent extraction systems.

**Keywords** Alpha irradiation · Radiolysis · TODGA · Absorbed dose · Solvent extraction · Cm and Am

## Introduction

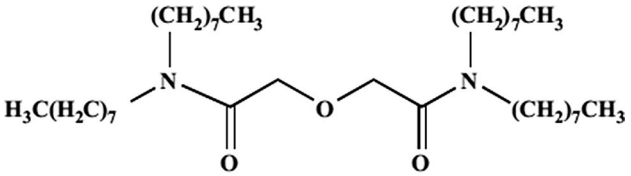
TODGA (N,N,N´,N´-tetraoctyldiglycolamide) (Fig. 1), especially when dissolved in a non- polar diluent and not in contact with a strong acidic system, is hydrolytically stable and has a high selectivity towards trivalent and tetravalent elements [1, 2]. TODGA based extraction systems have been proposed [3, 4] and developed for several nuclear hydrometallurgical partitioning processes [5], focussing mainly on the extraction and recovery of trivalent minor actinides [6, 7]. Recently a TODGA based system was also developed for the grouped actinide extraction process, the so called GANEX process [8–11].

In an extraction process, the solvent becomes highly radioactive when it is contacted with the actinide containing aqueous solution. Radioactive decay of extracted actinides induces damage to the solvent through radiolysis, which generally results in a breakdown of the active extracting molecule and formation of radiolysis products. Radiolysis usually has a negative influence on the performance of the organic solvent as the new species generated very often have extracting properties that markedly differ from those of the original ligands. In the case of TODGA, solvent degradation potentially also leads to enhanced risk of phase instability due to third phase formation [5, 12, 13]. The extraction properties of the solvent can only be maintained if the extractant is reasonable resistant to ionising radiation, delivered primarily through alpha decay of the extracted actinide metals. For extraction process development it is therefore a crucial issue to investigate and understand the radiolytic stability of the solvent.

Several studies have been dedicated to the gamma radiolytic stability of TODGA based solvents [14–17]. Sugo et al. [14] studied the radiolytic degradation of TODGA in n-dodecane based diluents by gamma irradiation of the organic phase. The degradation was found to be exponential versus the absorbed dose and the dose constant [18] strongly dependent on the initial TODGA concentration and the solvent composition. Further investigations with electron pulse irradiations concluded that n-dodecane radicals have a sensitization effect on the TODGA radiolysis [15]. Galán et al. [16] studied the radiolytic stability of external gamma irradiated TODGA/TPH solvents with varying compositions of octanol, uncontacted or pre-equilibrated with 3 mol/L $HNO_3$. A significant decrease of the TODGA concentration was found, especially when TODGA is not pre-equilibrated with $HNO_3$ and for solvent compositions containing octanol. The study went on to structurally identify seven of the main

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TODGA

**Fig. 1** TODGA: *N,N,N´,N* tetraoctyldiglycolamide

degradation products [14] and to prove that, compared to TODGA itself, they possess no or only small capabilities to extract An(III) or Ln(III). Only one study has reported on alpha degradation of the TODGA solvent system. Sugo et al. 2009. [17] compared alpha (by He ion bombardment) and gamma radiolytic degradation of externally irradiated TODGA solvents. However, to the best of our knowledge, radiolytic degradation by internal alpha emitters of a TODGA based solvent system has not yet been reported which has been investigated in this work.

## Materials and methods

### Chemicals

Deionized "MilliQ" water (18.2 MΩ·cm$^{-1}$) was used in all solution preparation. Dilute nitric acid solutions were prepared from concentrated nitric acid (supra pure, Merck) and used for preparation of feed solutions and dilution of samples. Tetraoctyldiglycolamide, TODGA was obtained from Tecnocom LTD, assayed to be of > 98% purity.

### Method

#### Choice of conditions

The organic solvent was prepared by diluting TODGA into n-dodecane to a concentration of 0.2 M. This concentration was chosen based on previous experience as it provides a stable extraction system at the chosen acidity with a well investigated extraction mechanism and stoichiometry [2, 5, 19].

The complexation strength of TODGA is in nitric acid remarkably high and follows for the actinides the order of M(III) ~ M(IV) > M(VI) ≫ M(V) [2, 5, 20]. The distribution ratios (D) of trivalent actinides are significant already at medium acidities and increases strongly with the $HNO_3$ concentration. The efficient extraction of actinides at high acidities makes it difficult to assess the stability of the TODGA/n-dodecane system by measurement of distribution factors. The reason is the low activity in the aqueous phase and the obvious risk of cross-contamination in the sampling procedure. Furthermore, TODGA dissolved in aliphatic systems has a tendency for third phase formation, particularly at high $HNO_3$ concentrations [21–23]. It has been shown that when the equilibrium concentration of nitric acid in the aqueous phase is less than 0.7 M, TODGA exists as a mixture of monomers and dimers [21]. As the aqueous phase acidity is increased, the extractant molecules form higher aggregates which can contain up to eight of seven molecules of TODGA [22, 23]. The formation of the larger TODGA aggregates takes place over the same range of aqueous acidities where the extraction of trivalent f-element cations displays a hyperstoichiometric nitric acid dependence. Acid-driven aggregation of TODGA is likely responsible for the increased phase instability and unusual acid and extractant dependencies observed for the extraction of trivalent metal nitrates at high acidity [21–23]. A lower nitric acidity is therefore preferred for an alpha radiolytic stability study of the TODGA/n-dodecane system based on distribution factors. In this work 0.5 M $HNO_3$ was the chosen acidity.

#### Solvent extraction experiments

In total three 2 mL glass vials with Teflon sealed screwcaps were prepared for the extraction experiments. Two contained high alpha activity for the purpose of radiolysis, the third was prepared with low alpha activity to act as reference. All liquid additions or samplings were gravimetrically done using an analytical balance with a weighing precision of 0.1 mg. In the extraction vials, 750 µL of the organic phase was added together with 750 µL of aqueous phase with an acidity of 0.5 M $HNO_3$. The aqueous phase was either 750 µL of a Cm solution or 50 µL of an $^{241}$Am solution together with 700 µL 0.5 M $HNO_3$ for the reference vial. The radiolysis experiment (Fig. 2) was started by intensely mixing the three extraction vials using a Bioshake IQ (1800 rpm, T = 23 °C) for 1 h. This mixing was followed by sampling of all three extraction vials. In order to ensure perfect phase separation, the extraction vials were centrifuged (1700 g for 10 min) before 50 ul of each phase was removed, weighed and diluted into 950 µL n-dodecane or 1 M $HNO_3$ into sample tubes for gamma measurement. The aqueous sampling had to be especially carefully executed to avoid cross contamination. In total 7 samples (350 µL) could be extracted out of each extraction vial generating a series of 14 samples with incremental absorbed alpha dose over 91 days. The reference extraction vial was sampled every second time. The sample scheme is outlined in Table 1.

#### $^{241}$Am and $^{244}$Cm solutions preparation

A 150 ppm $^{241}$Am solution in 8 M $HNO_3$, initially obtained from purification of old Pu, was converted to 0.5 M $HNO_3$

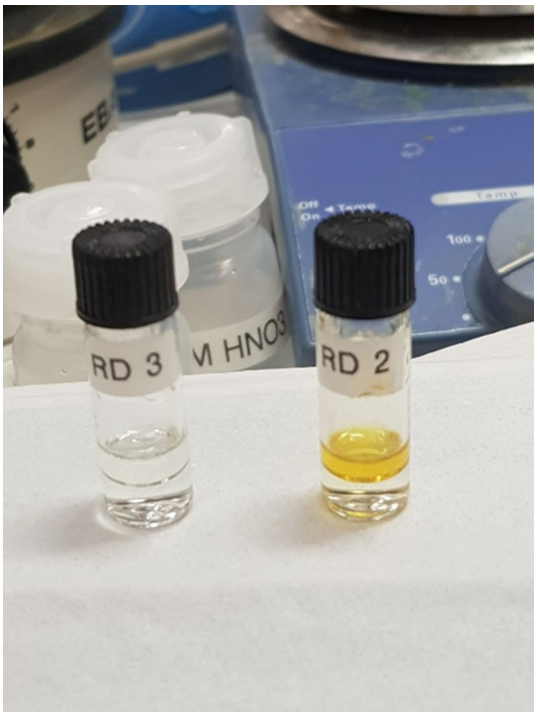



**Fig. 2** Internal alpha irradiation of a 0.2 M TODGA solvent in n-dodecane in contact with 0.5 M $HNO_3$. Left sample: reference (low alpha doses), Right sample: Alpha radiolysis 6.2 MGy

**Table 1** Experimental layout for the radiolysis study

| Time [days] | Org = 0.2 M TODGA<br>Aq = 0.5 M $HNO_3$ | | |
|---|---|---|---|
| | Vial 1 | Vial 2 | Ref |
| 0 | X | X | X |
| 5 | X | | |
| 8 | X | | X |
| 12 | X | | |
| 15 | X | | X |
| 19 | X | | |
| 27 | X | | X |
| 36 | | X | |
| 42 | | X | X |
| 47 | | X | |
| 55 | | X | X |
| 64 | | X | |
| 70 | | X | X |
| 91 | | X | |

by evaporation to dryness in the corresponding acid three times. The solution was gravimetrically diluted, and the specific activity characterised by gamma spectrometry.

The Cm solution contained 0.5 M $NaNO_3$, and was therefore initially purified by extraction chromatography using the DGA branched resin to remove Na. 1.5 mL of the Cm solution was acidified to approximately 3 M $HNO_3$ and added as feed to an extraction chromatography column (Biorad) prepared with DGA branched resin (50 – 100 µm particle size) with a free column volume of ~ 0.2 mL. The Cm was washed with 2 mL 3 M $HNO_3$ followed by 0.5 mL 1 M $HNO_3$ and thereafter eluted into 3 mL 0.01 M $HNO_3$. An americium impurity in the Cm solution enabled to follow the separation by gamma spectrometry. The purified Cm was converted to 0.5 $HNO_3$ by repeated evaporation to dryness. The specific activity of the Cm solution was accurately characterised by Pu reference spiking and a corresponding alpha ratio measurement. With the Cm isotopic vector accurately known, the specific activities of all Cm isotopes could be accurately determined, see Table 2. The alpha emission is totally dominated by $^{244}$Cm (99.5%) and $^{243}$Cm (0.42%). The specific activity of the Am impurity, both $^{241}$Am and $^{243}$Am, was determined by ICP-MS and gamma spectrometry and was high enough to enable gamma measurements of radiolysis series without addition of $^{241}$Am as gamma tracer. The alpha dose rate after extraction of Cm into the organic phase was around 2.8 kGy/h. The dose rate from $^{241}$Am to the organic solvent in the reference samples was $3.4 \bullet 10^{-3}$ kGy/h.

## Radiochemical analysis

Gamma spectrometric measurements of $^{241}$Am were done using a planar HPGe borewell detector (Canberra GCW2022) with an energy resolution of about 1.3 eV (FWHM) at 122 keV. All gamma spectrometry samples (1 mL) were identically prepared by dilution of the original samples into

**Table 2** Main composition of the of the feed $^{241}$Am and Cm solutions

| | Concentration [ppm] | Specific alpha activity [Bq/g] | α-branching | Main α-emission [MeV] |
|---|---|---|---|---|
| Am solution (50 µL added to the reference experiment as gamma tracer) | | | | |
| $^{241}$Am | 140 | $1.80 \times 10^7$ | 1.00 | 5.486, 5.443 |
| Cm solution (750 µL added to the experiment) | | | | |
| $^{243}$Cm | 1.5 | $2.80 \bullet 10^6$ | 0.997 | 5.785, 5.472 |
| $^{244}$Cm | 220 | $6.70 \times 10^8$ | 1.00 | 5.805, 5.763 |
| $^{245}$Cm | 16,53 | $1.1 \times 10^5$ | 1 | 5.362, 5.304 |
| $^{246}$Cm | 26,26 | $3.0 \times 10^5$ | 0.9997 | 5.386, 5.344 |
| $^{247}$Cm | 0,34 | 1.2 | 1 | 4.868, 5.265 |
| $^{248}$Cm | 0,08 | 12 | 0.916 | 5.078, 5.035 |
| $^{241}$Am | 13 | $1.70 \times 10^6$ | 1.00 | 5.486, 5.443 |
| $^{243}$Am | 130 | $1.0 \times 10^6$ | 1.00 | 5.276, 5.233 |

The relative uncertainty in concentration and specific activity is < 11% for $^{241}$Am and $^{243}$Am (evaluated by gamma spectrometry and ICP-MS analyses) and < 1% for the main Cm isotopes (evaluated by Pu/Cm alpha ratio measurements), coverage factor = 2

1 M nitric acid (or 1 mL n-dodecane). A custom made, resin embedded calibration source (Eckert & Ziegler) prepared in the same type of sample tube and geometry as used in the measurements of the samples was used for energy and efficiency calibration of the detector. The given uncertainty for $^{241}$Am (59.6 keV) emission of the source was 5% (k = 1).

For alpha measurements an Ortec duo system was used equipped with passivated ion implanted Si detectors (PIPS, ULTRA-AS) detectors having an active area of 450 mm$^2$ and a specified resolution of 20 keV. The samples were measured at different source to detector distances varying from 10 to 20 mm with counting efficiencies varying from a few to 10-12% at < 1 mbar pressures in the measurement chambers. A standard reference counting source (Eckert & Ziegler) with electrodeposited $^{239}$Pu, $^{241}$Am, and $^{244}$Cm was used for energy and efficiency calibration of the measurement position.

## Results and discussion

The phase stability remained intact during the experiment and samples could easily be removed by pipetting throughout the complete series. The radiolytic degradation of the solvent induced a colour change and for the last sample with approximately 6.2 MGy, the solvent had turned yellow but showed otherwise no indication of damage.

In each sampling the distribution coefficient, $D = C_{Am,org}/C_{Am,aq}$ was calculated from the measured organic and aqueous samples. At low metal loading, the reaction mechanism and the stoichimetry involved in the extraction of a trivalent metal with TODGA is well known [2, 5, 21, 22]. From the extraction reaction mechanism, Eq 1, with $A^- = NO_3^-$ and L = TODGA, the equilibrium constant can be defined according to Eq 2.

$$Am + 3A^- + xL \leftrightarrow AmA_3L_x \tag{1}$$

$$K = \frac{\left[\overline{AmA_3L_2}\right]}{\left[Am^{3+}\right] \cdot [A^-]^3 \cdot [L]^x} \tag{2}$$

where $x$ is the number of TODGA molecules extracted per metal which for Am(III) under the chosen conditions takes a number between 3 and 4 [2]. Defining $D = C_{Am,org}/C_{Am,aq} = [AmA_3L_x]/[Am^{3+}]$, Eq. 2 can be rearranged and the distribution coefficient expressed as a function of nitrate $[A^-]$ and free ligand concentration, $[L]$

$$D = \frac{\left[\overline{AmA_3L_2}\right]}{\left[Am^{3+}\right]} = K \cdot [A^-]^3 \cdot [L]^x \tag{3}$$

Equation 3 shows that the distribution coefficient is at constant acidity proportional to the free ligand concentration. During the experiment the concentration of free ligand will decrease as a result internal alpha radiolysis of the organic phase. Under the assumption that radiolysis products of TODGA do not significantly compete are incapable to form extractable complexes with Am [16] and that the acidity remains constant, the measured distribution coefficient is directly related to the concentration decrease of TODGA which in turn is induced by radiolytic damage from alpha decay of Cm and Am extracted into the organic solvent.

The estimated range of $^{241}$Am or $^{244}$Cm alpha particles in the organic solvent is around 35 µm [24]. It is a reasonable assumption that the full energy of each alpha decay is deposited in the organic solvent. The absorbed alpha doses [J/kg] over time to the organic solvent can thus be calculated from the specific alpha activity, the measured distribution coefficient and the time, according to:

$$AbsDose\left[\frac{\mathrm{J}}{\mathrm{kg}}\right] = \sum_{i=241}^{248} S_i \cdot \left(\frac{D_t}{D_t + 1}\right) \cdot r \cdot E_\alpha \cdot 1.602 \cdot 10^{-19} \cdot t \cdot 1000 \tag{4}$$

where $S_i$ = specific activity of Cm or Am isotope in the aqueous phase (i) [Bq/g], $D_t$ = distribution coefficient at sampling time t, calculated as the average D of the previous sample and the sample at time t, $r = \rho_{aq}/\rho_{org}$ density ratio aqueous to organic phases = 1.25, $E_a$ the average alpha decay energy [eV], 1.602e−19 [J/eV], $t$ = irradiation time [s]. The expression involving the distribution coefficient is the fraction of the activity extracted into the organic phase. The average D is an attempt to compensate the calculated absorbed doses for the decrease in distribution coefficient and the corresponding decreasing dose-rate during the irradiation time in between the samples.

Figure 3 shows the distribution coefficient as a function of the absorbed alpha dose to the solvent. An exponential decrease with the absorbed alpha doses is clearly seen. The distribution coefficients obtained from reference samples seem to initially decrease and to reach a stable level at a somewhat lower value. This behaviour is not easily

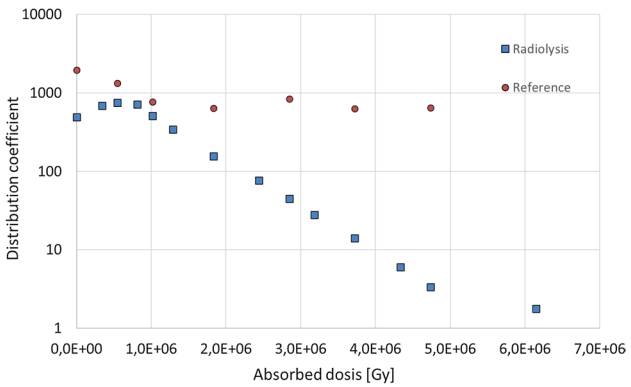



**Fig. 3** The distribution coefficients vs absorbed alpha dose [Gy] in the TODGA solvent

explainable as a continuous decrease of the distribution coefficients is to be expected if the stability of TODGA is influenced from other parameters than radiolysis over the experiment period of 91 days. Radiolytic degradation can be excluded as the total absorbed alpha dose to the solvent of the reference sample is low with a maximum of 5 kGy calculated for the last reference point. It is likely that the results are influence by experimental uncertainties. The reference sample series nevertheless show that that the stability of the extraction system towards acid hydrolysis or oxidation is high enough not to significantly influence the results of the radiolysis experiment. This is in agreement with findings in previous studies [14–17]. The initial 3 points from the radiolysis series were found to be affected by cross-contamination and the sampling procedure was further optimised to avoid this problem. With the general high solution activity in this experiment and the obvious risk of cross-contamination as the low activity aqueous sample is removed by pipetting through the high activity organic phase, an upper limit of around $10^3$ in determination of distribution coefficients is probably reasonable.

The last point deviates from the trend. In the end of the experiments most TODGA molecules in the organic solvent have been inactivated by radiolysis. Weak extraction of Am has been shown by one of the radiolysis products of TODGA [14, 16]. A significant effect on the distribution coefficient trend can only be seen when the extraction of Am starts to be dominated by a radiolysis product of TODGA instead of TODGA itself.

The concentration decrease of TODGA due to radiolysis is exponential as the rate of radiolysis is dependent on the TODGA concentration [14, 16, 18]. The radiolysis rate equation is defined as

$$-\frac{dC_{TODGA}}{dD} = C_{TODGA} \cdot k \tag{5}$$

where $C_{\mathrm{TODGA}}$ is the organic concentration [M], $D$ = absorbed alpha dose [Gy] and $k$ is the radiolysis degradation rate constant [$\mathrm{Gy}^{-1}$]. The solution to the rate equation is

$$C_{TODGA} = C^0_{TODGA} \cdot e^{-kD} \tag{6}$$

or rearranged as the concentration ratio;

$$\ln\left(\frac{C_{TODGA}}{C^0_{TODGA}}\right) = -k \cdot D \tag{7}$$

With an initial TODGA concentration, $C^0_{\mathrm{TODGA}}$, of 0.2 M, the TODGA concentration as a function of D can be calculated, through Eq [3]. In Fig. 4, the logarithm of the TODGA concentration ratio, $\ln(C/C^0)$, has been plotted against the total absorbed alpha dose. The initial three points of the radiolysis

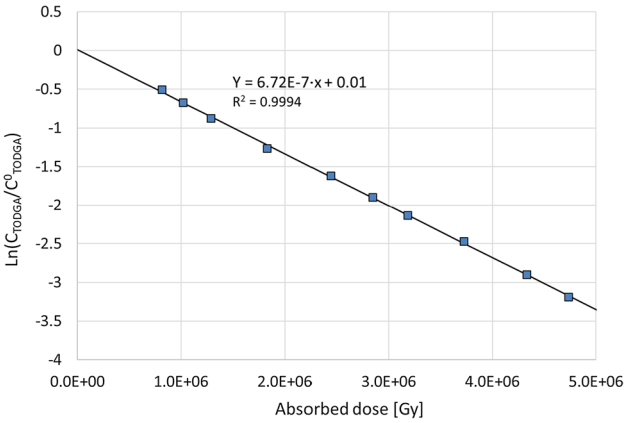



**Fig. 4** Alpha radiolysis of TODGA solvent. Concentration ratio as a function of absorbed dose to the solvent

series have been removed as they are likely affected by cross contamination and the first point replaced with the distribution coefficient of the first reference value. An exponential decrease, according to Eq 6, is clearly observed. A least square fitting of the data points, excluding the last point, gives the pseudo first order degradation rate or dose constant [19] of $k = 6.7 \times 10^{-7}$ [$\mathrm{Gy}^{-1}$]. The intercept value very close to 0 indicates that the distribution coefficient of the first reference point added to the curve is probably correct.

The dose rate constant of $6.7 \times 10^{-7}$ $\mathrm{Gy}^{-1}$ compares well to $2.2 \times 10^{-6}$ $\mathrm{Gy}^{-1}$ obtained by Sugo et al. [17] in gamma radiolysis of non-acid equilibrated 0.2 M TODGA dissolved in n-dodecane. The lower value in this work is because nitric acid has a stabilising effect on the radiolysis of TODGA [16]. Sugo et al. [17] compared alpha (by He ion bombardment) and gamma radiolytic degradation of externally irradiated TODGA solvents not equilibrated with nitric acid. In this study $G$ values of 0.022 and 0.08 [μ mol $\mathrm{J}^{-1}$] were reported for alpha and gamma irradiation, respectively which can be recalculated to approximately $1.8 \times 10^{-6}$ [$\mathrm{Gy}^{-1}$] and $6.4 \times 10^{-6}$ [$\mathrm{Gy}^{-1}$]. The TODGA concentration and radiolysis products were in both cases analytically determined, and the radiolysis products found to be different between alpha and gamma radiolysis. It was concluded that TODGA has an approximate four-fold higher resistance to alpha compared to gamma irradiation. The higher resistance to alpha radiolysis is a conclusion which was also found in a similar study although involving a different solvent system [25]. The denser radiation tracks involved in higher LET radiation (alpha) allows radical recombination to a higher degree than for gamma radiation. It is therefore likely that alpha radiolytic degradation of TODGA solvents is seriously overestimated when the radiolytic stability is investigated by gamma radiation.

Investigation of radiolytic degradation by internal alpha emitters (compared to He bombardment) has the advantage

that the specific activities of the alpha emitting nuclides can be well characterised and thus also the absorbed alpha dose over time. For a TODGA based organic solvent equilibrated with nitric acid the radiolytic degradation is exponential with a dose constant of $6.7 \times 10^{-7}$ $Gy^{-1}$. This value indicates good stability of TODGA towards alpha radiolytic degradation. Through the dose constant and Eq. 5, it is possible to estimate the number of molecules that are destroyed per alpha decay. At the start of the experiment when the TODGA concentration is 0.2 M, around $2.4 \times 10^5$ TODGA molecules are destroyed per alpha decay. The absorbed doses required to reduce the TODGA concentration to a factor 2 is around 1.0 MGy.

## Conclusion

The radiolytic stability of 0.2 M TODGA dissolved in n-dodecane was investigated by internal alpha radiolysis over a 3 months period at a dose rate of 2.8 kGy/h. The distribution ratio of Cm(III) and Am(III) decreased exponentially with increased radiation dose up to ca. 4700 kGy. Where most of the TODGA molecules have been destroyed by alpha radiolysis. The exponential decrease of the distribution coefficients is a strong indication that extraction of minor actinides by products formed in the radiolysis of TODGA is insignificant. The degradation constant, $6.7 \times 10^{-7}$ $Gy^{-1}$ as evaluated from the exponential decrease, proves good alpha radiolytic stability of TODGA based solvents. The constant indicates that the free TODGA concentration is halved with every 1.0 MGy deposited by internal alpha decay.

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## References

1. Sasaki Y, Sugo Y, Suzuki S, Tachimori S (2001) The novel extractants, Diglycolamides, for the extraction of lanthanides and actinides in $HNO_3$–N-Dodecane system. Solvent Extr Ion Exch 19(1):91–103
2. Ansari SA, Pathak P, Mohapatra PK, Manchanda VK (2012) Chemistry of diglycolamides: promising extractants for actinide partitioning. Chem Rev 112(3):1751–1772
3. Modolo G, Asp H, Schreinemachers C, Vijgen H (2007) Development of a TODGA based process for partitioning of actinides from a PUREX Raffinate Part I: batch extraction optimization studies and stability tests. Solvent Extr Ion Exch 25(6):703–721
4. Ansari SA, Pathak PN, Manchanda VK, Husain M, Prasad AK, Parmar VS (2005) N, N, N′, N′-Tetraoctyl Diglycolamide (TODGA): a promising extractant for actinide-partitioning from high-level waste (HLW). Solvent Extr Ion Exch 23(4):463–479
5. Whittaker D, Geist A, Modolo G, Taylor R, Sarsfield M, Wilden A (2018) Applications of Diglycolamide based solvent extraction processes in spent nuclear fuel reprocessing, Part 1: TODGA. Solv Extr Ion Exchange 36:223–229
6. Magnusson D, Christiansen B, Malmbeck R, Glatz J-P, Modolo G, Serrano-Purroy D, Sorel C (2009) Demonstration of a TODGA based extraction process for the partitioning of minor actinides from a PUREX Raffinate—part III: centrifugal contactor run using genuine fuel solution. Solv Extr Ion Exchange 27:26–35
7. Modolo G, Wilden A, Kaufholz P, Bosbach D, Geist A (2014) Development and demonstration of innovative partitioning processes (I-Sanex and 1-Cycle SANEX) for actinide partitioning. Prog Nucl Energy 72:107–114
8. Adnet J-M, Miguirditchian M, Hill C, Heres X, Lecomte M, Masson M, Brossard P, Baron P (2005) Development of new hydrometallurgical processes for actinide recovery: GANEX concept. In: Proceedings of GLOBAL, Atomic Energy Society of Japan
9. Taylor R, Carrott M, Galan H, Geist A, Hères X, Maher C, Wilden A (2016) The EURO-GANEX process: current status of flowsheet development and process safety studies. Procedia Chem 21:524–529
10. Malmbeck R, Magnusson D, Bourg S, Carrott M, Geist A, Hérès X, Wilden A (2019) Homogenous recycling of transuranium elements from irradiated fast reactor fuel by the EURO-GANEX solvent extraction process. Radiochim Acta 107:917–929
11. Bell K, Carpentier C, Carrotta M, Geist A, Gregsona C, Hérèsc X, Magnusson D, Malmbeck R, McLachlan F, Modolo G, Müllich U, Sypula M, Taylor R, Wilden A (2012) Progress towards the development of a new GANEX process. Procedia Chem 7:392–398
12. Ravi J, Venkatesan KA, Antony MP, Srinivasan TG, Vasudeva Rao PR (2015) Solvent extraction behavior of trivalent metal ions in diglycolamides having same carbon to oxygen ratio. Sep Sci Technol 51(1):32–40
13. Tachimori S, Sasaki Y, Suzuki SI (2002) Modification of TODGA-n-dodecane solvent with a monoamide for high loading of lanthanides (III) and actinides (III). Solvent Extr Ion Exch 20(6):687–699
14. Sugo Y, Sasaki Y, Tachimori S (2002) Studies on hydrolysis and radiolysis of N, N, N′, N′-tetraoctyl-3-oxapentane-1,5-diamide. Radiochim Acta 90:161–165
15. Sugo Y, Izum Y, Yoshida Y, Nishijima S, Sasaki Y, Kimura T, Sekine T, Kudo H (2007) Influence of diluent on radiolysis of amides in organic solution. Radiat Phys Chem 76:794–800
16. Galán H, Nunez A, Sedano R, Espartero AG, Durana A, Mendoza JD (2012) Radiolytic stability of TODGA: characterization of degraded samples under different experimental conditions. Procedia Chem 7:195–201
17. Sugo Y, Taguchi M, Sasaki Y, Hirota K, Kimura T (2009) Radiolysis study of actinide complexing agent by irradiation with helium ion beam. Radiat Phys Chem 78:1140–1144
18. Mincher BJ, Curry RD (2000) Considerations for choice of a kinetic figure of merit in process radiation chemistry for waste treatment. Appl Radiat Isot 52(2):189–199
19. Weßling P, Trumm M, Geist A, Panak PJ (2018) Stoichiometry of An(iii)–DMDOHEMA complexes formed during solvent extraction. Dalton Trans 47(32):10906–10914
20. Zhu Z-X, Sasaki Y, Suzuki H, Suzuki S, Kimura T (2004) Cumulative study on solvent extraction of elements by N, N, N′, N′-Tetraoctyl-3-Oxapentanediamide (TODGA) from nitric acid into N-Dodecane. Anal Chim Acta 527(2):163–168
21. Yaita T, Herlinger AW, Thiyagarajan P, Jensen MP (2004) Influence of extractant aggregation on the extraction of trivalent f-Element cations by a Tetraalkyldiglycolamide. Solv Extr Ion Exchange 22(4):553–571
22. Singh MB, Patil SR, Lohi AA, Gaikar VG (2018) Insight into nitric acid extraction and aggregation of N, N, N', N'-Tetraoctyl diglycolamide (TODGA) in organic solutions by molecular dynamics simulation. Sep Sci Technol 53(9):1361–1371

23. Pathak PN, Ansari SA, Kumar S, Tomar BS, Manchanda VK (2010) Dynamic light scattering study on the aggregation behaviour of N, N, N′, N′-tetraoctyl diglycolamide (TODGA) and its correlation with the extraction behaviour of metal ions. J Colloid Interface Sci 342(1):114–118
24. Magill J, Dreher R, Ünlü K, Heller-Zeisler S, Zeisler R (2009) The NUCLEONICA nuclear science portal. In: AIP conference proceedings
25. Magnusson D, Christiansen B, Malmbeck R, Glatz JP (2009) Investigation of the radiolytic stability of a CyMe4-BTBP based SANEX solvent. Radiochim Acta 97:497–502

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