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Effect of α-Radiolysis on TRUEX-NPH Solvent

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ABSTRACT

An unexpectedly high degradation of the TRUEX (TRansUranic EXtraction) solvent occurred during the treatment of waste solutions from the New Brunswick Laboratory. The waste solutions treated contained approximately 1 g/L of Pu-239 and 20 mg/L of Am-241. Earlier studies of α-radiolysis using carbon tetrachloride rather than normal paraffinic hydrocarbons (NPH) as a diluent indicated greater resistance to radiation damage than observed. For this study, the TRUEX-NPH solvent was loaded with Am-241 in nitric acid, irradiated with doses up to 3.5 Mrad, and monitored for decline in extraction capability as a function of absorbed dose. Results of this study are being used to improve the Generic TRUEX Model, a thermodynamic model that permits flow-sheet design for solvent extraction processing.

INTRODUCTION

For the past decade the TRUEX (TRansUranic EXtraction) solvent extraction process has been used to remove transuranic radionuclides from aqueous solutions. During this period the concentrations of the extractants and the diluent have been changed to produce a more benign organic solvent. The radiation damage to the solvent is expected to vary with different formulations. Earlier radiolysis studies (1–5) using different diluents or a high-intensity ^{60}Co γ-ray source indicated that radiolysis damage should be inconsequential for the radiation doses typically experienced. Nonetheless, the TRUEX-NPH (NPH = normal paraffinic hydrocarbons) solvent underwent unexpectedly high degradation during the treatment of waste solutions from the New Brunswick Laboratory (6). This finding motivated the present examination of α-radiolysis of the TRUEX-NPH solvent. The solvent was loaded with ^{241}Am in nitric acid solution, irradiated with doses up to 3.5 Mrad, and monitored for changes in extraction capability as a function of absorbed dose. The capacity of the TRUEX-NPH solvent to extract and be stripped of ^{241}Am did not change appreciably for the doses used. Low levels of plutonium in the ^{241}Am stock solution were extracted by the solvent but resisted removal. The gradual accumulation of nonamericium (i.e., plutonium) activity and its resistance to stripping from the solvent present the greatest obstacles in TRUEX processing.

BACKGROUND

The TRUEX solvent uses a neutral bifunctional organophosphorous extractant, octyl (phenyl) - *N*, *N*- diisobutylcarbamoylmethylphosphine oxide (CMPO), and a phase modifier, tributyl phosphate (TBP), in an organic diluent. Descriptions of the structure (Fig. 1) and chemistry of CMPO are in the literature (1, 2, 7). In an effort to eliminate chlorocarbons from waste processing, NPH has replaced carbon tetrachloride (CCl_4) and tetrachloroethylene (TCE) as the diluent.

The radiolytic degradation of TBP is well documented (8). Analysis of the degradation products of TBP from the γ-radiolysis of TRUEX-TCE (4) indicated that only dibutyl phosphoric acid (HDBP) and small amounts of monobutyl phosphoric acid are produced. Numerous degradation products of CMPO from the γ-radiolysis of TRUEX-CCl_4 and TRUEX-TCE have been observed (2, 4). The chemical bonds most often severed are those with the N bonds, the P—CH_2 bond, and the CH_2—CO bond. The hydrocarbon chains, P=O bond, and C=O bond are least likely to suffer radiolytic damage.

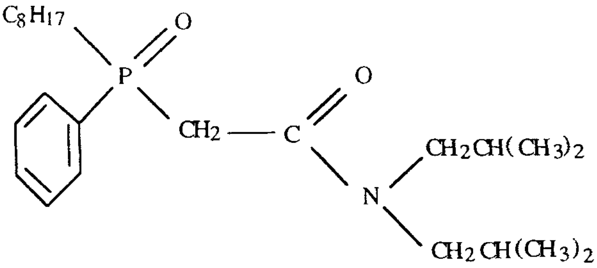

FIG. 1 Octyl(phenyl)-*N*,*N*-diisobutylcarbamoylmethylphosphine oxide (CMPO).

High-intensity ^{60}Co γ-ray sources are commonly used in radiolysis studies. Dose rates of 1 Mrad/h can deliver large doses in a relatively short time. These doses can be achieved without activating the sample, allowing in-depth chemical analysis of the irradiated solvents. Unfortunately, the energy deposition of α-particles emitted in solution by transuranic (TRU) radionuclides is vastly different from that of a ^{60}Co source. A 5-MeV α-particle deposits all of its kinetic energy over a path less than 50 μm, while the TRUEX solvent is essentially transparent to the 1173 and 1332 keV γ-rays emitted by ^{60}Co. The high linear energy transfer (LET) of α-particles concentrates the damage in the vicinity of the TRU radionuclide. Thus, decay of ^{241}Am bound to CMPO is likely to significantly damage the extractant. Loading the TRUEX solvent with an α-particle emitter is the only accurate way to assess the radiolytic damage encountered by the solvent.

The dose absorbed by the solvent is delivered by α-particles emitted by the ^{241}Am. It is reasonable to assume that all the kinetic energy from the α-particles is deposited in the solvent and to ignore the small dose contribution from ^{241}Am γ-ray emission. The rate of energy deposition is easily calculated from the specific activity of the ^{241}Am (7.8 × 10^{11} dpm/L), the kinetic energy of the alpha particle (5.48 MeV), and standard conversion factors:

$$\left(7.8 \times 10^{11}\,\frac{\text{dpm}}{\text{L}}\right)\left(\frac{5.48\ \text{MeV}}{\text{decay}}\right)\left(\frac{1.602 \times 10^{-13}\ \text{J}}{1\ \text{MeV}}\right)\left(\frac{1\ \text{min}}{60\ \text{s}}\right) = 0.0115\,\frac{\text{W}}{\text{L}} \tag{1}$$

The flow-sheet design of the TRUEX process employs the convention of expressing doses in W·h/L. The expression for power dissipation per unit volume of solvent (Eq. 1) does not account for differences in the densities of specific diluents. Incorporating the density of the diluents for TRUEX-CCl_4 and TRUEX-NPH (1420 and 850 g/L, respectively) yields significantly different dose rates:

$$\left(0.0115\,\frac{\text{J}}{\text{s·L}}\right)\left(\frac{\text{L}}{1.42\ \text{kg}}\right)\left(\frac{3600\ \text{s}}{\text{h}}\right) = 29.2\,\frac{\text{J}}{\text{kg·h}} = 2920\,\frac{\text{rad}}{\text{h}} \tag{2}$$

$$\left(0.0115\,\frac{\text{J}}{\text{s·L}}\right)\left(\frac{\text{L}}{0.85\ \text{kg}}\right)\left(\frac{3600\ \text{s}}{\text{h}}\right) = 48.7\,\frac{\text{J}}{\text{kg·h}} = 4870\,\frac{\text{rad}}{\text{h}} \tag{3}$$

Comparing the actual dose rates to TRUEX-CCl_4 (Eq. 2) and TRUEX-NPH (Eq. 3) indicates that the latter receives 67% more dose. Although doses will be given in W·h/L throughout this paper, traditional dose units can be computed using the conversion 1 W·h/L = 0.424 Mrad.

Four batches of waste solution from the New Brunswick Laboratory (NBL) and Argonne National Laboratory were treated by the TRUEX process over a 3-year period. These solutions contained approximately 1 g/L of ^{239}Pu and 20 mg/L of ^{241}Am. In addition to these four waste solutions, a large volume of waste containing much lower plutonium concentrations was treated using the TRUEX process. During one such run where acidified carbonate solutions were being processed, the process had to be shut down shortly after start-up because the solvent exiting the solvent cleanup section had changed from clear and slightly yellow to cloudy and a dark yellow-orange. The solvent also appeared to be more viscous, and samples counted by liquid scintillation showed elevated α-activity. When attempts to strip the solvent proved ineffective, the solvent was replaced with a freshly prepared solution, and the run was successfully completed. In addition, a 6 M HNO_3 solution containing a layer of TRUEX-NPH solvent, used to decontaminate glovebox equipment, was stored in the glovebox for an extended time (6–10 months); the solvent contained approximately 46 g/L plutonium. The organic in this sample had turned gelatinous and a dark brown-red.

The severe degradation of the loaded TRUEX-NPH was unexpected because the doses delivered to it were modest compared to those delivered in earlier γ-radiolysis studies (1, 2, 4, 5). These γ-radiolysis studies did not gelatinize the TRUEX solvent.

Data from the earlier radiolysis studies were used to predict solvent degradation with the generic TRUEX model (GTM), a thermodynamic model that permits flow-sheet design for solvent extraction processing (9). The inability of the GTM to account for the degradation of TRUEX-NPH in the first batch of NBL waste motivated this investigation of the α-radiolysis of TRUEX-NPH.

In the remainder of this paper the partitioning of ^{241}Am between the organic and aqueous phases of a solvent extraction system is expressed in terms of the distribution ratio D_{Am}:

$$D_{\mathrm{Am}} = \frac{\text{concentration of Am-241 in organic phase}}{\text{concentration of Am-241 in aqueous phase}} \tag{4}$$

The concentration of nitric acid in the aqueous phase strongly affects D_{Am} and was varied to effect the loading and stripping of ^{241}Am from the organic solvent. The distribution ratio of TRUEX-NPH in contact with 2.0 M HNO_3 ($D_{\mathrm{Am}}^{2.0} = 29$) allows ^{241}Am to be loaded into the organic phase from an acidic waste solution. We expected that $D_{\mathrm{Am}}^{2.0}$ will decrease as the radiation dose increases due to degradation of CMPO. Stripping of ^{241}Am from the organic solvent is typically achieved in multistage washes with 0.05 M HNO_3 ($D_{\mathrm{Am}}^{0.05} = 0.23$). The change in $D_{\mathrm{Am}}^{0.05}$ as the solvent receives an α-dose is generally small. Destruction of CMPO is balanced by buildup of acidic degradation products, such as HDBP, which are strong extractants of actinides. Changes in the distribution ratio of solvent contacted with 0.01 M HNO_3 ($D_{Am}^{0.01}$ = 0.01) are likely to be most dramatic as the buildup of strongly extracting degradation products dominates the chemistry.

EXPERIMENTAL

The TRUEX-NPH solvent used in these experiments is 0.2 M CMPO and 1.4 M TBP diluted in normal dodecane. The distribution ratios of ^{241}Am (D_{Am}) with this solvent and 0.01, 0.05, and 2.0 M HNO_3 are 0.01, 0.23, and 30, respectively, which are acceptable for our purposes (5).

The ^{241}Am stock solution was prepared from a solution of mixed TRU isotopes. The original solution was acidified to 8 M HNO_3 and twice run through a Dowex MP-1 anion resin column. The resulting solution was evaporated to dryness, and the ^{241}Am was then resuspended in 0.5 M HNO_3. The stock solution was assayed by alpha spectroscopy to contain 3.0×10^{10} dpm/mL of ^{241}Am at >99.9% purity. Concentrations of Pu, Np, and ^{243}Am were below detection limits of the α-spectrometer (C. J. Conner, Argonne National Laboratory, personal communication, 1994).

The TRUEX-NPH solvent was preequilibrated with 2 M HNO_3 prior to loading it with ^{241}Am. Culture tubes were filled with 4.0 mL of TRUEX-NPH and 0.5 mL aliquots of 2 M HNO_3 containing 6.3×10^9 dpm/mL of ^{241}Am. The samples were mixed vigorously for 60 seconds and then placed in a rack behind shielding until they were sampled. The specific activity of the loaded solvent was 7.8×10^8 dpm/mL.

Two tubes were sampled at 3, 6, 9, 11, 15, 22, and 30 days. (Radiation doses appear in Table 1.) Both 4.0 mL samples were washed three times with 2.0 mL of fresh water. One of these samples was scrubbed three times with 2.0 mL portions of 0.25 M Na_2CO_3, followed by five washes with 2.0 mL of water. From each of the samples, three 1.0 mL aliquots were withdrawn and placed in new culture tubes. The sets of three aliquots were preconditioned with two 3.0 mL portions of HNO_3, one each of 0.01, 0.05, and 2.0 M HNO_3. All the wash, scrub, and preconditioning solutions were collected for recovery of ^{241}Am. A final addition of 1.0 mL of fresh HNO_3 of appropriate concentration was placed in each sample, mixed vigorously, and allowed to equilibrate overnight. The samples scrubbed with Na_2CO_3 and the 0.01 M sample were spiked with approximately 10^6 dpm of ^{241}Am to facilitate accurate distribution ratio measurements. The 0.05 and 2.0 M HNO_3 samples not scrubbed with Na_2CO_3 retained sufficient americium from the irradiated solvent and did not re-

TABLE 1
Radiation Doses Deposited in TRUEX-NPH by Alpha Decay of ^{241}Am. Doses Are Based on a Specific Activity of 7.8 × 10^8 dpm/mL of ^{241}Am in 4.0 mL Samples of Solvent

		TRUEX-NPH dose	
Irradiation (days)	Time (hours)	(W·h/L)	(Mrad)
3	72	0.83	0.35
6	144	1.65	0.70
9	216	2.48	1.05
11	264	3.03	1.32
15	360	4.13	1.75
22	528	6.06	2.57
30	720	8.26	3.51

quire a spike of fresh ^{241}Am to measure D_{Am}. Aliquots were removed from all samples the following day and counted for α-activity with a Packard Tricarb 220 liquid scintillation (LS) system and for the 60 keV ^{241}Am γ-ray with a Canberra high purity germanium (HPGe) well detector. The aliquots varied in size from 5 to 50 μL, depending on the activity of the aqueous and organic phases.

One extra sample was initially loaded in case a culture tube failed during the sampling schedule. When scrubbing the sample with 0.25 M Na_2CO_3 to recover the americium during cleanup at the completion of irradiations, the solvent suddenly became a gelatinous yellow foam similar to that encountered during the processing of the first NBL batch. This sample received a dose of approximately 20 W·h/L.

RESULTS AND DISCUSSION

As in the previous radiolysis studies of TRUEX solvent, D_{Am} was not strongly affected by the acid concentrations normally used in waste processing, 0.05 and 2.0 M HNO_3. A steady increase in $D^{0.01}_{\mathrm{Am}}$ was observed with increasing dose and was attributed to buildup of ^{241}Am in the organic phase. Scrubbing the irradiated solvent with 0.25 M Na_2CO_3 removed many of the acidic degradation products and most of the ^{241}Am from the loaded solvent. The carbonate scrub was less effective in removing plutonium from the irradiated solvent. Comparison of the α-activity with the ^{241}Am γ-activity showed that significant nonamericium (i.e., plutonium) α-activity was present in all the aqueous and organic phases. The plutonium concentrations were highest for nonscrubbed samples and increased as nitric acid strength in the contacts increased.

The measured D_{Am} values for TRUEX-NPH not scrubbed with carbonate are graphed versus absorbed dose for contacts with 0.01, 0.05, and 2.0 M HNO_3 in Figs. 2, 3, and 4, respectively. Also plotted are the curves derived from γ-radiolysis of TRUEX-NPH (5). Although these curves are skewed by the high doses (10–200 W·h/L) and wide scatter of the data from Simonzadeh et al. (5), they represent the existing model for radiolysis of TRUEX-NPH. The retention of ^{241}Am in the organic phase of the 0.01 and 0.05 M HNO_3 contacts (Figs. 2 and 3) elevated the D_{Am} values compared to fresh solvent and indicated the need to employ a carbonate scrub for stripping TRU from the solvent. The samples preequilibrated and back-extracted with 2.0 M HNO_3 retained most of their α-activity and required aliquot dilution by a factor of 100 before counting. The presence of TRU complexing degradation products and/or impurities soluble in aqueous acidic solutions is likely responsible for the lower-than-expected D_{Am} for the 2.0 M HNO_3 contact.

The carbonate scrub removed most of the ^{241}Am initially loaded into the solvent, yielding much lower ^{241}Am and total α-activity levels in the contact solutions used to determine D_{Am}. The organic phase from the 0.01 M HNO_3 contact contained little ^{241}Am and required 50 μL aliquots to obtain γ-ray peaks with the HPGe detector. However, the liquid scintillation counting system detected significant α-activity in this phase, indicat-

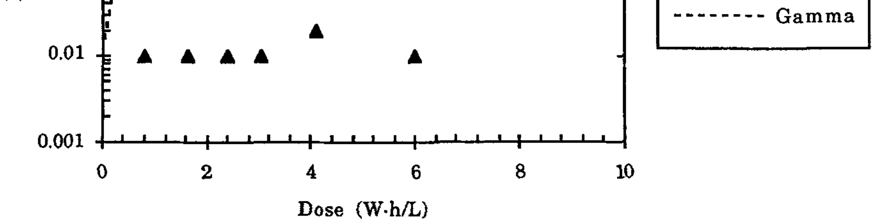

FIG. 2 Change in D_{Am} with dose for 0.01 M HNO_3 and nonscrubbed TRUEX-NPH. The α-radiolysis samples were counted for α-activity using liquid scintillation (LS) and for ^{241}Am γ-activity using a high-purity germanium detector (HPGe). The (gamma) curve is based on the γ-radiolysis of TRUEX-NPH (5).

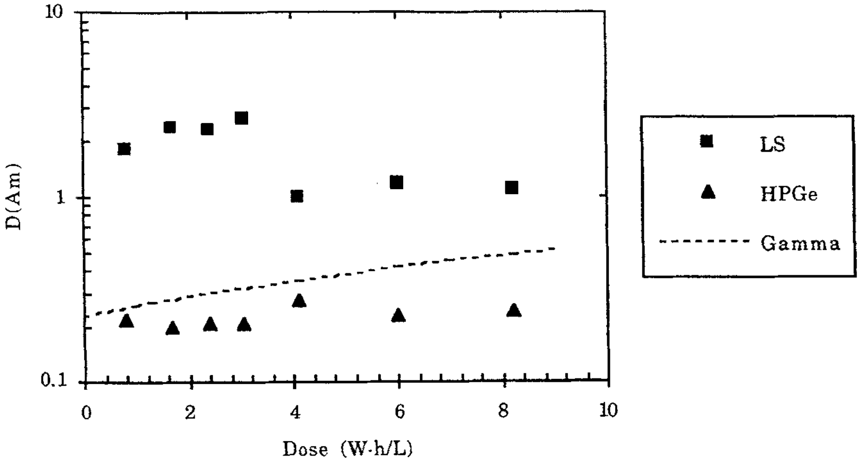

FIG. 3 Change in D_{Am} with dose for 0.05 M HNO_3 and nonscrubbed TRUEX-NPH. The α-radiolysis samples were counted for α-activity using liquid scintillation (LS) and for ^{241}Am γ-activity using a high-purity germanium detector (HPGe). The (gamma) curve is based on the γ-radiolysis of TRUEX-NPH (5).

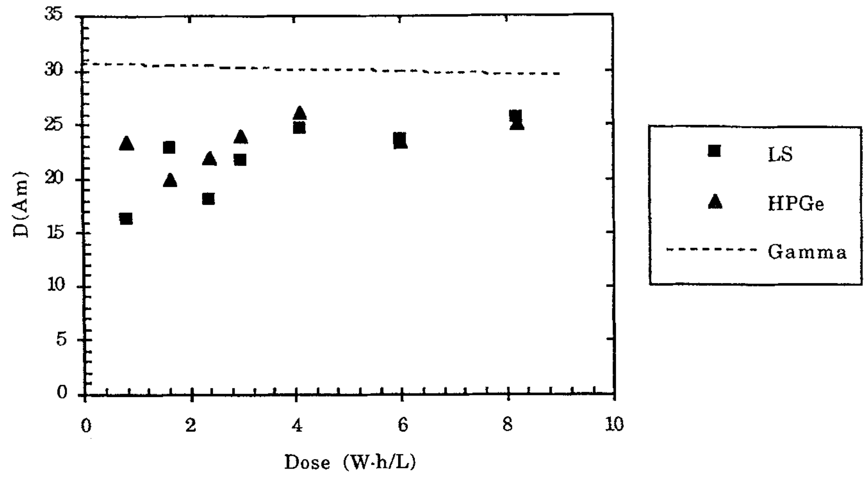

FIG. 4 Change in D_{Am} with dose for 2.0 M HNO_3 and nonscrubbed TRUEX-NPH. The α-radiolysis samples were counted for α-activity using liquid scintillation (LS) and for ^{241}Am γ-activity using a high-purity germanium detector (HPGe). The (gamma) curve is based on the γ-radiolysis of TRUEX-NPH (5).

ing that plutonium remained in the irradiated solvent after the carbonate scrub. As shown in Fig. 5, the change in $D_{\text{Am}}^{0.01}$ with radiation dose is similar for γ- and α-radiolysis (LS and HPGe). The γ-radiolysis curve represents the existing model for radiolysis damage to TRUEX-NPH, It is based upon doses from 10–200 W·h/L and is not reliable for the dose range examined here. The increase in D_{Am} with increasing dose indicates that this irradiation yields powerful acidic extractants for actinides that are resistant to stripping by 0.25 M Na_2CO_3.

The change in $D_{\text{Am}}^{0.05}$ with dose is small for the LS and HPGe data sets plotted in Fig. 6. The "gamma" curve and LS data agree, but both surpass the slope shown for the HPGe data. We concluded that buildup of americium in the strip stages should not be a problem in TRUEX processing. However, inclusion of an alumina column or other aggressive means of stripping plutonium from the solvent is required to ensure reliable recycling of the solvent. The values of $D_{\text{Am}}^{2.0}$ were generally lower for the LS than HPGe data (Fig. 7). Rather than following a clear trend as in the weaker acid contacts, the 2.0 M HNO_3 D values experienced a fairly uniform 5–20% depression from fresh solvent in partitioning of americium and plutonium for all doses.

Many of the samples of irradiated solvent were resampled 4–6 weeks after the initial aliquots were measured for ^{241}Am activity. The aqueous

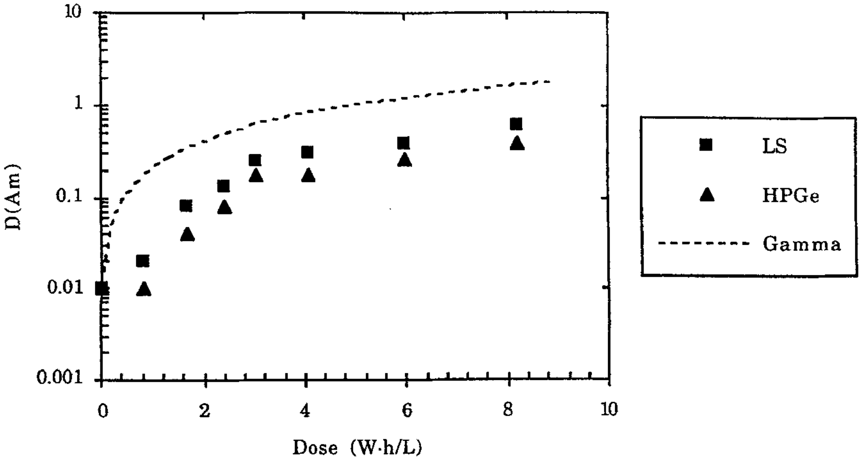

FIG. 5 Change in D_{Am} with dose for 0.01 M HNO_3 and for TRUEX-NPH scrubbed with 0.25 M Na_2CO_3. The α-radiolysis samples were counted for α-activity using liquid scintillation (LS) and for ^{241}Am γ-activity using a high-purity germanium detector (HPGe). The (gamma) curve is based on the γ-radiolysis of TRUEX-NPH (5).

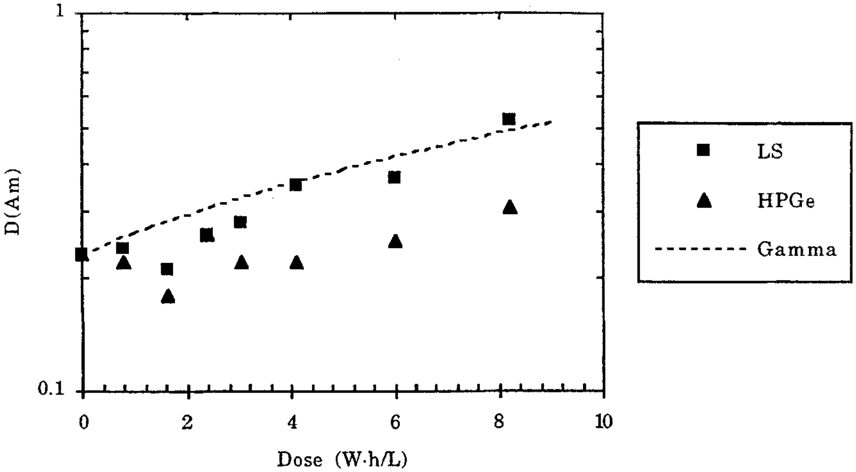

FIG. 6 Change in D_{Am} with dose for 0.05 M HNO_3 TRUEX-NPH scrubbed with 0.25 M Na_2CO_3. The α-radiolysis samples were counted for α-activity using liquid scintillation (LS) and for ^{241}Am γ-activity using a high-purity germanium detector (HPGe). The (gamma) curve is based on the γ-radiolysis of TRUEX-NPH (5).

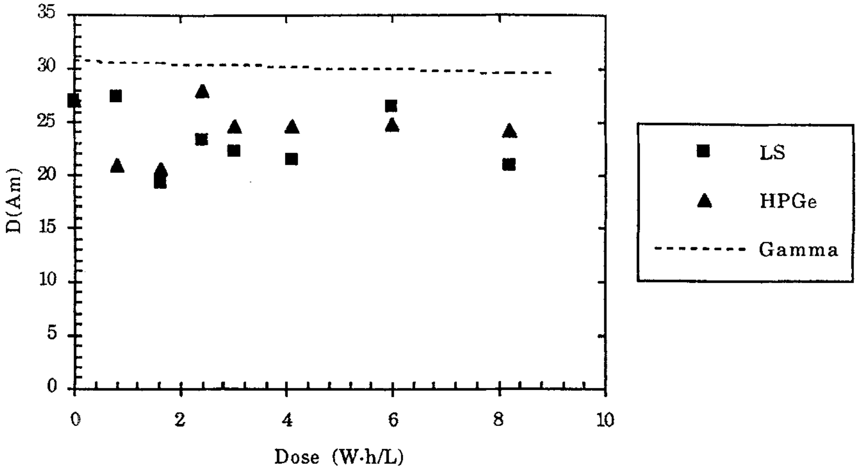

FIG. 7 Change in D_{Am} with dose for 2.0 M HNO_3 TRUEX-NPH scrubbed with 0.25 M Na_2CO_3. The α-radiolysis samples were counted for α-activity using liquid scintillation (LS) and for ^{241}Am γ-activity using a high-purity germanium detector (HPGe). The (gamma) curve is based on the γ-radiolysis of TRUEX-NPH (5).

and organic phases were not separated after irradiation but were kept in contact in sealed culture tubes stored in a rack inside a glovebox. Surprisingly, most of the samples scrubbed with 0.25 M Na_2CO_3 showed changes in the distribution of ^{241}Am between the aqueous and organic phases. The D_{Am} values approached those obtained for unirradiated solvent for each of the acid concentrations (0.01, 0.05, and 2.0 M HNO_3).

These samples of scrubbed solvent were checked for precipitation or third phase formation, but neither was visible. The mass balance of ^{241}Am between the phases was also checked. The total amount of americium was constant within 10%. It appears as though a slow migration of americium occurred between the phases, or perhaps some other mechanism slowly changed the americium concentration. Since a small change in americium concentration can cause a significant change in D_{Am}, surface adhesion or a very thin third phase may be responsible for the change in D_{Am}.

The concentration of nonamericium α-activity retained by the irradiated solvent was surprising. Based on the source of the ^{241}Am stock and its α-spectrum, we assumed that all the nonamericium α-activity was due to plutonium. Despite being below detection limits in the stock americium solution, the plutonium concentrations often exceeded americium in the contact solutions used to determine D_{Am} (Fig. 8).

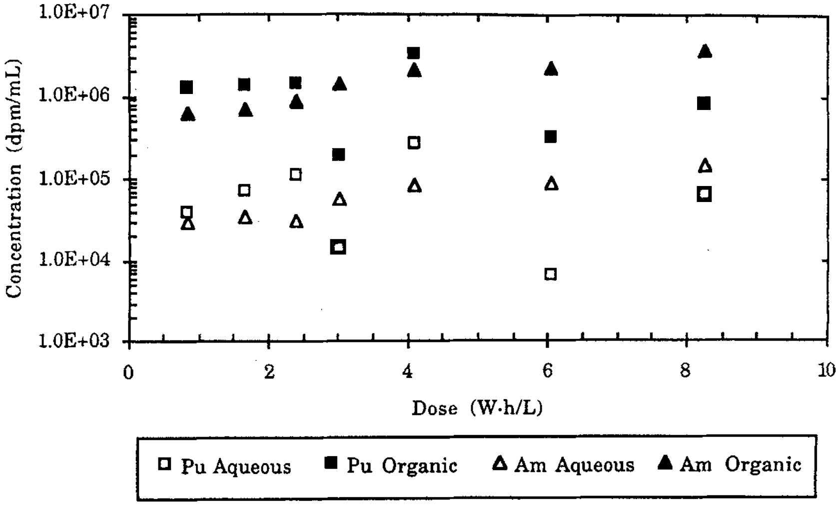

FIG. 8 Concentrations of americium and plutonium in aqueous and organic phases for contacts of 2.0 M HNO_3 with carbonate-scrubbed TRUEX-NPH. It was assumed that all nonamericium α-activity was due to plutonium.

CONCLUSIONS

α-Radiolysis of TRUEX-NPH solvent does not significantly affect the distribution ratios of americium with the HNO_3 concentrations (0.05 and 2.0 M) normally used in extraction and stripping during TRUEX processing. The radiation-induced degradation products do not resist americium stripping. This is at least partially due to acidic products being complexed to plutonium isotopes. The retention of plutonium in the solvent appears to be responsible for most of the changes observed in distribution ratios. The trace level of plutonium initially loaded into the solvent is retained and surpasses the americium concentration for some contacts. Competition between plutonium and americium may be responsible for the consistent depression of D_{Am} for all doses with 2.0 M HNO_3. Since plutonium retention is problematic, an α-radiolysis study monitoring D_{Pu} and using plutonium as the radiation source would be useful.

ACKNOWLEDGMENT

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