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Studies on separation of ^{90}Y and ^{90}Sr separation from hydrochloric acid solutions using TODGA as the extractant by SLM method

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Abstract

Yttrium-90 is an important radionuclide known for its therapeutic application in nuclear medicine. Solvent extraction studies with N,N,N',N'-tetra-octyldiglycolamide (TODGA) has shown that Y(III) is well extracted in 6 M HCl while at the same time, extraction of Sr(II) is very low leading to a separation factor (D_Y/D_{Sr} = 60,000). This property of TODGA can be exploited for the separation of Y from Sr.

The aim of this present work is to produce carrier free ^{90}Y by using Supported Liquid Membrane (SLM) based separation of Y and Sr with TODGA as the carrier. Solvent extraction studies with various diluents viz. 1-decanol, xylene, MIBK, chloroform etc. indicated that xylene and n-dodecane are most suitable as S.F. >50,000 are obtained. Based on the results, a SLM based separation scheme was developed using 0.1 M TODGA in xylene loaded on a microporous PTFE membrane as a polymeric support and 6 M HCl as feed and 0.01 M HCl as strip phase. The results appear promising for the development of SLM based Y-90 generator. The purity of the product was ascertained by the half life method.

Keywords: Ytteium-90, TODGA, Separation factor, SLM

1. Introduction

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Separation of Y-90 from Sr-90 is an important area of research due to the application of the former in the field of radiopharmaceuticals because of its short half-life ($t_{1/2}$ = 64.2 hrs) and high β emissions (E_{max} = 2.28 MeV) [1]. Out of the various separation methods, solvent extraction methods using diglycolamides have shown promise due to unusually high distribution coefficients of Y(III) from acidic medium while Sr(II) extraction being insignificant under identical experimental conditions [2,3].

Solvent extraction studies with diglycolamide extractants such as TODGA (N,N,N',N'-tetraoctyl diglycolamide) have shown that trivalent Y is well extracted from HCl medium while the extraction of Sr(II) is negligible [2]. A separation factor of ~60,000 was reported at 6 M HCl and a method has been developed for the separation of carrier- free Y-90 [3]. It is well known that diluents play a significant role in the metal ion extraction. In the present studies, an attempt has been made to understand the role of organic diluents on the extraction of ^{90}Y and its separation behaviour from ^{90}Sr. Based on the solvent extraction data; a supported liquid membrane based separation method was developed for obtaining carrier-free ^{90}Y. The purity of the separated product (by solvent extraction as well as SLM method) was ascertained by half life method.

2. Experimental

TODGA was procured from Thermax Ltd, Pune and its purity was checked using IR, PMR, HPLC and elemental analysis. Other chemicals used were of analytical reagent grade. The radiotracers, ^{85,89}Sr and ^{90}Sr were procured from BRIT, Mumbai. ^{90}Y was obtained by irradiating natural Y_2O_3 in Dhruva reactor (100MW Research Reactor) at a flux of 10^{13} n/cm^2/s.

Metal ions distribution studies were carried out by equilibrating suitable volumes of aqueous phase (1mL) in 6 M HCl spiked with radiotracer with equal volume of organic phase (in different diluents) in stoppered glass test tubes for 60 minutes in a thermostated water bath at 25±1°C. The two phases were then centrifuged and assayed radiometrically.

Supported liquid membrane studies were carried out using 20mL glass transport cells with feed / strip solutions stirred at 200 rpm as mentioned in an earlier report [4]. The assay of ^{85,89}Sr was carried out employing a well type NaI(Tl) counter coupled with a multichannel analyzer. On the other hand, estimation of ^{90}Y was carried out by liquid scintillation counter. All the experiments were repeated at least twice and the accepted data were within the error limits of relative standard deviation of 5%.

3. Results and Discussion

3.1. Solvent extraction studies:

The solvent extraction studies with ^{90}Y and ^{85,89}Sr in 6 M HCl were carried out using 0.1 M TODGA in presence of different organic diluents and the results are listed in Table 1. As indicated in the table, non-polar diluents, with the exception of hexone (a polar diluent), were more suitable for the extraction of Y-90 and the highest distribution coefficient was obtained with n-dodecane. We have reported separation factor (S.F.) value of 60,000 with n-dodecane in an earlier report which used slightly higher concentration of TODGA compared to the present case [2]. In this study, the S.F. value was found to be more favourable for xylene. Hence, the subsequent studies were carried out using xylene as the diluent.

Based on this, a separation method was developed for the recovery of pure ^{90}Y from a mixture of ^{90}Y + ^{90}Sr. The method involved preferential extraction of ^{90}Y by 0.1 M TODGA in xylene from 6 M HCl followed by its stripping using 0.01 M HCl. The purity of the product was ascertained from its half-life measurements. Several products samples of ^{90}Y such as: i) after extraction from 6 M HCl, ii) after extraction from 6 M HCl followed by

stripping with distilled water and iii) after several cycles consisting of the extraction and stripping steps were obtained. The half-lives of the products from strip fractions were calculated from the slope of the semi-log plot and were found to be in the range 64.17 to 64.47 suggesting reasonably high purity of the product (Table 2). On the other hand, the ^{90}Y containing extract resulted in much higher half-life suggesting partial ^{90}Sr contamination.

3.2. Transport studies:

The SLM studies were carried out with 0.1 M TODGA in xylene using a microporous PTFE membrane as a polymeric support. The studies on the transport of Sr and Y by TODGA-SLM for a feed phase containing 6 M HCl and receiver phase containing 0.01 M HCl suggested >99% transport of ^{90}Y in 2 h while ^{85,89}Sr transport was almost negligible. The results of the transport experiments indicated the feasibility of the separation of ^{90}Y from ^{90}Sr using a simple transport cell.

^{90}Y samples were removed from the receiver phase after different time intervals (after 40, 60 and 120 minutes) from an SLM experiment carried out using ^{90}Sr and ^{90}Y mixture and the purity of the product was ascertained as a function of time by the half-life method as indicated above under the solvent extraction studies. The half lives were found to be in the range of 64 to 70 (Fig. 1) suggesting that the product was contaminated with ^{90}Sr with increasing sampling time (from receiver phase). This is due to permeation of small amount of ^{90}Sr from the feed side to the receiver side. The product can be purified by passing through a crown ether column as reported by us previously [5].

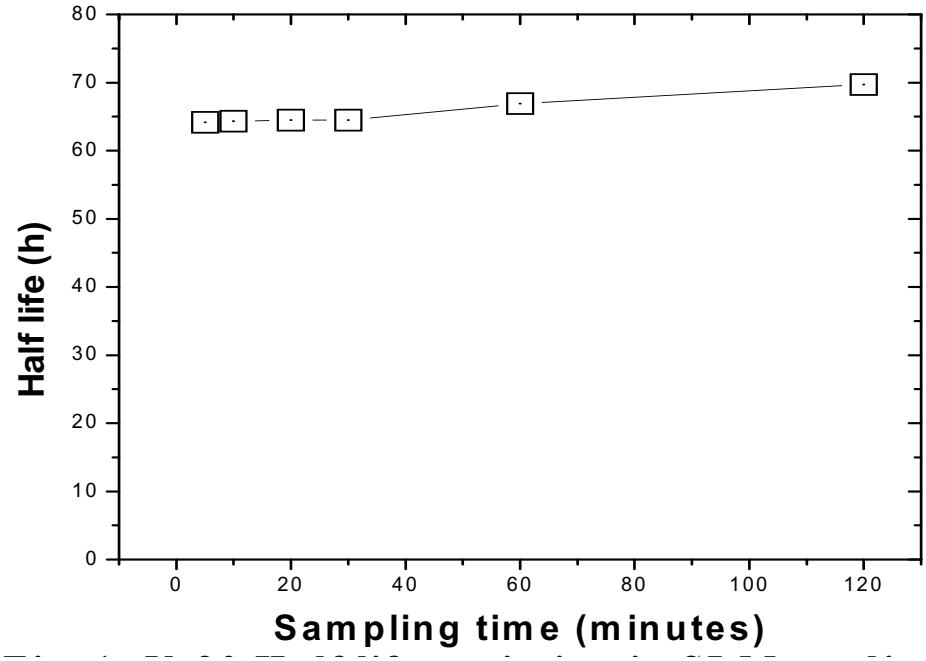

Fig. 1: Y-90 Half life variation in SLM studies

4. Conclusions

The present study indicated that solvent extraction method was more promising compared to the supported liquid membrane based separation method. The product obtained from the SLM method can be purified further by passing through a crown ether column for possible applications [5].

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