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A novel method for the preparation of large-area $^{90}\text{Sr}/^{90}\text{Y}$ sources for the calibration of hand contamination monitors



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HIGHLIGHTS

- Preparation of large-area $^{90}\text{Sr}/^{90}\text{Y}$ sources for the calibration of hand contamination monitors.
- Extraction of $^{90}\text{Sr}/^{90}\text{Y}$ activity followed by preparation of a polymeric solution.
- Pouring the radioactive polymer solution on a surface of a defined area.
- Quality evaluation of the sources.

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ABSTRACT

This paper describes a method for the preparation of large-area $^{90}\text{Sr}/^{90}\text{Y}$ polymer film sources for the calibration of hand contamination monitors. The process consists of solvent extraction of predictable quantity of ^{90}Sr into an organic solvent containing di-tert-butyl-cyclohexano-18-crown-6 (DCH18C6), formation of a polymeric solution of poly(methyl methacrylate) (PMMA), pouring the ^{90}Sr -embedded polymer solution over a surface of a defined area followed by evaporation to create a thin film and peeling-off of the radioactive PMMA film. Quality control tests of the radioactive films were carried out to ensure nonleachability, uniform distribution of activity and stability. Sources having $5 \text{ kBq} \pm 428 \text{ Bq}$ were prepared using this method and routinely used for calibration of contamination monitors.

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1. Introduction

Hand contamination monitors are used to check the presence of radioactive contamination adhered on working personnel's hand while leaving the radioactive laboratory. For routine calibration of hand monitors for beta radiation, an external radioactive source of large area containing ^{90}Sr is required. We have undertaken the development of a new method for the fabrication of large area ^{90}Sr sources for calibration of hand monitors. Preparation of such sources require judicious selection of a technique, careful preparation of source to meet the specifications and evaluation of quality of the source to meet regulatory requirement.

When sources of uniform dimension, suitable activity and acceptable quality from radiological safety point of view are required, a reliable, reproducible and precise method is to be

chosen. Two successful preparative-scale preparations of $^{90}\text{Sr}/^{90}\text{Y}$ reference sources have been reported in the literature by Sundara Rao et al. (1983) and Tsoupko-Sitniko et al. (2002). The former method (Sundara Rao et al., 1983) is based on the deposition of liquid solution of $^{90}\text{Sr}/^{90}\text{Y}$ on VYNS films followed by slow evaporation of solution is applicable to the preparation of small area source. The method reported by Tsoupko-Sitniko et al. (2002) is based on the incorporation of the radionuclide in a thin film of a conducting ion-exchanger polymeric film grown on a metal support. Although there is no technical impediment to adopt the latter method, the implicit need of an intricate electrochemical polymerization process to prepare polymer film of desired attributes from corresponding monomer is a major deterrent. Methods based on the inkjet printers in which the radioactive material is mixed into the ink of inkjet printers which is then used to print patterns on a material substrate such as a sheet of paper is also a promising technique for the preparation surface sources (El-Ali et al., 2003; Hino et al., 2002; Sato et al., 2004; Scafè et al., 2011; Sossi et al., 2005; van Staden et al., 2007). This strategy involves

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many intricate steps including the formulation of radioactive ink of printing density, setting colored inks with different radioactive concentrations, adjustment function of printing concentrations for each ink color, selection of nozzles of printing head to deliver printable radioactivity, assessment of surface contouring and the absorbing capability of the printing medium. While methods based on the inkjet printers are fascinating for the preparation of radioactive source, considerable R&D is required in order to realize its utility in the preparation of ^{90}Sr source. Nevertheless, each radioactive source preparation method has its own distinct advantages and disadvantages but since every institution has different scientific and technical resources, each option has a place.

In the pursuit for a facile approach to undertake the preparation of large-area $^{90}\text{Sr}/^{90}\text{Y}$ reference sources, the scope of using solvent extraction process to extract required quantity of $^{90}\text{Sr}/^{90}\text{Y}$ activity into an organic phase followed by immobilization with a polymer and the radioactive composite making a large area film using the radioactive polymer similar to that used for preparing ^{60}Co sources (Udhayakumar et al., 2008) could be viewed as an interesting proposition. This approach has been simplified in the present work and directed towards the development of a viable technique for the preparation of large-area $^{90}\text{Sr}/^{90}\text{Y}$ reference sources. In the quest for an effective extractant to extract $^{90}\text{Sr}/^{90}\text{Y}$, we have focused our interest towards the use of di-tert-butyl-cyclohexano-18-crown-6 (DCH18C6) owing to its reported selectivity to extract ^{90}Sr from an acidic solution (Horwitz et al., 1990a, 1990b, 1991, 1999; Law et al., 1997).

Herein, we report the development of a novel process which consists of solvent extraction process using DCH18C6 to extract required quantity of $^{90}\text{Sr}/^{90}\text{Y}$ activity into an organic phase, addition of a soluble polymer into the organic solvent, spreading of the radioactive polymer solution on a planar substrate and evaporation of the solution to form a large area film. The factors that influence the extraction of $^{90}\text{Sr}/^{90}\text{Y}$ into the organic phase were identified and a careful control has been exercised to arrive at the conditions resulting optimum extraction. Evaluation of the quality of the radioactive film is also described in this work. To the best of the authors' knowledge, this report is the first to describe the use of DCH18C6 to make such radioactive film.

2. Experimental

2.1. Materials & methods

DCH18C6 (analytical grade) and methyl methacrylate polymers (PMMA), were supplied by Sigma-Aldrich Chemicals, Mumbai, India. Unless otherwise stated, all other reagents used in this study were at least of analytical grade and obtained from Merck India Limited, Mumbai, India. The radioactive isotope $^{85+89}\text{Sr}$, $^{90}\text{Sr}/^{90}\text{Y}$ were available in the Radiopharmaceuticals Division. A shaker consisting of a SS frame with an arm fitted with terry clips at equidistance for holding tubes was used for extraction experiments. A spirit level bubble was used to level the surface. Viscosity of the composite solution was measured using a Rheometer of Model MCR 101 made by Antonpaar, Germany. Radioactivity measurement of ^{90}Sr solution was carried out in a liquid scintillation counter. Radioactive measurement of swipe etc. containing ^{90}Sr was carried out in a GM counter made by Electronics Corporation of India Ltd (ECIL), Hyderabad, India.

2.2. Studies on the extraction of ^{90}Sr into the organic phase

The solvent extraction studies of Sr^{2+} by DCH18C6 in chloroform from nitric acid solutions were carried out using $^{85+89}\text{Sr}$

tracer. In order to study the extractability behavior of Sr in chloroform, equal volumes (1.0 mL) of the aqueous phase spiked with $^{85+89}\text{Sr}$ and the organic phase were taken in cylindrical stoppered glass tubes of 5 mL capacity and shaken in a shaker at room temperature ($25 \pm 0.1^\circ\text{C}$) for 10 min to ensure complete equilibration. Given the simplicity of producing $^{85+89}\text{Sr}$ by neutron irradiation of inexpensive natural SrCO_3 target and convenient means for assaying the activity using a well type NaI (Tl) scintillation counter interphased to a multi-channel analyzer, the scope of using $^{85+89}\text{Sr}$ tracer for solvent extraction experiments seemed very attractive and thus adapted in this investigation.

The two phases were then centrifuged, separated and suitable aliquots of each phase were assayed for radioactivity by liquid scintillation counter. The percent extraction (%) was calculated using the following expression relationship:

$$\% \text{ Extraction} = \frac{A_i - A_f}{A_i} \times 100$$

where A_i and A_f are the initial and final radioactivity of $^{85+89}\text{Sr}$ (used as a surrogate for ^{90}Sr), respectively in the aqueous phase. The radioactivity content (A_i and A_f) was assayed using a well type NaI(Tl) scintillation counter interphased to a multi-channel analyzer (514 keV gamma line, characteristic of ^{85}Sr) by drawing suitable aliquots before and after extraction. Each experiment was repeated three times and data are presented as the average of three experiments. The counting time 400 s was sufficient for obtaining data with the relative detection error < 3%.

In order to study the dependence of the extraction equilibria with time, percentage extraction was determined at 3 M HNO_3 using 0.04 M solution of DCH18C6. The dependence of percentage extraction on nitric acid concentration (0.5–4 M) was studied at 0.04 M solution of DCH18C6. The effect of concentration of the extractant on the distribution of Sr^{2+} was studied at 3 M HNO_3 using DCH18C6 in the concentration range of 0.02–0.1 M. $^{85+89}\text{Sr}$ tracer was used as a surrogate for ^{90}Sr .

2.3. SEM and EDS analysis

The quality of the polymeric film was examined by scanning electron microscope (SEM) and Energy Dispersive X-ray (EDS) analysis of a dummy film prepared in an identical manner as of the active source. EDS microanalysis technique was used to identify the elemental constituents of the electrodeposited samples. We have performed SEM and EDS on specimen coated with a thin (ca. 4 nm) over layer of gold.

2.4. Preparation of the source

Preparation of large-area rectangular $^{90}\text{Sr}/^{90}\text{Y}$ sources were carried out in a specially designed device of 174 mm (l) \times 125 mm (b) \times 61.5 mm (h) made of mild steel with a rectangular cross-section. The device comprise of a base plate 150 mm (l) \times 100 mm (b) \times 20 mm (h); radiography film, a rectangular cut neoprene gasket, a removable rectangular frame of peripheral configuration to be mounted back-to-back on gasket. The device provides a "flexible platform" for mounting a flat rectangular bottom plate. The rectangular frame sits along the outer perimeter of the surface of the flat rectangular base plate to form a rectangular cavity. The base plate and rectangular frame are clamped together with butterfly screws and essentially forms a four walled rectangular structure with a sealed rectangular cavity. A radiography film of dimension similar to the base plate and a rectangular shaped cross-section of neoprene gasket are sequentially placed just above the base plate and mounted on the frame using the butterfly screws. This forms a substantially rectangular

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