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Extraction chromatographic studies of metal ions using *N*,*N*,*N*′,*N*′-tetraoctyl diglycolamide as the stationary phase

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Abstract

N,*N*,*N*′,*N*′-tetraoctyl diglycolamide (TODGA) has been used as the stationary phase in the extraction chromatographic separation of actinides and other metal ions from pure nitric acid as well as from simulated high-level waste (SHLW). Chromosorb-W was found to be a better support material amongst the different solid supports evaluated viz. chromosorb-W, chromosorb-102, XAD-4 and XAD-7. Uptake profiles of various metal ions, such as U(VI), Pu(IV), Am(III), Eu(III), Fe(III), Sr(II) and Cs(I) were obtained as a function of acidity by batch studies using TODGA/chromosorb-W. Effect of macro concentration of Nd, Fe and U suggested that the uptake of Am(III) is mainly influenced by the presence of trivalent lanthanide ions. Breakthrough capacity of the resin material for Am(III) in presence of macro amount of Eu(III) was determined in the successive cycles of loading and elution. Loading capacity of the column was found to be 20 mg of Eu/g of the resin material. Elution studies of Am(III) suggested that 0.01 M EDTA was effective amongst different eluents used.

Keywords: TODGA; Extraction chromatography; Actinide-partitioning; Waste management

1. Introduction

The high-level waste (HLW) solution generated during the PUREX reprocessing of the spent nuclear fuel contains long-lived minor actinides, such as ^{241}Am, ^{243}Am, ^{245}Cm and ^{237}Np apart from the small amounts of unrecovered plutonium and uranium. At present, the most accepted concept for the management of HLW is to vitrify it in the glass matrix followed by disposal in deep geological repositories. Since the half lives of the minor actinides concerned range between a few hundred to millions of years, the surveillance of this high active waste for such a long period of time is rather difficult. An alternative or a complementary concept is the partitioning and transmutation (P&T) option, which envisages the complete removal of minor actinides from HLW solution and their consequent burning in reactors as mixed oxide fuels [1–3]. This process will lead to generation of extra energy and at the same time would alleviate the waste disposal problems. In the last two decades, all over the world, efforts have been made in several international laboratories to use the already known reagents and also to synthesize several new class of reagents for the efficient partitioning of minor actinides from high-level waste solutions. A number of solvent extraction processes, e.g. TRUEX, TRPO, DIDPA and DIAMEX utilizing the reagents octyl (phenyl) *N*,*N*-diisobutylcarbamoylmethyl phosphine oxide (CMPO), trialkyl phosphine oxide (TRPO), diisodecyl phosphoric acid (DIDPA) and *N*,*N*′,*N*,*N*′-dimethyl dibutyl tetradecyl malonamide (DMDBTDMA), respectively, have shown great promise and few of them have been tested upto the pilot plant scale [4–11]. Amongst these extractants, diamides have been found to be particularly promising in view of their improved back extraction properties towards Am(III)/Cm(III), their complete incinerability and the innocuous nature of their radiolytic degradation products (mainly carboxylic acids and

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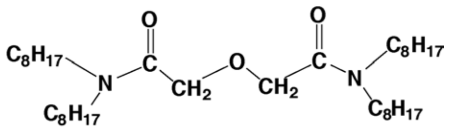

Fig. 1. *N*,*N*,*N*′,*N*′-tetraoctyl diglycolamide (TODGA).

amines) that can be easily washed out [12–14]. To increase the efficiency of diamides towards the forward extraction of Am(III) and Cm(III), several modifications have been attempted. It has been observed that the introduction of one etheric oxygen in between the two amide groups (diglycolamide) causes significant enhancement in the extraction efficiency of minor actinides due to their tridentate nature [15]. Recently developed, tridentate ligand, *N*,*N*,*N*′,*N*′-tetraoctyl diglycolamide (TODGA, Fig. 1) has been identified as one of the most powerful extractants being considered for the partitioning of trivalent actinides and lanthanides from HLW solutions [16,17].

In view of their continuous nature, solvent extraction processes are extensively employed for plant scale operations for the recovery of metal ions in quantities larger than kilograms. However, the major problem associated with this technique is the generation of large volume of secondary waste and handling of large volume of inflammable diluents, particularly when the metals quantities involved are in the grams/milligrams range. It is, therefore, imperative to look for an alternate technique where the secondary waste volume is lower. In this connection, several techniques like liquid membrane, magnetically assisted chemical separation (MACS) and extraction chromatography (EC) are being evaluated for the partitioning of minor actinides from HLW solutions [18–25]. Amongst these techniques, EC offers distinct advantages, such as minimum generation of secondary waste and ease of operation. This technique combines the versality of solvent extraction and chromatographic techniques. One of the differences between solvent extraction and chromatography process is the change in the activities of the extractant and the extracted complex due to the influence of support. Other difference is the non-attainment of the thermodynamic equilibrium when EC is performed in a column. However, this does not influence the chemical steps of transfer of the solute from one phase to another [26]. Recently, one EC-based study has been reported employing TODGA as the stationary phase employing Amberchrom® CG-71C as support material. The performance of the resin material has been compared with that prepared by using a malonamide viz. 2-(2-hexyloxy-ethyl)-*N*,*N*′-dimethyl-*N*,*N*′-dioctyl-malonamide (DMDOEHMA) [27].

In the present paper, extraction chromatographic material prepared by impregnating TODGA on chromosorb-W has been evaluated with respect to the uptake of Am(III) for the first time. Comparison of this material was also made with extraction chromatographic materials prepared using Cyanex-923, CMPO and DMDBTDMA for Am(III) uptake studies. Batch studies have been carried out for the uptake of Am(III), Eu(III), Pu(IV), U(VI), Fe(III), Sr(II) and Cs(I) ions from pure nitric acid as well as from simulated high-level waste (SHLW) solution. In the column studies, breakthrough curves for Am(III) were obtained under different loading conditions.

2. Experimental

2.1. Reagents and radionuclides

N,*N*,*N*′,*N*′-tetraoctyl diglycolamide was synthesized at Chemistry Department, University of Delhi. The details of the synthesis procedure is described elsewhere [28]. Chromosorb-W (dimethyl dichlorosilane treated acid washed celite diatomaceous silica) and chromosorb-102 (styrene divinylbenzene polymer) obtained from Johns Manville, Amberlite XAD-4 (styrene divinylbenzene polymer) and Amberlite XAD-7 (polyacrylic ester) procured from Sigma Chemicals Co., USA, were washed with distilled water and acetone followed by air-drying before use. The mesh size was 60–80 for all the solid supports. All the other reagents used were of analytical reagent grade. ^{233}U tracer was purified by anion exchange in HCl medium to eliminate the daughter products of ^{232}U (formed as a byproduct during the irradiation of ^{232}Th for ^{233}U production) and its purity was confirmed by α spectrometry [29]. Pu (principally ^{239}Pu) was purified from ^{241}Am in HNO_3 medium and its radiochemical purity was ascertained by gamma spectrometry for the absence of ^{241}Am [30]. Pu(IV) was extracted by 0.5 M HTTA (2-thenoyl trifluoroacetone) in xylene at 1 M HNO_3 and stripped by 8 M HNO_3 and was used as stock for Pu(IV) [31]. Further, plutonium valency in the aqueous phase was adjusted and maintained in tetravalent state by the addition of 0.05 M $NaNO_2$ + 0.005 M NH_4VO_3 (holding oxidants). Other radionuclides viz. ^{241}Am, ^{85,89}Sr, ^{152,154}Eu, ^{59}Fe and ^{137}Cs were procured from Board of Radiation and Isotope Technology (BRIT), Mumbai, India. ^{59}Fe tracer was purified by a method developed in our laboratory employing Cyanex-923 as extractant and its purity was ascertained by gamma spectrometry using HPGe detector for the absence of ^{60}Co [32].

2.2. Preparation of chromatographic resin material

The extraction chromatographic resin material was prepared by impregnating TODGA on chromosorb-W, chromosorb-102, XAD-4 and XAD-7. A known amount of TODGA was diluted in acetone (1:1) and was mixed with equal weights of the above-mentioned solid supports. The slurry was equilibrated for 24 h in a mechanical shaker followed by the solvent removal by flushing nitrogen gas with gentle stirring. The resultant material was vacuum dried to constant weight. The weight percentage of the extractant loaded on the resins was calculated from the difference in

Table 1
Composition of simulated high-level waste (SHLW) for pressurised heavy water reactor (PHWR)

Constituent	Concentration (mg/L)	Constituent	Concentration (mg/L)
Se[a]	12.3	Rb[b]	74.5
Sr[b]	186.3	Y[c]	99
Zr[b]	771.3	Mo[a]	731.3
Ru[c]	463.8	Co[b,##]	127.5
Pd[c]	267.5	Ag[b]	18.6
Cd[b]	16.3	Sn[a]	15.6
Sb[a]	4.7	Te[a]	102.8
Cs[b]	543.8	Ba[b]	308.8
La[c,#]	263.8	Ce[b]	532.5
Nd[d]	862.5	Eu[d]	22.6
Sm[d]	163.8	Fe[a]	500
Na[b]	3000	Cr[b]	100
Ni[b]	100	Mn[b,###]	181.3
U[b]	20000	Tb[d]	5.0
Pr[d]	243.8	Dy[d]	2.0
Gd[d]	165		

[a] Metal powder.
[b] Nitrate salt.
[c] Chloride salt.
[d] Oxide.
[#] Inclusive of Pm.
[##] Inclusive of Rh.
[###] Taken in place of Tc.

the weight of the resin before and after equilibration and was found to be ∼50% (w/w) in all the cases. Loading percentage of the extractant was also confirmed by elemental (C, H, N) analysis.

2.3. *Preparation of simulated high-level waste*

The concentration of various metal ions in a typical SHLW solution is given in Table 1. Salts in the nitrate form were preferred and in the cases where nitrate salts could not be arranged, metal powder, metal oxide and chloride salts were employed. Care was taken to dissolve each of them separately in hot concentrated nitric acid before their addition to the mixture. Acidity of the mixture was ascertained by alkalimetry in the presence of neutral saturated $K_2C_2O_4$ solution and the overall acidity of SHLW was adjusted as desired with suitable concentration of nitric acid. The concentration of fission products in SHLW corresponds to the burn up of 6500 MWd/Te of natural UO_2 in a pressurised heavy water reactor (PHWR) [5].

2.4. *Distribution studies*

The sorption of radionuclides from nitric acid medium as well as from SHLW was investigated by equilibrating a known volume of solution (1.0 mL) with a known amount of resin material (∼25 mg). Agitation of the two phases was carried out in a thermostated water bath maintained at 25 ± 0.1 °C for 45 min. This time was found to be sufficient to attain the equilibrium condition for Am(III) and Fe(III) (Table 2). Subsequently, the tubes were centrifuged; the aqueous layer separated and centrifuged second time. Suitable aliquots of the aqueous phase were taken before and after equilibration for assaying radiometrically. Assay of ^{241}Am, ^{85,89}Sr, ^{152,154}Eu, ^{59}Fe and ^{137}Cs was carried out by gamma counting in a well type NaI(Tl) scintillation counter. Alpha counting for ^{233}U and ^{239}Pu was carried out by liquid scintillation counter employing a toluene-based scintillator containing 10% (v/v) di(2-ethylhexyl) phosphoric acid (HD2EHP), 0.7% (w/v) 2,5-diphenyloxazole (PPO), 0.03% (w/v) 1,4-di-[2-(5-phenyloxazoyl)]-benzene (POPOP). Material balance was within ±5%. The distribution ratio (K_d) was calculated by employing the following formula,

Table 2
Uptake of Am(III) and Fe(III) as a function of time by TODGA sorbed on chromosorb-W at 1 M HNO_3; temperature: 25 °C

Time of equilibration (min)	K_d	
	Am(III)	Fe(III)
2	1949	0.58
5	3001	0.67
10	5935	0.73
20	6129	0.53
30	6118	0.57
45	6384	0.64
60	6059	0.61
90	5371	0.65
120	6064	0.67

$$K_d = \left[\frac{C_0 - C}{C}\right]\frac{V}{W} \tag{1}$$

where C_0 and C are the concentrations of metal ions (in counts per unit time per unit volume) before and after equilibration, V is the volume of aqueous phase used (mL) and W is the weight of the resin material employed (g). For determining K_d-HNO_3, equilibration experiments were carried out in similar way in the absence of tracer. The concentration of HNO_3 in the aqueous phase was determined volumetrically before and after equilibration.

2.5. *Column preparation and characterisation*

The characteristics of the chromatographic resin material and of packed column are given in Table 3. The column was prepared by packing ∼500 mg of chromatographic resin material in a borosilicate glass column of about 4 mm in diameter. The bed volume and the bed density were calculated from the column dimensions and the weight of the packed chromatographic resin material. The volume of stationary phase (V_s) and volume of mobile phase (V_m) were estimated by the reported method [33]. The column was preconditioned by passing excess of appropriate nitric acid solutions, prior to the introduction of the sample solutions. The column operations were carried out at ambient temperature (25.0 ± 0.1 °C) at the flow rate of about 0.4 mL/min. The breakthrough curves and elution profiles were obtained by plotting radioactivity (in terms of counts per unit time per unit volume) of differ-

Table 3
Characteristics of chromatographic resin material and packed column

Resin material	
Stationary phase	TODGA
Support material	Chromosorb-W
Mesh size	60–80
Extractant loading	47% (w/w)
Average density of resin	1.08 g/mL
Packed column	
Bed volume	1.257 mL
Bed density	0.398 g/mL
Density of stationary phase	0.891 g/mL
Volume of stationary phase	0.210 mL/mL of bed
Volume of mobile phase	0.632 mL/mL of bed

ent volume increments of the mobile phase versus volume of solution passed.

3. Results and discussion

3.1. Batch studies

3.1.1. Evaluation of resin materials

Table 4 gives the K_d values of Am(III) by TODGA impregnated (loading 48 ± 2%) on the four different inert supports viz. chromosorb-W, chromosorb-102, Amberlite XAD-4 and Amberlite XAD-7 from 1 M HNO_3. It should be noted that except chromosorb-W, which is a celite diatomaceous silica, the rest of the inert supports employed are nonionic polymeric resins. It can be seen that the highest K_d value was obtained for chromosorb-W as solid support, possibly due to the polar nature of the material. The leaching of the extractant was independently investigated by equilibrating the equal amount of resin materials with 1 M HNO_3 for 1 h in a mechanical shaker. After removing the aqueous phase, it was contacted with a minimum volume (1 mL) of *n*-dodecane to dissolve the leached extractant followed by Am(III) distribution studies using above organic phase. For the different resin materials, order of D_{Am} observed was: chromosorb-102 (6.8×10^{-3}) > XAD-4 (6.1×10^{-3}) > XAD-7 (3.0×10^{-3}) > chromosorb-W (2.7×10^{-3}). Lowest D_{Am} in case of chromosorb-W reflected the minimum leaching out of the extractant from the support. This order of leaching was further confirmed by the elemental analysis of the acid washed chromatographic resin materials. Chromosorb-W has also been reported as an excellent solid support for actinide-partitioning employing DMDBTDMA as the stationary phase [25]. All further studies (batch as well as column) were, therefore, carried out using this solid support.

Table 4
Uptake of Am(III) by TODGA sorbed on different solid supports at 1 M HNO_3; temperature: 25 °C; mesh size: 60–80

Support material	K_d, Am(III)
Chromosorb-102	3433
XAD-4	3527
XAD-7	4085
Chromosorb-W	6125

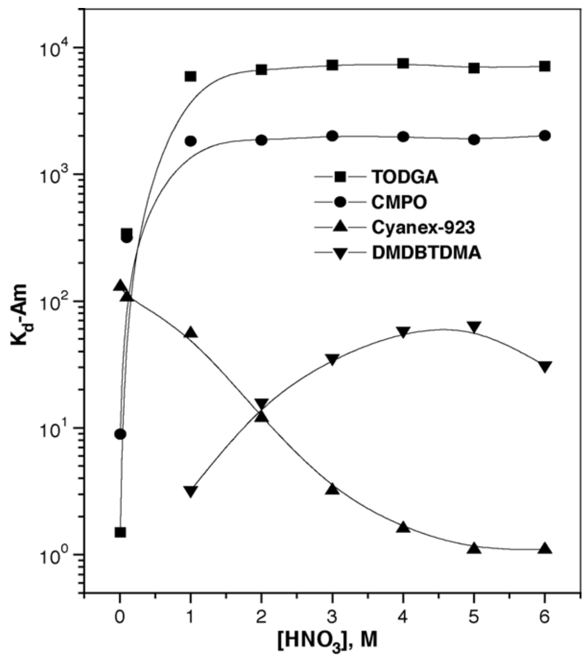

Fig. 2. Uptake of Am as a function of HNO_3 concentration by different extractants sorbed on chromosorb-W; temperature: 25 °C.

Fig. 2 shows the extraction profile of Am(III) as a function of nitric acid concentration for different extractants viz. TODGA, CMPO, Cyanex-923 and DMDBTDMA sorbed on chromosorb-W. In case of TODGA and CMPO, there was a sharp increase in the K_d values of Am(III) upto 1 M HNO_3 beyond which saturation was observed. For DMDBTDMA, Am(III) uptake increased gradually with nitric acid concentration and reached only to moderate values above 3 M HNO_3. On the other hand, Cyanex-923 shows moderate K_d value only at low acidity ($K_d = 140$ at 0.5 M HNO_3). The K_d values of Am(III) at 3 M HNO_3 followed the order: 7200 (TODGA) > 2000 (CMPO) > 35.3 (DMDBTDMA) > 3.2 (TRPO), suggesting TODGA as the most promising extractant for trivalent actinide.

3.1.2. Uptake studies of metal ions

The uptake profiles of metal ions, such as Am(III), Eu(III), Pu(IV), U(VI), Fe(III), Sr(II) and Cs(I) were obtained from pure nitric acid as well as from SHLW employing TODGA/chromosorb-W. Fig. 3 shows the variation in K_d values of metal ions as a function of HNO_3 concentration. It can be seen that the uptake of Am(III), Eu(III) and Pu(IV) increases sharply with the acidity upto 2 M HNO_3 beyond which saturation was observed. The order of uptake (K_d) observed for lanthanides and actinides was similar to that obtained in solvent extraction studies, i.e. Eu(III) > Am(III) > Pu(IV) > U(VI). These results clearly

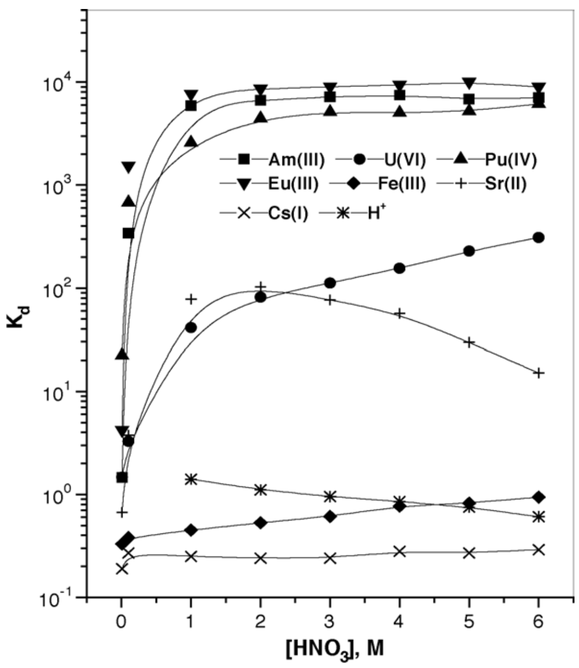

Fig. 3. Uptake of metal ions as a function of HNO_3 concentration by TODGA sorbed on chromosorb-W; temperature: 25 °C.

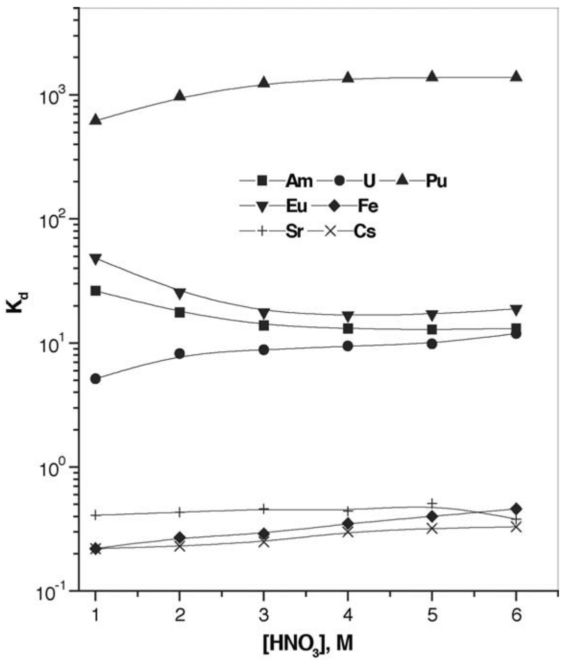

Fig. 4. Uptake of metal ions as a function of HNO_3 concentration from SHLW by TODGA sorbed on chromosorb-W; temperature: 25 °C.

demonstrate that TODGA molecule sorbed on chromosorb-W behaves similar to that observed in liquid–liquid extraction systems. In case of Sr(II), the K_d value increased with the aqueous phase acidity up to 3 M HNO_3 and decreased thereafter. For Cs(I) and Fe(III), the K_d values were less than 0.5 in the entire range of acidity investigated, suggesting insignificant uptake of these metal ions. For HNO_3, K_d value was found to decrease from 1.4 at 1 M HNO_3 to 0.6 at 6 M HNO_3. Table 5 gives the K_d values of Am(III), Pu(IV) and U(VI) for TODGA and compares with the other commonly used extractants, such as DMDBTDMA and CMPO sorbed on chromosorb-W at 4 M HNO_3. It can be seen that TODGA represents better uptake (K_d) for Am(III) and Pu(IV) as compared to that of DIAMEX solvent DMDBTDMA. It is worth noting that the commonly used neutral diamides act as bidentate ligand whereas the presence of additional etheric oxygen (in addition to two carbonyl oxygen, Fig. 1) in TODGA makes the molecule tridentate thereby increasing the complexation with the metal ion. This clearly demonstrates that tridentate diglycolamide represents excellent extraction properties for actinide(III) over bidentate diamides. It is worth noting that for DMDBTDMA and CMPO, the order of K_d was Pu(IV) > U(VI) > Am(III). This is the general order observed for many extractants, which follow the order of their ionic potential. In case of TODGA, the uptake of U(VI) is less as compared to Am(III) probably due to the presence of two oxygen atoms which might cause hindrance in the complexation with tridentate ligand. Though TODGA and CMPO display comparable extraction properties for actinides, there is distinct advantage of employing TODGA over phosphorous-based extractant in the secondary waste management due to its complete incinerability.

Table 5
Uptake of metal ions by different extractants sorbed on chromosorb-W at 4 M HNO_3; temperature: 25 °C

Extractant loaded on chromosorb-W	K_d for different metal ions			References
	Am(III)	Pu(IV)	U(VI)	
TODGA	7.5×10^3	5×10^3	1.5×10^2	Present work
DMDBTDMA	50	4×10^3	1.6×10^3	[25]
CMPO	3.5×10^3	3×10^5	1×10^5	[22]

Fig. 4 shows the variation in the K_d values of Am(III), Eu(III), Pu(IV), U(VI), Fe(III), Sr(II) and Cs(I) as a function of HNO_3 concentration from SHLW solution by TODGA sorbed on chromosorb-W. Interestingly, the K_d values are distinctly higher for Pu(IV) as compared to those of Am(III) and Eu(III). Also, at any acidity, the K_d value of a given metal ion is less in the SHLW as compared to that in pure HNO_3 (Fig. 3), apparently due to the co-extraction of other metal ions present in SHLW. The major cations responsible for the decrease in the Am(III) sorption by the resin could be the lanthanides present in the SHLW solution which show strong uptake. Also, no significant extraction of Sr(II) was observed from SHLW as maximum K_d, Sr(II) was ~0.5 in the entire range of acidity.

3.1.3. *Sorption of Am(III) under loading conditions of Nd, U and Fe*

Fig. 5 shows the uptake of Am(III) by TODGA/chromosorb-W in the presence of varying concentration of U(VI) and Nd(III). The results reflect that even the presence

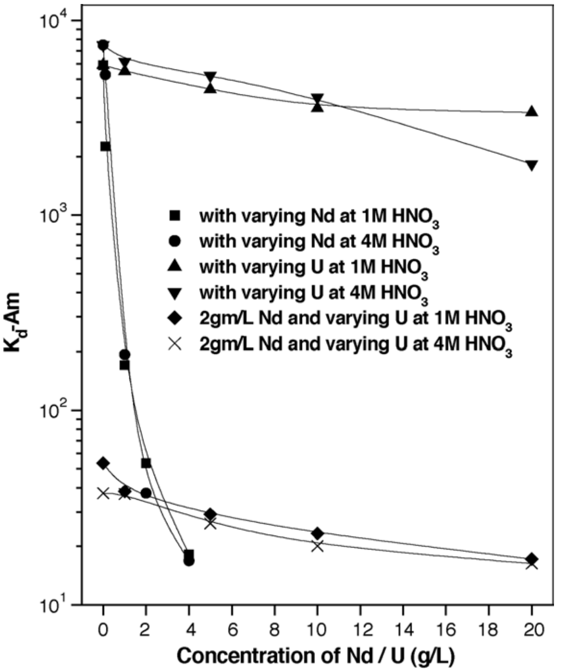

Fig. 5. Uptake of Am under loading condition of U and Nd by TODGA sorbed on chromosorb-W; temperature: 25 °C.

of 20 g/L of uranium does not affect the uptake behaviour of Am(III). This is attributed to the higher uptake of trivalent actinide as compared to hexavalent uranium as reflected in the batch studies. In case of Nd(III) (taken as a representative element of lanthanide), K_d value of Am(III) decreased sharply with increase in the lanthanide concentration. Similar effect could also be observed in the presence of 2 g/L of Nd and varying concentration of U. Literature survey has revealed that the presence of Fe(III) significantly affects the uptake of Am(III) as reported earlier for CMPO and DMDBTDMA [7,12,25]. Uptake of Am(III) was investigated, therefore, in the presence of macro amount of Fe (1–6 g/L). Table 6 shows that there is insignificant decrease in the uptake of Am(III) even in the presence of 6 g/L of Fe(III) investigated at 1 and 4 M HNO_3 suggesting that Fe(III) does not affect the recovery of minor actinides by TODGA. Behaviour of TODGA is quite different from those of CMPO and DMDBTDMA. It is related to the fact that whereas TODGA does not extract Fe(III); CMPO and DMDBTDMA have significant distribution ratio values for Fe(III) in the acidity range 1–6 M HNO_3.

Table 6
Uptake of Am(III) by TODGA sorbed on chromosorb-W in the presence of macro amounts of iron; temperature: 25 °C

[Fe] (g/L)	K_d, Am(III)	
	1 M HNO_3	4 M HNO_3
0.0	5895	7493
1.0	5902	7376
2.0	5704	7056
4.0	5893	6668
6.0	5401	6737

Table 7
Uptake of Am(III) by TODGA sorbed on chromosorb-W in the presence of nitrate and sulphate; temperature: 25 °C

In the presence of nitrate at 0.1 M HNO_3		In the presence of sulphate at 1 M HNO_3	
$[NO_3^-]$ (M)	K_d, Am(III)	$[SO_4^{2-}]$ (M)	K_d, Am(III)
0.0	341	0.00	5895
1.0	6610	0.02	6674
2.0	6820	0.04	7447
4.0	6776	0.06	7645
6.0	6556	0.08	7596
–	–	0.10	7513

3.1.4. Uptake behaviour of Am(III) from nitrate and sulphate media

The sorption of Am(III) in the presence of nitrate and sulphate is particularly relevant for the recovery of minor actinides from the nitrate and sulphate bearing waste solutions. Table 7 gives the batch distribution data for the uptake of Am(III) in the presence of varying concentration of nitrate and sulphate. It can be seen that the K_d value of Am(III) increased many fold (20 times) in the presence of 1 M nitrate which was attributed to the salting out effect. On the other hand, there was no significant change in the K_d value of Am(III) even in the presence of 10 g/L (0.1 M) of sulphate suggesting that minor actinides can be effectively separated from the sulphate bearing waste by TODGA/chromosorb-W.

3.2. Column Studies

3.2.1. Breakthrough curve for Am(III)

Fig. 6 shows the breakthrough curves for Am(III) from pure nitric acid medium as well as from SHLW under different loading conditions. It can be seen that about 10 mL solution containing 1 g/L of Eu(III) could be passed through the column without any leakage of ^{241}Am, both at 1 and 4 M HNO_3, suggesting that about 20 mg of Eu(III) can be loaded per 1 g of the chromatographic resin material (column contains 500 mg of the resin). It was interesting to note that no breakthrough for ^{241}Am was observed even after passing 100 mL of solution containing 20 g/L of U at 1 M HNO_3 spiked with ^{241}Am tracer. This suggests that U(VI) has no significant effect on the loading of Am(III) which was also reflected in the batch studies (mentioned in earlier section). However, when SHLW solution at 1 and 4 M HNO_3 was passed through the column, the breakthrough appeared with in 2 mL of the effluent solution due to the presence of large number of metal ions (including ~2 g/L of lanthanides), which are sorbed on the column. Batch studies as well as column studies suggested that the lanthanides are the major cations affecting the uptake of Am(III).

3.2.2. Elution studies of metal ions

In order to study the elution behaviour of Am(III) from the loaded column, different eluents were evaluated viz. 0.01 M HNO_3, 0.01 M EDTA, 0.1 M 2-Hydroxy Isobutyric Acid

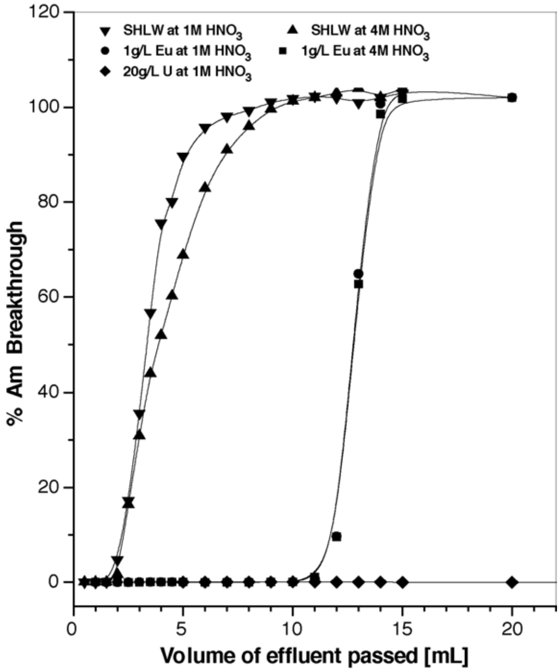

Fig. 6. Breakthrough curve for Am under different loading conditions on TODGA/chromosorb-W column; temperature: 25 °C.

(αHIBA) and 0.1 M Imino Diacetic Acid (IDA). The column was loaded with solution containing 1 g/L of Eu spiked with ^{241}Am tracer at 4 M HNO_3 and washed with 10 mL of 4 M HNO_3 before the elution. No activity release was observed in the washing solution. Constant flow rate of about 0.4 mL/min was maintained throughout the elution studies. Fig. 7 presents the elution curves for Am(III) for different eluents. It could be seen that more than 50 mL of 0.01 M HNO_3 was required for the complete recovery of Am(III). In case of complexing agents like EDTA and IDA, all Am(III) could be eluted within 20 mL of the eluent (>98%). Similarly, with αHIBA, recovery was possible with in the 25 mL of 0.1 M solution.

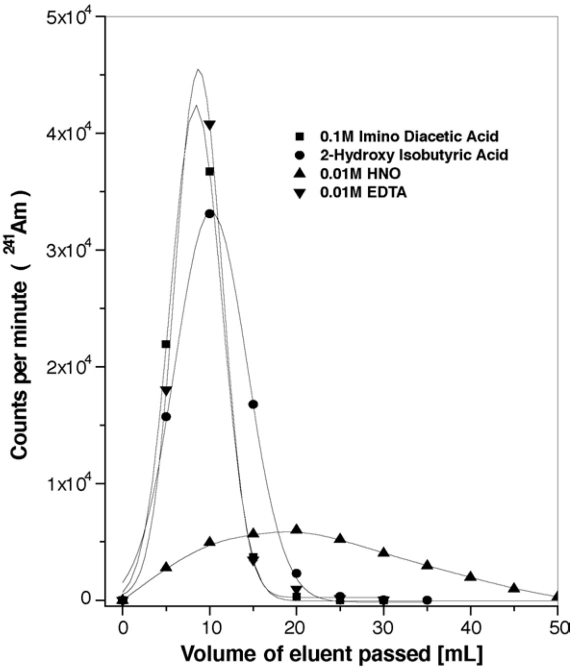

Fig. 7. Elution curve for Am by different eluents; loading solution: 1 g/L Eu containing Am tracer at 4 M HNO_3; temperature: 25 °C.

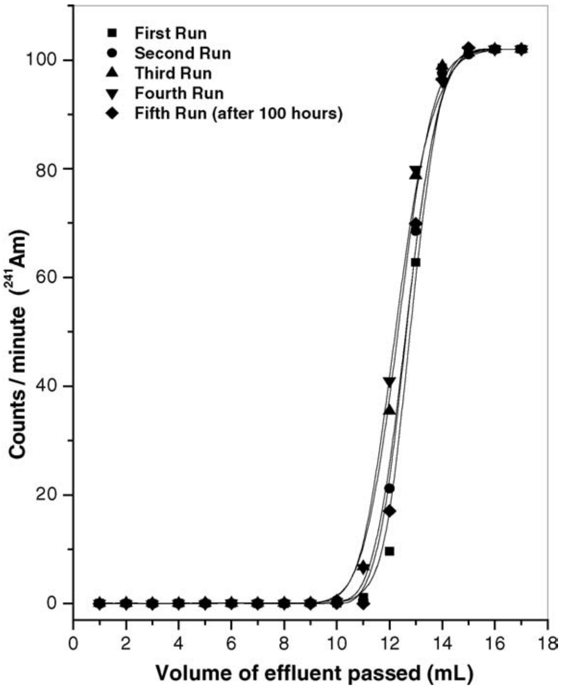

Fig. 8. Breakthrough curve for Am for successive loading; loading solution 1 g/L Eu at 4 M HNO_3; temperature: 25 °C.

3.2.3. Reusability of column

Loading solution, containing 1 g/L of Eu spiked with ^{241}Am tracer at 4 M HNO_3, was used to study the reusability of the column. Fig. 8 shows the breakthrough curves for the successive loadings of Am(III) using the same column. In the first loading, the column was loaded till the 100% breakthrough for Am(III) was observed and then Am(III) activity was eluted with 0.01 M EDTA. The column was subsequently washed with 15 mL water and conditioned with 4 M HNO_3 before the second loading was carried out. Subsequent loadings were carried out in the similar manner. It is interesting to note that there was no change in the breakthrough of Am(III) even after fifth cycle of loading which was performed after 100 h. The results clearly demonstrated the stability of the chromatographic resin material.

4. Conclusions

The chromatographic resin material prepared by impregnating TODGA on chromosorb-W was found promising for the partitioning of trivalent actinides. Chromosorb-W was found to be an excellent inert support. Whereas the macro concentration of U(VI) and Fe(III) do not influence the uptake of Am(III), trivalent lanthanides influence the uptake significantly. Loading capacity of the column was found to be 20 mg of Eu(III) per gram of the resin material. 0.01 M EDTA was found to be an efficient eluting agent for Am(III). Reusability of the column demonstrated the sufficient stability of the chromatographic resin material in recycling operations.

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