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## Applied Radiation and Isotopes

journal homepage: [www.elsevier.com/locate/apradiso](http://www.elsevier.com/locate/apradiso)Usefulness of nano-zirconia for purification and concentration of  $^{125}\text{I}$  solution for medical applications

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## HIGHLIGHTS

- Utility of nano-zirconia sorbent for purification and concentration of  $^{125}\text{I}$  is proposed.
- The process was demonstrated up to 3.7 GBq (100 mCi) activity level.
- Radiocesium impurities could be completely removed from the  $^{125}\text{I}$  solution.
- Greater than 10 fold increase in the radioactive concentration of  $^{125}\text{I}$  was achieved.
- Iodine-125 obtained was found suitable for preparation of brachytherapy sources.

## ARTICLE INFO

## Article history:

Received 10 May 2013

Received in revised form

30 August 2013

Accepted 20 September 2013

Available online 8 October 2013

## Keywords:

Iodine-125

Column chromatography

Nano-zirconia

Radionuclidic purity

Radioactive concentration

## ABSTRACT

This paper described the utility of tetragonal nano-zirconia ( $\text{t-ZrO}_2$ ) to remove trace levels of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  contaminants from  $^{125}\text{I}$  solution obtained from neutron irradiation of natural Xe target. A careful scrutiny of the adsorption parameters of  $\text{t-ZrO}_2$  was considered worthwhile investigating to arrive at the optimum conditions to perform the purification as well as concentration of  $^{125}\text{I}$  solution. The procedure proposed herein provides  $^{125}\text{I}$  of acceptable purity and radioactive concentration for clinical application. The compatibility of the separated product in the preparation of  $^{125}\text{I}$  brachytherapy sources was found to be satisfactory.

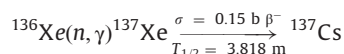
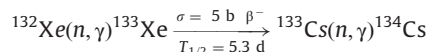
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## 1. Introduction

Iodine-125 is an attractive radionuclide widely used in the preparation of a host of radiolabeled molecules for a variety of *in-vitro* and *in-vivo* medical applications (Bodei et al., 2003; Adam and Wilbur, 2005). The general  $^{125}\text{I}$  production route is through the neutron irradiation of  $^{124}\text{Xe}$  gas to form  $^{125}\text{Xe}$  followed by decay to form  $^{125}\text{I}$  using either enriched  $^{124}\text{Xe}$  or natural Xe targets (Martinho et al., 1984; Joshi et al., 2012). In order to separate  $^{125}\text{I}$  from the irradiated gaseous Xe target into an appropriate chemical form of acceptable quality, two production methods are universally followed. The first method involves dissolution of the radioactivity contents obtained from irradiated target followed by distillation referred to as wet distillation technique (Joshi et al., 2012), while the second entails dry-distillation of the targets (IAEA-TECDOC-1340, 2003).

With an aim to realize the production of  $^{125}\text{I}$  in a small batch size, the scope of using natural Xe target along with the wet

chemical process seemed to be an attractive option of considerable value and followed by some institutions. Natural xenon is a mixture of nine stable isotopes in the following percentages; xenon-124 (0.096); xenon-126 (0.090); xenon-128 (1.92); xenon-129 (26.44); xenon-130 (4.08); xenon-131 (21.18); xenon-132 (26.89); xenon-134 (10.44); and xenon-136 (8.87) (Joshi et al., 2012). Although the isotopic abundance of  $^{124}\text{Xe}$  in natural targets (0.096%) is an inherent disadvantage, relatively high neutron adsorption cross-section of  $^{124}\text{Xe}$  ( $\sigma_{\text{th}} = 128 \pm 15$  barns and resonance activation integral of  $3600 \pm 500$  barns) is highly attractive because of which reasonable yields of the final product can be obtained. However, one of the major impediments in using natural xenon as target is the coproduction of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  owing to the neutron absorption of  $^{132}\text{Xe}$  and  $^{136}\text{Xe}$  as shown below:



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Cesium-134 and  $^{137}\text{Cs}$  would contribute as radionuclidic impurities in the final  $^{125}\text{I}$  product and thus necessitated its removal which is mostly accomplished by distillation (Joshi et al., 2012). Although effective, the need for high temperature operations (typically of the order of several hundred degrees centigrade) is a major disincentive. Suffice it to say that assessing an alternate path such as the column chromatographic separation process is not only an interesting prospect to circumvent such drawbacks, but might also be viewed as an effective strategy due to its technical simplicity, efficacy and amenability for remote operation. In the quest for an effective sorbent to accomplish the separation of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  from  $^{125}\text{I}$ , our attention was turned towards the use of nanomaterials owing to their unique features which can provide an opportunity to suit our requirement. Nanomaterial based sorbents have been explored in recent years as an alternative to more commonly used sorbents based on bulk materials (Liang and Zhang, 2011). The numerous ways in which the nanoscale features of such sorbents interact with ions in aqueous solution on the basis of size, shape, and charge of the particles, offer unmatched potential and could be exploited to achieve efficient separations. In this context, the use of t-ZrO<sub>2</sub> seemed to be an attractive proposition because it not only provides the scope of controlling the adsorptive activity but also selectivity by tuning the pH of the external solution (Chakravarty and Dash, 2013). In order to tap the potential of t-ZrO<sub>2</sub> in the column chromatography separation, we have successfully exploited its usefulness in a number of radiochemical separation and concentration procedures (Chakravarty, 2011; Chakravarty et al., 2012; Chakravarty and Dash, 2013). In view of the appealing attributes, assessing the prospect of using t-ZrO<sub>2</sub> for the separation of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  from  $^{125}\text{I}$  is not only an intuitive proposition but also serves as a model to demonstrate the credentials of nanomaterial based sorbents. This proposition was conceived and directed toward the development of an effective column chromatography based strategy for the separation of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  from  $^{125}\text{I}$  exploiting the fascinating properties of t-ZrO<sub>2</sub>.

In light of the perceived need to realize the scope of using t-ZrO<sub>2</sub>, a careful scrutiny of the sorption parameters of t-ZrO<sub>2</sub> was hence considered worthwhile investigating to arrive at the optimum conditions to perform the separation as well as concentration. In this communication, we describe an overview of the experimental procedure, overall separation procedure and assessment of the quality of the separated  $^{125}\text{I}$  solution for radiopharmaceutical use.

## 2. Materials

Reagents such as hydrochloric acid, ammonium hydroxide, etc. were of analytical grade and were procured from S.D. Fine Chemicals, Mumbai, India. ZrOCl<sub>2</sub>·8H<sub>2</sub>O (99.9%) was obtained from E. Merck, Darmstadt, Germany. Paper chromatography strips were purchased from Whatman International Limited, Maidstone, England. No-carrier added (NCA)  $^{125}\text{I}$  and  $^{137}\text{Cs}$  was readily available in the Radiopharmaceuticals Division as these radioisotopes are regularly used here for fabrication of brachytherapy sources.

Gamma activity of  $^{125}\text{I}$  was assayed using a NaI (TI) scintillation counter. The  $\gamma$ -spectra of  $^{125}\text{I}$  was taken using a Si(Li) detector (Canberra Eurisys, France) coupled with a multichannel analyzer (MCA). HPGe detector coupled with a MCA (Canberra Eurisys, France) was used for the analysis of trace levels of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{126}\text{I}$  impurities present in  $^{125}\text{I}$  and also for their quantitative estimation. The zeta-potential of the nanoparticles were measured using a Zetasizer (Nano ZS/ZEN3600, Malvern Instruments Ltd., UK). X-ray diffraction measurements (10–70°) were carried on the powder for the crystallite size estimation, using monochromatized

Cu-K $\alpha$  radiation on a Philips X-ray diffractometer, Model PW 1927. Silicon was used as an external standard for correction due to instrumental broadening. TEM data were recorded using TEM, JEOL FX microscope (Jeol Ltd., Tokyo, Japan) on the powder sample. The surface area and the pore size analysis were carried out by nitrogen adsorption (BET) technique at 77 K using Quanta-chrome, Autosorb-1 analyzer (Quantachrome Instruments, FL 33426 USA).

## 3. Experimental

### 3.1. Synthesis of t-ZrO<sub>2</sub>

The procedure outlined in our reported paper was followed to synthesize t-ZrO<sub>2</sub> (Chakravarty et al., 2011). In brief, the synthesis procedure involved controlled hydrolysis of zirconyl chloride in ammonical medium which essentially consists of drop wise addition of zirconyl chloride solution in to a solution of 2.5 M NH<sub>4</sub>OH maintaining the pH of the reaction mixture at ~12. The reaction mixture was refluxed at ~95 °C overnight, the precipitate obtained was filtered, washed, dried and calcined at 700 °C for 5 h. The calcined material was crushed and sieved to get particles of 50–100 mesh (149–297  $\mu\text{m}$ ).

The material was synthesized in several (> 10) batches and the structural characterization of the nanomaterial synthesized in each batch was carried out adopting the procedure reported by us earlier (Chakravarty et al., 2010).

### 3.2. Production of $^{125}\text{I}$

Iodine-125 was produced by neutron irradiation of natural Xe gas as reported earlier (Joshi et al., 2012). The irradiation containers made of aluminum were filled with natural Xe gas, sealed, tested for leak tightness and irradiated in the Dhruva reactor of our Institute at a thermal neutron flux of  $5 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$  for 2 weeks. The irradiated Xe containers were transferred to the processing facility, decayed for 50 d and then kept in a liquid nitrogen bath to condense  $^{125}\text{I}_2$  formed during neutron irradiation. After opening the containers containing solidified  $^{125}\text{I}_2$ , the radioactivity contents was dissolved in 0.03 M Na<sub>2</sub>SO<sub>3</sub> solution. The pH of the radioactive solution was subsequently adjusted to ~13 by addition of NaOH solution. The radioactivity of  $^{125}\text{I}$  was measured in a pre-calibrated ion chamber and the trace levels of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  present in it were determined by  $\gamma$ -spectrometry using HPGe detector coupled with MCA by monitoring the 605 keV and 662 keV peaks for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , respectively. The x-ray spectra of  $^{125}\text{I}$  was recorded using a Si(Li) detector coupled with MCA.

### 3.3. Batch sorption studies

#### 3.3.1. Determination of distribution coefficients of I<sup>-</sup> and Cs<sup>+</sup> ions on t-ZrO<sub>2</sub>

Distribution coefficients ( $K_d$ ) of Cs<sup>+</sup> and I<sup>-</sup> ions on t-ZrO<sub>2</sub> matrix were measured at different pH, using  $^{137}\text{Cs}$  and  $^{125}\text{I}$  radio-tracers. In each experiment, 200 mg of sorbent was suspended in 20 mL solution containing the radioactive metal ions, in a 50 mL stoppered conical flask. The flasks were shaken in a wrist arm mechanical shaker for 30 min at room temperature and then filtered. The activities of the solution before and after equilibration were measured in a NaI (TI) counter using appropriate window settings (0–100 keV for  $^{125}\text{I}$  and 600–700 keV for  $^{137}\text{Cs}$ ). The distribution coefficients were calculated using the following

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