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Chromatographic separation of carrier free ⁹⁰Y from ⁹⁰Sr using a diglycolamide based resin for possible pharmaceutical applications

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ABSTRACT

N,N,N',N'-tetraoctyl diglycolamide (TODGA) has been used as the stationary phase in an extraction chromatography resin (XCR) material prepared for evaluating the uptake and the separation behaviour of ⁹⁰Y and ⁹⁰Sr from acidic feeds. Chromosorb-W was used as the solid support material while the feed solution was usually 4 M HNO₃. The batch uptake studies have suggested almost no Sr(II) uptake while Y(III) uptake increased with acidity up to 4 M HNO₃ beyond which a decrease in the $K_{d,w}$ values were observed. Column studies were carried out and breakthrough profiles were obtained for both Y(III) and Sr(II). No breakthrough of Y(III) was noticed even when >50 column volumes of the feed (carrier free ⁹⁰Y at 4 M HNO₃) was passed through the column while about 20 column volumes were required for the breakthrough of Y(III) when the feed contained 1 g/L Y in 4 M HNO₃ spiked with ⁹⁰Y tracer. The reusability of the column was also studied which indicated in the deterioration of the column performance as shown by the sharp fall in the breakthrough volumes and was attributed to the probable leaching of the reagent from the support material. The role of absorbed dose was also investigated for Y(III) uptake. Separation of carrier free ⁹⁰Y tracer was carried out by loading the column with ⁹⁰Sr and eluting with 0.01 M solutions of HNO₃ as well as EDTA. The purity of the product was ascertained by half-life method.

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1. Introduction

 90 Y is a pure beta-emitter with a high E_{max} of 2.3 MeV and a rather short half-life of 64.1 h. Because of its favourable nuclear and chemical properties, ⁹⁰Y compounds are considered suitable as radiopharmaceutical for therapeutic applications [1,2]. The high complex formation constants of Y(III) with chelating ligands make ⁹⁰Y – chelates suitable for their use as radiopharmaceuticals [3–5]. Though ⁹⁰Y can be easily obtained by neutron irradiation of natural yttrium, low specific activity of the product due to the presence of large amount of the carrier (⁸⁹Y) makes this source unsuitable for radiopharmaceutical applications. On the other hand, the alternative source of ⁹⁰Y is from the decay of ⁹⁰Sr which is abundantly available in spent nuclear fuel. ⁹⁰Sr ($t_{1/2}$ = 28.5 y) attains secular equilibrium with ⁹⁰Y in a short period and can serve as a long term source for the latter radionuclide. There are several methods such as solvent extraction. extraction chromatography and liquid membrane, etc., reported for the separation of ⁹⁰Y from ⁹⁰Sr using a variety of reagents [6-13]. Out of these, solvent extraction based methods appear promising due to their fast kinetics, simplicity and continuous nature. However, due to the growing concern for the

environment, solvent extraction methods are not preferable as they use large volumes of volatile organic compounds and generate large amounts of secondary wastes and there is a search for alternative 'green' separation methods. Techniques such as extraction chromatography on the other hand, are becoming increasingly popular as they thrive on very low volumes of the solvent.

N,N,N',N'-tetraoctyldiglycolamide (TODGA, Fig. 1) displays unique extraction behaviour as the lanthanide (Ln³⁺) and actinide (An³⁺) elements are extracted to a much higher extent as compared to the metal ions such as Th^{4+} and UO_2^{2+} and alkaline earth ions such as Sr²⁺ [14]. Several literature reports are available which employ TODGA as the extractant for trivalent lanthanide and actinide ions [15-17]. However, we have developed solvent extraction methods using diglycolamide extractants for the separation of ⁹⁰Y from ⁹⁰Sr for the first time [18,19]. In view of better acceptability of the extraction chromatography based methods over the solvent extraction based methods. TODGA impregnated extraction chromatographic resin (XCR) materials have been studied for metal ion uptake in a variety of systems [20-23]. Out of these, Horwitz et al. [21] have carried out several separation studies including one involving ⁹⁰Y separation from ⁹⁰Sr. However, it was required to make a systematic investigation of ⁹⁰Y and ⁹⁰Sr separation using TODGA impregnated XCR and evaluation of this system for any practical application. Though ion-exchange chromatography method using a cation exchanger and a complexing agent as the

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Fig. 1. Structural formula of TODGA.

eluent was reported to be promising [13], the extraction chromatographic method utilizing the unique selectivity features of TODGA was needed to be studied from both basic chemistry as well as application point of view. Therefore, it was thought of interest to prepare TODGA impregnated XCR and investigate the separation behaviour of ⁹⁰Y from ⁹⁰Sr.

The present work deals with the relative sorption behaviour of Y(III) and Sr(II) in nitric acid and hydrochloric acid using a TODGA impregnated XCR made with Chromosorb W as the solid support. On the basis of the batch uptake date, column studies have also been carried out. The separation 90 Y from 90 Sr was also investigated and the reusability and radiation stability of the resin was evaluated. Comparison of the results with those obtained by Horwitz et al. using a similar extraction chromatographic material has also been made.

2. Experimental

2.1. Reagents

(TODGA) N,N,N',N'-tetraoctyldiglycolamide was procured from Thermax, India and was characterized by elemental analysis, NMR, IR, HPLC and distribution ratio measurements. Chromosorb W (60-80 mesh) was obtained from Johns Manville, USA and was used after thorough washing (vide infra). All the other reagents were of AR grade and were used as procured. Carrier free ⁹⁰Y tracer was prepared by milking from a crown ether column as described in a previous publication [18]. For other studies involving ⁹⁰Y tracer, the radionuclide was prepared by irradiating natural Y₂O₃ in Dhruva reactor at a flux of 1 × 10¹³ neutrons/cm²/s. ^{85,89}Sr tracer was procured from BRIT, Mumbai and was used as a surrogate for ⁹⁰Sr. For the limited studies involving ⁹⁰Sr, the radionuclide was procured from BRIT, Mumbai. Assaying of ⁹⁰Y and ^{85,89}Sr was done by beta counting using liquid scintillation counter (Hidex, Finland) and gamma counting using NaI(Tl) scintillation counter (Para Electronics, India) inter phased to a multi-channel analyzer (ECIL, India), respectively. Toluene based commercial liquid scintillator cocktail (Sisco Research Laboratory, Mumbai) was used for the radiometric assay of ⁹⁰Y.

2.2. Procedure

2.2.1. Preparation of extraction chromatography resin

Chromosorb W was cleaned by thorough washing with methanol, dilute hydrochloric acid and deionized water. The resin beads were dried at 60 °C to constant weight before loading the extractant. The extraction chromatographic resin material was prepared by equilibrating 6 g of the solid support material (Chromosorb W) with 4 g TODGA in methanol for 24 h. The solvent was evaporated by flushing nitrogen gas and the resultant solid was kept under vacuum in a desiccator till constant weight. The loading of TODGA in the resin material was 40% which was confirmed from weight measurements.

2.2.2. Batch distribution ratio measurements

⁹⁰Y and ^{85,89}Sr were used as the radioactive tracers in the extraction chromatographic studies. The distribution studies were performed in a tube containing about 100 mg of the resin coated

 $(K_{d,w})$ values were determined by the following formula,

$$K_{\rm d,w} = \frac{(C_0 - C)/W}{C/V}$$
 (1)

where C_0 and C are the initial and equilibrium concentration of metal ion, W, the weight of the resin and V, the volume of the aqueous phase. Assaying of the radiotracers was done as described above. Decay corrections were made for ⁹⁰Y counting measurements. All the experiments were carried out in duplicate and the distribution data represent the average of the two results. The material balance in all the experiments was within an error limit of $\pm 5\%$.

2.2.3. Column studies

The column containing the TODGA based XCR was prepared by packing about 1 g of the resin material in a glass column of about 4 mm diameter. In the present studies the flow rate was adjusted to about 4–6 drops (0.3 mL/min) by changing the packing density of the glass wool plug in the column and the liquid column height above the glass wool plug. The bed volume of the column was estimated from the length of the bed and the i.d. of the column. The elution profiles were obtained after loading a known amount of the ⁹⁰Y tracer followed by elution with 0.01 M nitric acid. All the runs were carried out at ambient temperature (25 ± 0.1 °C).

2.2.4. Separation studies

The separation factor (S.F.) is defined as the ratio of $K_{d,w}$ values of Y(III) to that of Sr(II). Batch separation studies were carried out using 90 Y and 85,89 Sr tracers. Based on the batch data, column studies were carried out and the separation of 90 Y from 90 Sr was accomplished under varying experimental conditions as mentioned below. Purity of the product (90 Y) was ascertained by monitoring the decay profile over a period of at least 3 half lives and calculating the half life from the slope of the semi-log plot of activity vs time as follows:

$$Half-life = \frac{0.693}{slope}$$
(2)

3. Results and discussion

3.1. Batch sorption studies

Our previous studies using several solid supports have indicated that Chromosorb W was more suitable than the other support materials [24,25]. In view of this, Chromosorb W was used in the present study as the support material. The characteristics of the resin material are listed in Table 1.

3.1.1. Effect of equilibration time

Prior to the batch sorption and column studies, it was required to study the kinetics of metal ion uptake which is very important in extraction chromatography studies. The optimum equilibration time for Y^{3+} ion (Sr^{2+} has much lower extraction with TODGA as the extractant [18]) uptake by the extraction chromatographic resin material was studied by agitating the tubes containing the resin with the feed containing the radiotracer (90 Y) for different time intervals. Though equilibrium was attained in about 5 min (Fig. 2), for all subsequent experiments an equilibration time of 30 min was used. The time taken for attaining equilibrium metal ion uptake value was significantly lower than that reported earlier by us for the uptake of Am³⁺ ion (similar in chemical behaviour to Y^{3+}) with

Table 1

Characteristics of the TODGA impregnated resin material and the column used in the present study.

Resin characteristics		
Stationary phase	TODGA	
Support material	Chromosorb W	
Mesh size	60-80	
Particle size	100–150 μm	
Extractant loading	43%	
Average density of the resin	1.06 g/mL	
Saturation loading capacity	0.7 mequiv./g	
Column characteristics		
Column dimension	0.4cm imes 13.8cm	
Resin weight	0.93 g	
Bed volume	1.73 mL	
Capacity of column	0.65 mequiv.	

DMDBTDMA, a tetra alkyl malonamide extractant [24]. However, the results obtained in the present studies agreed fairly well with that reported by Horwitz et al., in an analogous system using a different solid support material [21].

3.1.2. Batch sorption studies at varying acidities

Our solvent extraction studies have indicated that HCl medium is more suitable for the separation studies involving ⁹⁰Y and ⁹⁰Sr as compared to HNO₃ medium using TODGA as the extractant [18]. The batch sorption data from nitric acid and hydrochloric acid media indicated that the sorption of Sr(II) was insignificant with nitric acid as the feed and slightly higher $K_{d,w}$ values were reported with HCl at almost the entire range of acidity. The $K_{d,w}$ values for Y(III) were much higher as compared to those for Sr(II) and this was in the same line of their solvent extraction behaviour reported earlier by us [18]. It was intriguing to note that the $K_{d,w}$ values for Y(III) are significantly lower as compared to those reported by Ansari et al. involving Am(III) which was attributed to lower loading of the present XCR [20].

Higher $K_{d,w}$ values of Y(III) were obtained with HNO₃ as the feed as compared to HCl up to 3 M HNO₃ beyond which a crossover was seen (Fig. 3). While the peak value of $K_{d,w}$ for Y(III) was attained at 4 M HCl, the same was attained at 3 M acidity for nitric acid feeds. Moreover, the decrease beyond the peak value was less steep for HCl medium as compared to that in HNO₃ medium. This is reflected in the S.F. (separation factor, defined as the ratio of the $K_{d,w}$ values) values which peaked at 3 M for both the mineral acid medium



Fig. 2. Effect of equilibration time on the batch uptake of Y^{3+} from 3 M HNO₃ and 4 M HCl. The results indicate average of triplicate measurements.



Fig. 3. Batch distribution data of Sr(II) and Y(III) using the extraction chromatographic resin material from varying concentrations of HCl and HNO₃.

(Table 2). Though the decrease in the $K_{d,w}$ values for Y(III) was to a much lower extent in HCl medium as compared to that observed in HNO₃ medium, a steady increase in $K_{d,w}$ value of Sr(II) in HCl medium resulted in a peak S.F. value at 4 M HCl. In view of the higher separation factor values observed at 3 M HNO₃ as the feed solution, separation studies using columns were attempted subsequently at this acidity.

3.1.3. Saturation uptake capacity

The saturation metal ion uptake capacity with respect to Y(III) sorption was estimated by equilibrating 5 mL of the feed solution (containing 1 g/LY in 4 M HNO₃ spiked with ⁹⁰Y tracer) with 500 mg of the resin for 48 h. The radioactivity in the feed at the start of the experiment and the supernatant after equilibrating for 48 h were used for calculating the saturation uptake capacity of the TODGA resin which was found to be 0.7 mequiv./g of the extraction chromatography resin material. In view of low $K_{d,w}$ values encountered with Sr(II) no such uptake data was generated for Sr.

3.2. Column studies

Column studies were carried out using a column made with the specifications given in Table 1. For all the column studies, it was ensured that the column was uniform (without any air gap) and the flow rates were uniform. For all the column studies the flow rate was fixed at 0.3 mL per minute and the experiments were carried out at ambient temperature.

3.2.1. Breakthrough profiles

The breakthrough profiles of Sr(II) and Y(III) from HCl and HNO₃ medium using a 0.4 cm \times 13.8 cm column containing about 1 g of

Table 2

Separation behaviour of 90 Y and 90 Sr (studied using 85,89 Sr as the surrogate) from HCl and HNO₃ feed solutions. The results indicate average of triplicate measurements.

Acid concentration (M)	Separation factor (S.F.) values	
	HCl	HNO ₃
0.01	0.92 ± 0.01	2.11 ± 0.02
0.1	0.48 ± 0.01	143 ± 0.99
1	4.77 ± 0.05	741 ± 5.76
2	63.2 ± 0.3	863 ± 3.32
3	139 ± 1.12	1170 ± 6.71
4	95.1 ± 0.91	189 ± 1.22
6	62.3 ± 0.07	702 ± 3.55



Fig. 4. Breakthrough profiles of Sr(II) (using 85,89 Sr tracer) and Y(III) (using carrier free 90 Y) from 3 M HCl and 3 M HNO₃ feed solutions. Flow rate: 0.3 mL/min; temp. = 24 ± 0.1 °C.

resin material (Table 1) suggested that ^{85,89}Sr comes out of the column almost immediately after being loaded on to the column suggesting negligible uptake in the resin which is in conformity with the batch sorption data presented above. As the ^{85,89}Sr tracer used to carry out these experiments contained significant amount of carrier (concentration of Sr in the solutions was about 10^{-4} M) only slight retention was seen. As will be seen in subsequent studies (Section 3.2.5), ⁹⁰Sr was retained in the column was not eluted in the loading and washing stages using 3 M HNO₃. On the other hand, no breakthrough was observed even after passing 50 column volumes when carrier free ⁹⁰Y was used in the feed (3 M HNO₃). The breakthrough profiles are plotted together in Fig. 4. As no breakthrough of ⁹⁰Y was seen, a feed containing 1 g/L Y in 4 M HNO₃ which was spiked with 90Y tracer was used for the subsequent breakthrough studies. Similar to the results published by Ansari et al. who used Eu(III) carrier in their extraction chromatography studies [20], about 20 column volumes were required before any breakthrough of ⁹⁰Y.

3.2.2. Column capacity and elution behaviour

The capacity of the column (details given in Table 1) with respect to Y(III) was evaluated using the feed solution containing 1 g/L Yspiked with 90 Y. In a triplicate experiment it was observed that 0.65 ± 0.01 mequiv. of Y could be loaded on the column at 4 M HNO₃ without any discharge of the 90 Y activity which is in agreement with the saturation uptake capacity reported above. Elution experiments were performed after loading the column with the desired pure tracers. The elution behaviour of Y(III) was investigated by employing 0.01 M HNO₃ and 0.01 M EDTA (pH 2.0). The elution with 0.01 M HNO₃ was found to be extremely slow and a broad band was observed and about 20% elution was in 15 bed volumes (Fig. 5). On the other hand, a sharp band with quantitative elution in about 6 bed volumes was noticed when 0.01 M EDTA was used as the eluent (Fig. 5).



Fig. 5. Elution profiles of Y(III) from the column using 0.01 M HNO₃ and 0.01 M EDTA at pH 2.0. Flow rate: 0.3 mL/min; temp. = 24 ± 0.1 °C.

3.2.3. Reusability of the column

The reusability of the extraction chromatographic resin material was required to be investigated by repeatedly loading and eluting 90 Y. The feed solution contained 1 g/L Y(III) solution in 4 M HNO₃ spiked with 90 Y tracer. The breakthrough profiles from the column studies are superimposed in Fig. 6 from 3 successive runs which indicated that the breakthrough capacity decreased steadily with continuous use from 20 column volumes in the first run to 8 column volumes in the third run. This observation indicated possible leaching of the loaded extractant. It is required, therefore, to develop resin materials with the diglycolamide (DGA) moiety grafted to the resin surface.

3.2.4. Effect of absorbed dose

Due to the high energy beta particles associated with 90 Y, it was pertinent to study the performance of the resin after it was exposed to varying amounts of the dose in 60 Co irradiator with a



Fig. 6. Reusability data of the extraction chromatographic resin as indicated by successive runs. Feed: 1 g/LY in 3 MHNO_3 . Flow rate: 0.3 mL/min; temp. = $24 \pm 0.1 \degree$ C.



Fig. 7. Irradiation stability of the resin as indicated by the breakthrough profiles as a function of the absorbed dose. Feed: 1 g/L Y in 3 M HNO_3 . Flow rate: 0.3 mL/min; temp. = $24 \pm 0.1 \degree$ C.

dose rate of 3.2 kGy per hour. Effect of gamma irradiation on the resin performance was studied up to an absorbed dose of 50 Mrad. The results are shown in Fig. 7 and the decrease in the breakthrough volumes conclusively proved degradation of the resin with increasing absorbed dose. Zhang et al. [23] have also reported significant degradation of the TODGA based XCR from batch uptake studies involving Nd(III).

3.2.5. Separation of carrier free ⁹⁰Y from ⁹⁰Sr

Separation of carrier free ⁹⁰Y from the mixture of ⁹⁰Sr and ⁹⁰Y (present as the daughter product of ⁹⁰Sr) was accomplished by loading a known amount of the tracer mixture onto a column containing about 1 g of the XCR as mentioned above (Section 3.2.1). Loading of the tracer mixture was done at 4 M HNO₃ and washing up to 10 column volumes indicated negligible activity coming out of the column. Subsequently, elution was carried out using 0.01 M HNO₃ and the profiles of elution are given in Fig. 8. The first peak was due to ⁹⁰Sr and the second broad peak was due to ⁹⁰Y. Lack of getting a sharp ⁹⁰Y peak is attributed to two factors, (i) slow elution due to inefficient stripping and (ii) tailing of ⁹⁰Sr peak which added up and led to a broader ⁹⁰Y peak. Moreover, large eluent volume was required for getting quantitative elution of the product making this method unsuitable. The half-lives of the product obtained in the 10th, 15th and 20th fractions (of 1 mL each) were obtained from slope of the ln(activity) vs time plots (Fig. 9) using Eq. (2) and the data are listed in Table 3. As indicated, the products are contaminated with small fractions of ⁹⁰Sr making them unsuitable for use

A combination of the eluent, 0.01 M HNO₃ for 90 Sr elution and 0.01 M EDTA (at pH 2.0) for 90 Y elution was used for a better product separation as indicated in Fig. 10. The 90 Y fraction is recovered in about 3 column volumes or about 6 mL. The recovered products for the 17th, 22nd and 25th fractions (of 1 mL each) were assayed continuously for about 10 days and the half-life of 90 Y are also listed in Table 3. The half-life data for the 17th and 22nd fractions indicated reasonably good purity of the product. This separation method, therefore, can find possible application for the separation of 90 Y from 90 Sr for subsequent use in radiopharmaceuticals. However, it is required to carry out decomplexation reaction (of the



Fig. 8. Elution profiles of 90 Sr and 90 Y from the column. Loading and washing: 3 M HNO₃; elution: 0.01 M HNO₃. Flow rate: 0.3 mL/min; temp. = 24 ± 0.1 °C.



Fig.9. Decay profile of the purified ⁹⁰Y after second extraction followed by stripping used for half-life calculation.



Fig. 10. Elution profiles of 90 Sr and 90 Y from the column. Loading and washing: 3 M HNO₃; elution: 0.01 M HNO₃ (90 Sr) and 0.01 M EDTA at pH 2.0 (90 Y). Flow rate: 0.3 mL/min; temp. = 24 ± 0.1 °C.

Table 3

Purity test data of the eluted fractions as checked by the half-life method. The errors in the half-life are due to errors in the slope values.

Eluent	Sample identification ^a	Half-life (h)	Remarks
0.01 M HNO ₃	10 mL 15 mL 20 mL	$\begin{array}{c} 82.9\pm 0.2\\ 65.9\pm 0.1\\ 65.4\pm 0.1\end{array}$	Contaminated ^b Contaminated ^b Contaminated ^b
0.01 M EDTA	17 mL 22 mL 25 mL	$\begin{array}{c} 64.0 \pm 0.1 \\ 64.4 \pm 0.1 \\ 65.6 \pm 0.2 \end{array}$	Acceptable purity ^c Acceptable purity ^c Contaminated ^b

^a One mL samples taken from the eluted fractions.

^b Contaminated with small traces of ⁹⁰Sr.

 $^{\rm c}\,$ The purity of these products is acceptable as the half life is very close to that of pure $^{90}{\rm Y}.$

Y-EDTA complex) by acidification prior to subsequent complexation with DTPA, DOTA or their analogs. Though the 90 Y/ 90 Sr activity ratio in the product (after allowing the complete decay of 90 Y followed by liquid scintillation counting of the sample) was about 3×10^4 , purity level of 10^6 (required for radiopharmaceutical uses) can be obtained by coupling with another separation technique as reported previously [26]. Literature reports using an organoceramic hybrid material based chromatography method showed 90 Y/ 90 Sr ratio in the range of 1100-80,000 [12] compared to which the present method is superior. Moreover, the present separation method is far superior to the separation method reported by Horwitz et al. [21] using a similar resin but 0.1 M HCl as the eluent.

4. Conclusions

The present study involved the uptake studies involving ⁹⁰Y and ^{85,89}Sr (surrogate for ⁹⁰Sr) and evaluated the possibility of separating them using extraction chromatography resin containing TODGA as the extractant. The results suggested that it is possible to separate ⁹⁰Y from ⁹⁰Sr using a column containing the XCR material with reasonable purity when the elution of ⁹⁰Sr and ⁹⁰Y is done by 0.01 M HNO₃ and 0.01 M EDTA at pH 2.0, respectively. The order of elution suggests that making the conventional generator is not possible by the present method. However, the recovered ⁹⁰Sr fraction can be reloaded onto the column for a subsequent batch of ⁹⁰Y generation. Marginal contamination due to the presence of ⁹⁰Sr in the product is not ruled out and can be possibly separated by loading on to a crown ether column as reported earlier. However, the reusability of the column has serious limitations as a fresh column needs to be used for each separation.

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