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Extraction of Radio-Strontium from Nitric Acid Medium Using Di-*tert*-Butyl Cyclohexano-18-Crown-6 (DTBCH18C6) in Toluene–1-Octanol Diluent Mixture

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An alternative extraction system to the SREX solvent using a diluent mixture comprising 4,4'(5')di-*tert*-butylcyclohexano-18-crown-6 (DTBCH18C6) in 80% toluene–20% 1-octanol was developed and evaluated for Sr(II) extraction from pressurized heavy water reactor simulated high level waste (PHWR-SHLW). The acid uptake (5.7%) by the present solvent was significantly lower as compared to that by the SREX solvent (21%) which used 100% 1-octanol as the diluent. The extracted species conformed to the ion-pair $[\text{Sr}(\text{DTBCH18C6})(\text{H}_2\text{O})_x]^{2+} \cdot 2[(\text{NO}_3)(\text{H}_2\text{O})_y]^-$. Studies on Sr(II) extraction as a function of nitric acid concentration indicated more favorable extraction and stripping with the present solvent as compared to the SREX solvent. Loading studies with 0.025 M DTBCH18C6 in the diluent mixture, carried out using the Sr carrier, indicated a decrease in D_{Sr} from 3.1 with 10 ppm Sr carrier to 1.62 with 100 ppm Sr carrier. Other important physical parameters relevant for the extraction processes such as phase separation time (dispersion number), viscosity, and density were also measured. The radiation stability and reusability of the solvent was also investigated. In sharp contrast to the SREX solvent, with increasing absorbed dose the proposed solvent showed an increase in Sr extraction and an increased acid uptake.

Keywords crown ether; diluent effect; high level waste; radio strontium; solvent extraction

INTRODUCTION

One of the strategies for the remediation of high level nuclear waste (HLW) involves the selective separation of minor actinides such as ^{241}Am , ^{243}Am , ^{245}Cm , and long-lived fission products such as ^{129}I , ^{135}Cs , ^{99}Tc , and ^{93}Zr prior to their ultimate burning/transmutation in high flux reactors or accelerator driven systems (1,2). However, the major source of radiation in HLW is from fission products such as ^{90}Sr and ^{137}Cs . Their removal prior to vitrification of the waste oxide in glass matrices not only helps in reducing the dose, it also helps in reducing the deformations caused by the heat dissipation of glass during its long term

storage. Moreover, the separated radio strontium, ^{90}Sr ($t_{1/2} = 28.5$ years), due to its high power density (0.93 watt/g), has many applications viz. as a fuel for thermoelectric and thermo-mechanical power generators (RTGs and RTMGs) (3).

Several methods like precipitation, ion exchange, and solvent extraction have been reported for the separation of Sr from HLW (which usually contains apart from residual U, Pu, minor actinides, activation/fission products, as mention above, along with structural materials such as Fe, Ni, Cr, Al, and Zr in about 3–4 M nitric acid) (4). Amongst the various separation methods known, solvent extraction is a widely accepted technique due to its ease of operation, rapidity and continuous nature. Out of the various reagents proposed for Sr extraction using solvent extraction, 18-membered crown ethers exhibit high extractability and selectivity based on metal ion and ligand cavity size compatibility (5,6). Literature reports have indicated that the choice of diluent is critical when Sr-extraction is carried out from picrate or moderate nitric acid medium using crown ethers. A major part of the earlier work, on Sr-extraction from picrate medium, involved the extraction of ion-pairs from alkaline or weakly acidic aqueous solutions (7,8). A literature report (9) showed that substituted dicyclohexano-18-crown-6 ligands are selective extractants for Sr^{2+} ion from nitric acid medium when a complex diluent mixture of di-*n*-dodecyl naphthalene sulphonic acid (DNNS), tri-*n*-butyl phosphate (TBP), and kerosene was used. Horwitz et al. (10–12) on the other hand, utilized the Sr(II)-specific extraction property of the di-*tert*-butyl cyclohexano 18-crown-6 (DTBCH18C6) in 1-octanol while developing the SREX process. They evaluated many aliphatic alcohols, ketones, carboxylic acids, and esters as diluents and showed a direct correlation of Sr extraction with their water uptake ability in the order: alcohol > carboxylic acid ~ ketone > ester (13). We have reported that the reagent inventory can be drastically reduced by using a more polar diluent mixture, such as 80% 1-butanol and 20% 1-octanol (14). However, long chain aliphatic alcohols also extract significant amounts

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of acid into the organic phase and hence require a greater number of extraction and stripping stages. It was required, therefore, to evaluate alternative diluents with improved stagewise extraction and stripping performance. It was reported that dicyclohexano-18-crown-6 (DC18C6) in a mixture of *iso*-dodecanol and *n*-hexyl benzene could be an effective diluent for the recovery of Sr from nuclear fuel reprocessing concentrate solutions (15). It appears, therefore, that a mixture of a long chain aliphatic alcohol with an aromatic diluent can be a promising diluent in view of their low acid uptake ability compared to solvents containing only aliphatic alcohols.

In the present work, a solution of DTBCH18C6 in a diluent mixture of 80% toluene and 20% 1-octanol was evaluated for the selective extraction of Sr^{2+} from pressurized heavy water reactor (PHWR) simulated high level waste (SHLW) whose composition is given in Table 1. Batch multistage extraction studies were also carried out for quantitative extraction as well as for stripping. Studies on the effect of crown ether concentration, nitric acid concentration, Sr carrier concentration, and irradiation stability of the solvent were also carried out.

EXPERIMENTAL

Reagents

DTBCH18C6 (>97%) was procured from Fluka Chemie AG, Switzerland and was used as received. A.R. grade 1-octanol and toluene, procured from BDH, were used as received. $^{85,89}\text{Sr}$ tracer was procured from BRIT, Mumbai. All other reagents were of A.R. grade. SHLW solution (PHWR type) was prepared by dissolving various constituents as desired (Table 1). Nitrate salts were preferred for this preparation.

TABLE 1
Composition of a PHWR simulated high level waste (SHLW); Acidity: 3.12 M HNO_3

Metal ions	Concentration (g/L)	Metal ions	Concentration (g/L)
Na	5.500	Mo	0.137
K	0.224	Y	0.060
Sr	0.030	La	0.180
Ba	0.064	Cs	0.320
Fe	0.720	U	0.640
Cr	0.119	Ce	0.060
Ni	0.107	Pr	0.090
Mn	0.426	Nd	0.120
Zr	0.004	Sm	0.085

Distribution Studies

Distribution of Sr

The distribution data for Sr were obtained by equilibrating 1 mL of the ligand solution in the suitable diluent with an equal volume of the aqueous phase (usually PHWR-SHLW) spiked with $^{85,89}\text{Sr}$ tracer in a thermostated water bath at $25 \pm 0.1^\circ\text{C}$ for about 1 hour. After centrifugation, the phases were separated and assayed radiometrically (514 keV gamma peak) using a well-type NaI(Tl) scintillator counter. The distribution ratio, D_{Sr} was defined as the ratio of the activity per unit volume in the organic phase to that in the aqueous phase. The experiments were carried out in duplicate and the precision was within $\pm 5\%$.

Distribution of the Extractant

The distribution ratio of the ligand DTBCH18C6 was estimated by a procedure reported by us earlier (16). A known concentration of the ligand solution was equilibrated with an equal volume (usually 5 mL) of 3 M nitric acid. After centrifugation and separation of the phases, about 1 mL of the aqueous phase was separated and spiked with a known amount of the radiotracer $^{85,89}\text{Sr}$ followed by equilibration in a tube containing an equal volume of the diluent (without the ligand). From the D_{Sr} obtained this way the ligand concentration in the aqueous phase was estimated using a previously calibrated log-log plot of D_{Sr} vs ligand concentration.

Distribution of Nitric Acid

The concentration of nitric acid in the organic phase was estimated by volumetric method by titrating a known aliquot in 50% water–50% methanol mixture using phenolphthalein as the indicator. Nitric acid concentration in the aqueous phase was also estimated in a similar manner using saturated potassium oxalate to complex the metal ions present in SHLW. From the equilibrium concentration of nitric acid in both the phases, the distribution ratio for nitric acid was calculated.

Measurement of Physical Parameters

The dispersion number, N_{Di} , for the diluent mixture was estimated from the following equation:

$$N_{Di} = \frac{1}{t_B} \sqrt{\frac{\Delta Z}{g}} \quad (1)$$

where t_B is the phase disengagement time in seconds, ΔZ , the total height of two phases in meters and $g = 981 \text{ cm} \cdot \text{sec}^{-2}$. The dynamic viscosity and density values at room temperature were measured using an Anton Paar, Austria viscometer (model number SVN 3000). All physical measurements were carried out in duplicate and the data presented are the mean values.

Irradiation Studies

The crown ether in the diluent mixture comprising of 20% 1-octanol + 80% toluene as well as in 100% 1-octanol alone were irradiated up to a dose of 100 MRad in a Co-60 gamma chamber. The Sr(II) extraction studies and the acid uptake studies using the irradiated solvent were carried out in a manner reported above.

RESULTS AND DISCUSSION

Earlier reports have indicated that the crown ether, 4',4''(5'')-di-*tert*-butyl-dicyclo hexano-18-crown-6 (DTBCH18C6) in 1-octanol, when used as the solvent, showed a high extraction efficiency from acidic feeds (10,11). With DTBCH18C6 in 1-octanol as the diluent, the SREX process for removal of Sr was developed at the Argonne National Laboratory with 1 M of extractant. However, in order to decrease the ligand inventory, while maintaining the efficiency of extraction, one of the options is to use a lower fraction of the aliphatic alcohol (15,17).

Diluent Effect

We have carried out a detailed study on the effect of diluent composition on Sr(II) and acid extraction (17). The results indicated that toluene–1-octanol mixture gave not only higher Sr(II) extraction, but also extracted nitric acid to a much lower extent. It was also reported that the composition 80% toluene + 20% 1-octanol was optimum as it gave higher D_{Sr} as compared to the solvent containing 1-octanol alone. On the other hand, the acid uptake by the diluent mixture was also significantly lower (17). Table 2 gives the data on the extraction of Sr(II) from a simulated high level waste (SHLW). The D_{Sr} values with the diluent mixture were almost twice of those obtained with 1-octanol alone as the diluent. For both the solvents, when SHLW was used as the aqueous phase, there was slight decrease in the D_{Sr} values as compared to those obtained with 3 M HNO_3 as the feed. On the other hand, the acid uptake with the diluent mixture was >3 times lower as compared to the solvent comprising pure 1-octanol as the diluent.

TABLE 2

Optimization of 1-octanol + toluene composition for Sr(II) extraction from HNO_3 medium (or SHLW) using 0.025 M DTBCH18C6

Diluent composition	D_{Sr}	% acid extraction
100% 1-octanol	1.72 ^a	21.2 ^a
100% 1-octanol	1.49 ^b	21 ^b
20% 1-octanol + 80%-toluene	3.18 ^a	6.4 ^a
20% 1-octanol + 80%-toluene	2.75 ^b	5.7 ^b

Note. ^aStudies with 3 M HNO_3 ; ^bStudies with SHLW.

This was also reflected in the lower water uptake for the diluent mixture as will be discussed below.

Effect of Nitric Acid Concentration

The effect of nitric acid concentration on Sr(II) extraction from SHLW was investigated using 0.025 M DTBCH18C6 in both 100% 1-octanol and 20% toluene + 80% 1-octanol. For a feed containing SHLW at 3 M HNO_3 , the D_{Sr} values were 2.35 ± 0.03 and 2.54 ± 0.10 , for the two diluents, respectively. As shown in Fig. 1, the extraction profile of 20% toluene + 80% 1-octanol lies below that of 100% 1-octanol up to the feed acid concentration of 2.75 M HNO_3 , beyond which there was a reversal in the trend. This is of great relevance as using the proposed solvent it is possible to favorably extract radio strontium from the HLW which is usually at an acid concentration of 3–4 M. Similarly, stripping at lower acidity is also more favorable using the proposed solvent as compared to the SREX solvent (11). The acid uptake (in molarity) at different feed acidity is also presented in Fig. 1. As indicated, the acid extraction increased with the feed acidity in case of both the solvents. However, while a much larger concentration of acid uptake was observed in case of the solvent containing pure 1-octanol, the solvent mixture showed much lower acid extraction.

We have reported in an earlier publication that the acid extraction in case of the diluents containing aliphatic alcohols has a direct correlation with water uptake by the organic phase (18). It was expected that decreasing 1-octanol fraction would bring down the water uptake and hence the acid extraction. Table 3 gives the Karl–Fischer titration data for water content in the organic phase used in the present study for 3 M HNO_3 as well as

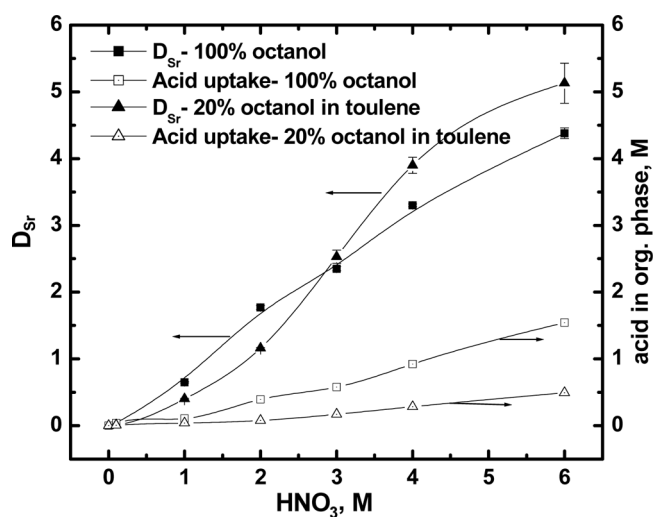


FIG. 1. Distribution data for Sr as well as nitric acid as a function of feed nitric acid concentration. [DTBCH18C6]=0.025 M.

TABLE 3
Water uptake data for different diluent mixtures for 3 M HNO₃ as the feed using 0.01 M DTBCH18C6

Diluent	Water content (%) ^a	Acid (%)	D _{Sr}
100% 1-octanol	5.71 ± 0.09 (5.91 ± 0.33)	11	1.03
80% 1-octanol + 20% toluene	3.33 ± 0.07 (3.18 ± 0.06)	8	1.02
60% 1-octanol + 40% toluene	2.31 ± 0.12 (2.56 ± 0.29)	6	1.16
50% 1-octanol + 50% toluene	2.29 ± 0.15 (2.15 ± 0.13)	3	1.22
40% 1-octanol + 60% toluene	1.50 ± 0.14 (1.73 ± 0.07)	3	1.30
20% 1-octanol + 80% toluene	1.10 ± 0.10 (1.00 ± 0.11)	1	1.18
100% toluene	NM ^c	NM ^b	NM ^b

Note. ^aValues in parentheses correspond to SHLW as the feed. ^bThe values were not measurable.

SHLW as the feed. The water content for the diluent mixture 20% 1-octanol + 80% toluene decreased to about 20% of the value obtained for 100% 1-octanol (Table 3). The water uptake for 3 M HNO₃ as the feed was comparable to that when SHLW was used as the feed (Table 3). In spite of the decrease in the water content and the acid uptake to about 1% for the diluent mixture, the D_{Sr} value was higher than that obtained for 100% 1-octanol as the diluent. This is in sharp contrast to literature reports correlating D_{Sr} with the water content in the organic phase (13).

Effect of Crown Ether Concentration

In the present case, an increase in D_{Sr} values with increasing crown ether concentration was observed. In order to evaluate the stoichiometry of the extracted species, crown ether concentration variation experiments were carried out using 0.025 M DTBCH18C6 in 20% 1-octanol + 80% toluene. The results are shown in Fig. 2. As previously reported by us and also by Horwitz et al.,

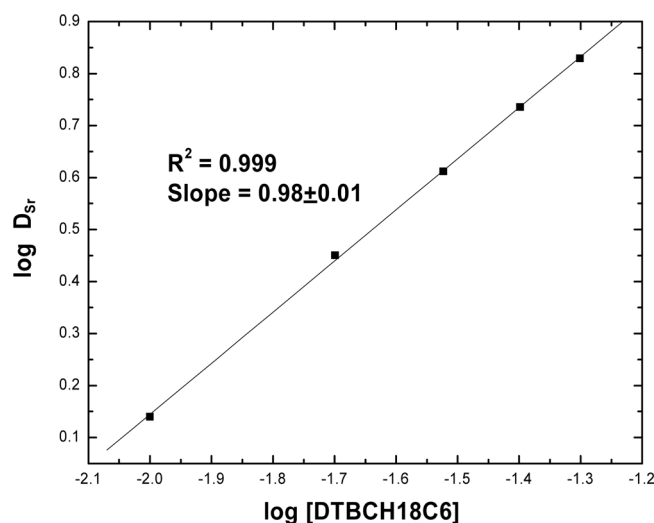
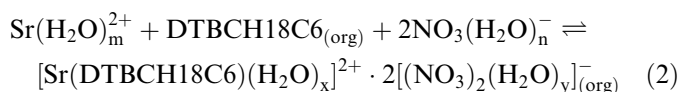


FIG. 2. Dependence of D_{Sr} on the crown ether concentration. Diluent: 20% 1-octanol + 80% toluene. Feed: 3 M HNO₃.

the ligand dependency on Sr(II) extraction was close to 1 indicating the following extraction equilibrium was prevalent (10,18).



where $m > x$ and $n > y$. The species in the organic phase was represented by the subscript '(org)' while those without any subscript refer to those in the aqueous phase. As the water extraction into the organic phase is dependent on the alcohol content, it is expected that the extracted species using the diluent mixture would contain lower number of associated water molecules ($x + y$) as compared to those in the species extracted by pure 1-octanol. For process application, it is required to evaluate the reusability of the solvent. Therefore, loss of the extractant into the aqueous phase needs to be determined experimentally. The partition coefficient values of the crown ether alone were measured as explained above. It was observed that about 0.005% of the extractant was partitioned into the aqueous phase from the diluent mixture as the diluent, which was much lower as compared to about 0.02% in case of the solvent using 100% 1-octanol as the diluent. This is again indicative of the fact that the present solvent is more suitable for the process applications as the losses towards the aqueous phase are much lower as compared to those with the SREX solvent.

Effect of Sr Carrier Concentration in the Feed

The PHWR-HLW contains about 30 ppm Sr (all isotopes) which is valid for a burn up of 7000 MWd/t and a cooling period of five years. The concentration of Sr is much higher in PWR and BWR high level wastes. In view of this, the extraction of Sr(II) was studied using Sr carrier solutions (10–100 ppm) spiked with ^{85,89}Sr tracer. As indicated in Fig. 3, a steady decrease in the D_{Sr} values was observed which is attributed to the loading effect.

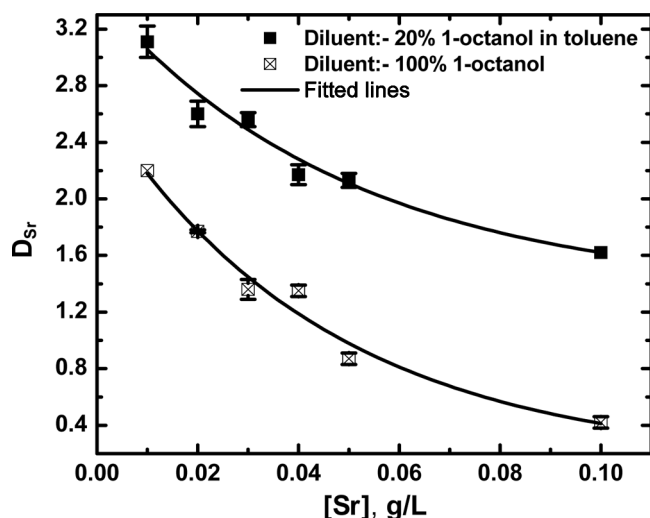


FIG. 3. Effect of Sr loading on D_{Sr} values. Organic phase: 0.025 M DTBCH18C6 in the diluent mixture as well as in 1-octanol. Feed: 3 M HNO_3 .

The results indicate reasonable D_{Sr} value (1.62) even for HLW containing 100 ppm Sr when 0.025 M DTBCH18C6 in 20% 1-octanol + 80% toluene was used as the solvent.

Batch Co-Current Extraction Studies using SHLW

The batch co-current extraction studies were carried out in order to evaluate the performance of the proposed solvent and the data were compared with those obtained using the solvent containing pure 1-octanol. Table 4 lists the extraction data using both the solvents. As indicated, while >99.3% extraction of Sr from SHLW is possible in about 4 stages using the proposed solvent (0.025 M DTBCH18C6 in 20% 1-octanol + 80% toluene), under identical condition about 96.5% Sr extraction was possible using the solvent containing pure 1-octanol. In five stages, the latter solvent yielded 98% extraction while the proposed solvent resulted in quantitative Sr extraction (>99.9%). As per the ongoing discussion, this was ascribed to a much lower extraction of acid by the proposed solvent

as compared to the SREX solvent (Table 4). In view of these results, it is suggested that the solvent developed in the present studies using 20% 1-octanol + 80% toluene is a superior solvent as compared to the one suggested earlier with pure 1-octanol [11].

Batch Co-Current Stripping Studies

Batch co-current stripping studies were carried out using the extracted Sr (spiked with $^{85,89}Sr$ tracer) from SHLW as per the method described in the previous section and the results are listed in Table 5. As indicated, while 3 stages of stripping were required for quantitative stripping using pure 1-octanol as the diluent, only 2 stages were needed using the diluent mixture used during the present study. This too is ascribed to the lower amount of nitric acid extracted by the diluent mixture as compared to that by pure 1-octanol.

Reusability of the Solvent

The reusability of the solvent was tested by carrying out the extraction and stripping studies and using the regenerated solvent by a second cycle of extraction and stripping and so on. The data on the reusability is presented in Table 6. It is clear from the data that there was reproducibility in the D_{Sr} , and acid uptake values even after 5 cycles. The stripping data from the reusability studies is presented in Fig. 4. As can be seen, quantitative stripping (>99.9%) was achieved in two cycles in all the four batches. Moreover, the difference in the stripping data is within the limits of experimental error. These results suggest that the proposed solvent can be used for long term process applications. However, for such applications, physical parameters such as the dispersion number, viscosity, and density data along with the irradiation stability of the solvent should be favourable (*vide infra*).

Irradiation Effect

The radiation stability is an important parameter for acceptance of a solvent for process applications. This, along with the reusability data, can decide the suitability

TABLE 4
Batch extraction data on Sr(II) recovery from simulated high level waste using 0.025 M DTBCH18C6 using 100% 1-octanol as well as 20% 1-octanol + 80% toluene

No. of stages	20% 1-octanol + 80% toluene		100% 1-octanol	
	% Sr extraction	% acid uptake	% Sr extraction	% acid uptake
I	73.4	6	56.9	20
II	92.9	6	86.8	16
III	98.1	5	92.8	14
IV	99.3	5	96.5	10
V	>99.9	5	98.0	9

TABLE 5

Batch stripping data for Sr(II) recovery from simulated high level waste using 0.025 M DTBCH18C6 using 100% 1-octanol as well as 20% 1-octanol + 80% toluene

No. of stages	20% 1-octanol + 80% toluene		100% 1-octanol	
	D_{Sr}	%Sr stripped	D_{Sr}	%Sr stripped
I	0.03	97.09	0.83	54.51
II	0.03	99.91	0.04	98.31
III	—	—	0.07	99.97

TABLE 6

Reusability data on the solvent, 0.025 M DTBCH18C6 in 20% 1-octanol + 80% toluene

Number of cycles	D_{Sr}	% extraction	% acid uptake	Remark about organic phase
I	2.46	71.10	6.21	Fresh
II	2.40	70.59	6.90	Reused once
III	2.52	71.59	6.47	Reused twice
IV	2.40	70.56	6.64	Reused thrice
V	2.39	70.50	6.95	Reused four times

of the process. Irradiation studies were carried out by exposing the solvents (both containing 0.025 M DTBCH18C6) to an absorbed dose equivalent to 100 MRad. The data on the Sr extraction is presented in Fig. 5 while that on acid extraction (%) is shown in Fig. 6. Interestingly, the proposed solvent (0.025 M

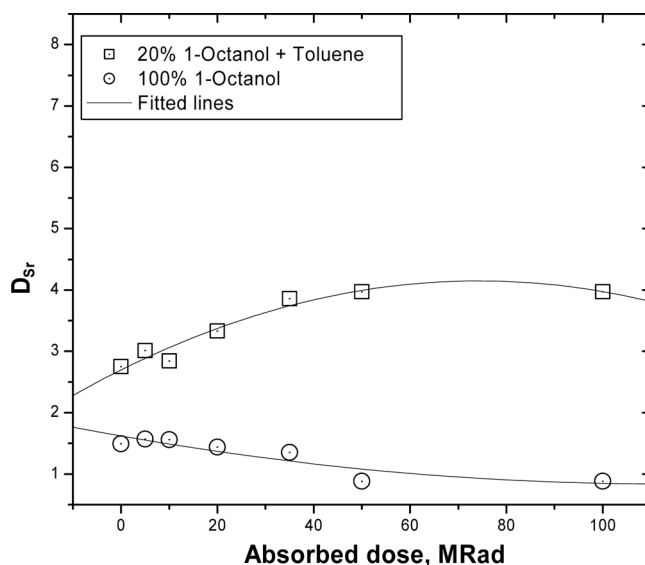


FIG. 5. Effect of absorbed dose on Sr extraction. Organic phase: 0.025 M DTBCH18C6 in 20% 1-octanol + 80% toluene. Feed: 3 M HNO_3 .

DTBCH18C6 in 20% 1-octanol + 80% toluene) showed an increase in its Sr extraction ability up to a dose of 40 MRad beyond which a plateau was noticed ($D_{Sr}=3.97$). On the other hand the solvent, 0.025 M DTBCH18C6 in 100% 1-octanol, showed little change in its Sr extraction ability up to a dose of 40 MRad beyond which gradual decrease in the D_{Sr} was noticed ($D_{Sr}=0.88$). The acid uptake data with irradiated solvents is presented in Fig. 6.

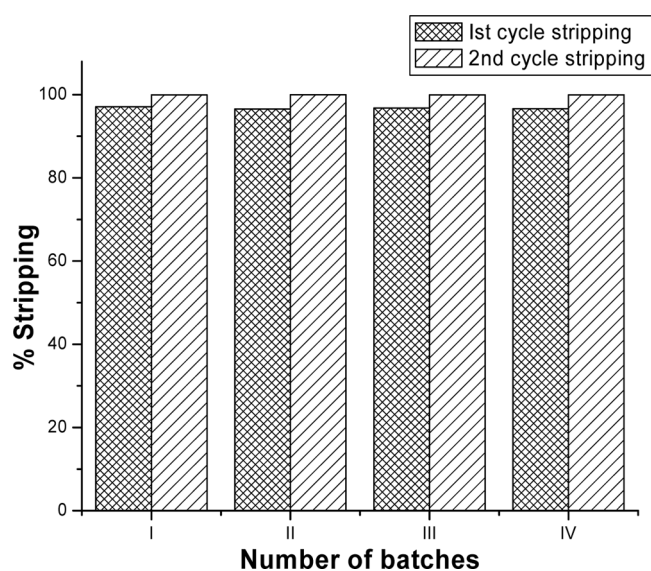


FIG. 4. Reusability data with 2 stages of stripping.

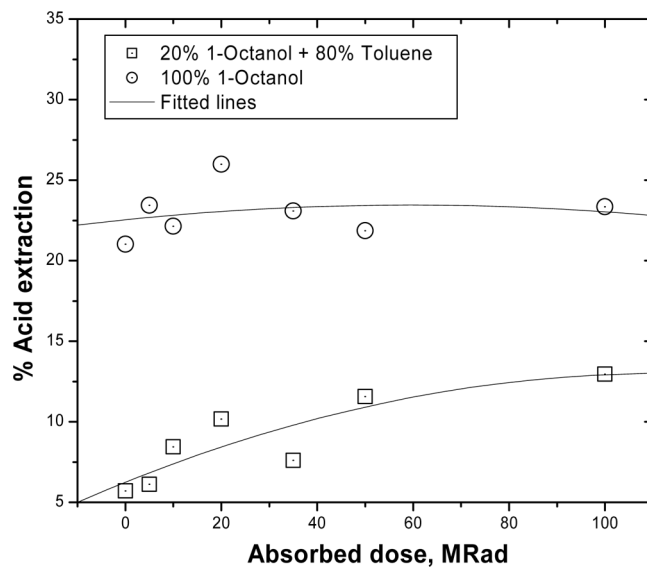


FIG. 6. Effect of absorbed dose on nitric acid extraction. Organic phase: 0.025 M DTBCH18C6 in 20% 1-octanol + 80% toluene. Feed: 3 M HNO_3 .

TABLE 7

Dispersion number, viscosity and density of the diluents containing varying fractions of toluene and 1-octanol

Diluent composition		Dispersion number (N_{Di})	Viscosity (mPa/S)	Density (g/cm ⁻³)
% 1-octanol	% toluene			
100	0	0.00472	8.869	0.8251
80	20	0.00319	4.4157	0.8318
60	40	0.00087	2.4200	0.8391
40	60	0.00299	1.4433	0.8466
20	80	0.00412	0.944	0.8534

The acid uptake by the pure 1-octanol solvent ($\sim 22 \pm 2\%$) from the SHLW did not change significantly with the absorbed dose while it gradually increased for the proposed solvent and attained a maximum of 13% at 100 MRad absorbed dose compared to the unirradiated solvent (5.7%). Sugo et al. have reported better radiation stability when aromatic diluents are used (19). Similar behavior is expected in case of a mixed diluent system used in the present system which contains 80% toluene and a slight improvement in Sr extraction with an increasing dose corroborates this. However, an increase in acid uptake with an increasing absorbed dose cannot be explained and the exact reason behind this interesting behavior needs to be investigated in greater detail.

Evaluation of Physical Parameters

The dispersion numbers (N_{Di}) obtained using 80% toluene–20% 1-octanol mixture and 100% 1-octanol are 4.12×10^{-3} and 4.72×10^{-3} , respectively suggesting that from the phase disengagement point of view 80% toluene–20% 1-octanol mixture is nearly comparable with 100% 1-octanol. The viscosity of the diluent mixture, on the other hand, was much more favorable compared to pure 1-octanol while the density values were comparable (Table 7).

CONCLUSIONS

From the present study it is clear that the solvent 0.025 M DTBCH18C6 in 80% toluene–20% 1-octanol mixture is superior to the SREX solvent (solution of the crown ether in 100% 1-octanol) in view of better extraction behavior, better stripping behavior, lower acid uptake, lower partitioning of the ligand towards the aqueous phase, higher irradiation stability, and favorable viscosity. Surprisingly, the irradiation stability indicated higher extraction efficiency at higher absorbed dose (100 MRad) for the proposed solvent as compared to the SREX solvent which showed almost 50% decrease in the Sr extraction efficiency. It may be of interest to carry out hot mixer-settler runs using this solvent to recover radio strontium from actual high level waste.

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