High pure, carrier free ⁸⁵Sr and ⁸³Rb tracers obtained with AIC-I44 cyclotron

Ryszard Misiak, Paweł Gaca, Mirosław Bartyzel, Jerzy W. Mietelski

Abstract The method of obtaining carrier free tracers, ⁸⁵Sr and ⁸³Rb from proton-irradiated ^{nat}RbCl target is described. The separation of the radionuclides was done using Sr-Resin, the resin based on a crown ether. Current and some other possible applications of the tracers are discussed.

Key words AIC-144 cyclotron • carrier free tracers • environmental radioactivity • ion exchange chromatography • rubidium-83 • strontium-82 • strontium-85

Introduction

The metallic Rb [1] and RbCl [2, 4] targets are used for the production of ⁸²Sr ($T_{1/2} = 25$ days) [1, 4] and ⁸⁵Sr ($T_{1/2} = 64.8$ days) [2] in carrier-free amounts, in (p,xn) reactions. ⁸²Sr is of interest as a generator for ⁸²Rb which is finding increasing use in positron emission tomography of the myocardium. Metallic rubidium has advantages over RbCl as a target material (higher yield of production) but also carries some risks and complications. The ⁸⁵Sr was separated from the RbCl target [2] by precipitation method using lead carrier. Another chemical separation of ^{82,85}Sr procedure utilizes the technique of ion exchange chromatography [1]. The separation of strontium from rubidium target was done using a Chelex-100 resin in hydrochloric acid media.

Need for ⁸⁵Sr tracer appeared in the course of our search for the best method for chemical yield determination used in ⁹⁰Sr analyses in environmental samples [6]. The ⁸⁵Sr nuclide is well known for this purpose and it is commercially available. However, we tried to obtain it with the new AIC-144 cyclotron at our Institute.

⁸³Sr is formed in the irradiated RbCl target as well. ⁸³Sr is a generator for a carrier-free ⁸³Rb tracer which can be useful for radiochemical works on ⁸⁷Rb in the environment.

Experimental

Four subsequent attempts were done to optimise obtaining of ⁸⁵Sr from ^{nat}RbCl (p.a.) target in (p,xn) reactions at about 30 MeV using the inner beam of AIC-144 cyclotron in Kraków. The irradiation time varied from 1 to 2 h and the nominal beam current was about 1 mA. The irradiated target was dissolved in water and filtered. The filtrate was

R. Misiak[⊠], P. Gaca, M. Bartyzel, J. W. Mietelski The Henryk Niewodniczański Institute of Nuclear Physics, 152 Radzikowskiego Str., 31-342 Kraków, Poland, Tel.: +48 12/ 662 83 94, Fax: +48 12/ 662 84 58, e-mail: Ryszard.Misiak@ifj.edu.pl

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evaporated to dryness and the resulting residue dissolved in 5 M HNO₃ (during the first attempt) or in 3 M HNO₃ + $0.01 \text{ M} (\text{COOH})_2$ (during the next three attempts). The separation and purification of ⁸⁵Sr were made by means of extraction chromatography. At the first stage, to separate the macroamount of target material from the microamounts of obtained radionuclides a Sr-Resin column (0.7 cm in diameter, 7.5 cm long, 100–150 µ, EIChroM) was applied. The columns were conditioned with an appropriate solution $(5 \text{ M HNO}_3 \text{ or } 3 \text{ M HNO}_3 + 0.01 \text{ M (COOH)}_2)$. The Sr-Resin column retained the strontium and barium isotopes while allowing the macroamount of rubidium isotopes and radiocaesium to pass through to waste storage. Ba^{2+} was eluted from the Sr-Resin column with 3 M HNO₃ + 0.01 M (COOH)₂. The ^{83,85}Sr nuclides were eluted with deionised water. After 21 days, which made ⁸³Sr almost completely decayed to ⁸³Rb, the second purification was done using a small Sr-Resin column (0.3 cm in diameter, 3.0 cm long, 100–150 µ, EIChroM). This way pure and carrier-free ⁸³Rb tracer was separated from the pure carrier-free ⁸⁵Sr tracer.

Results

Apart from ⁸⁵Sr ($T_{1/2} = 64.8$ days) also ⁸³Sr ($T_{1/2} = 32.4$ h) was formed in the irradiated targets. The following Rb isotopes were also present: ⁸³Rb ($T_{1/2} = 86.2$ days) the decay product of ⁸³Sr, formed also in the reaction ⁸⁵Rb(p,p2n)⁸³Rb; ⁸⁴Rb ($T_{1/2} = 32.9$ days) formed in the reaction ⁸⁵Rb(p,p2n)⁸⁴Rb; and ⁸⁶Rb ($T_{1/2} = 18.7$ days) formed in the ⁸⁷Rb(p,pn)⁸⁶Rb and/or ⁸⁵Rb(n, γ)⁸⁶Rb reactions. Some other impurities, like ¹³¹Ba ($T_{1/2} = 11.8$ days), ¹³³Ba ($T_{1/2} = 10.5$ a) and ¹³²Cs ($T_{1/2} = 6.48$ days) were formed in Cs(p,xn)Ba or Cs(p,pn)Cs processes from stable caesium contamination of the RbCl target as well. The yield of irradiation for ⁸⁵Sr was about 2 MBq/µAh and that for ⁸³Rb, originating from ⁸³Sr decay, was about 0.7 MBq/µAh.

Initially applied 5 M HNO₃ did not lead to the separation of Sr and Ba. It was successfully replaced by $3 \text{ M} \text{ HNO}_3 + 0.01 \text{ M} (\text{COOH})_2$, according to the distribution



Fig. 1. Elution of Rb⁺, Ba²⁺ and Sr²⁺ ions on the Sr-Resin column. The units on the horizontal axis are the resin volumes ($V_R = 2.9 \text{ cm}^3$) of the column. Flow-rate $0.8 \pm 0.1 \text{ cm}^3$ per minute (for the small Sr-Resin column: flow-rate is about 0.1 cm³ per minute and $V_R = 0.21 \text{ cm}^3$).



Fig. 2. a – Gamma spectrum of the separated ⁸⁵Sr tracer, measured with a low background shielded germanium detector. Beside the main ⁸⁵Sr photopeak at 513.99 keV (98.3%), the summing effect peak at 1028 keV and a weak line at 868 keV (0.012%) are visible. Counting time 1 h. b – Gamma spectrum of the separated ⁸³Rb tracer, measured with a low background shielded germanium detector. Counting time 5.2 h.

coefficients published by Horwitz et al. [3]. The separation process is illustrated in Fig. 1. Strontium and barium retained on the column. 99.8% of radioactive waste consisted of 83,84,86 Rb isotopes, were found in the 2 V_R (V_R - resin bed volume) of effluent from the Sr-Resin column. Ba^{2+} was quantitatively eluted after passing 26 V_R of the 3 M HNO₃ + 0.01 M (COOH)₂ solution. However, as shown in Fig. 1, Sr²⁺ started to be eluted beginning already after passing 20 V_R of the 3 M HNO₃ + 0.01 M (COOH)₂ solution. To obtain ⁸⁵Sr purified from any traces of radiobarium a part of ⁸⁵Sr had to be lost, about 2% of the total activity. Final elution of ⁸⁵Sr was done with deionised water. The contamination of ⁸⁵Sr with Rb or Ba in the eluted fraction after the first step was estimated to be less than 10^{-2} %. However, short-lived ($T_{1/2} = 32.4$ h) ⁸³Sr was also present in this fraction. After 21 days, more than fifteen half-lives, ⁸³Sr has practically completely decayed and ⁸³Rb accumulated. Therefore, the second purification on the smaller column (see "Experimental") was done to separate the accumulated ⁸³Rb from ⁸⁵Sr. The gamma spectra of the finally purified ⁸⁵Sr (Fig. 2a) and ⁸³Rb (Fig. 2b) show very high radionuclidic purity of the nuclides, respectively.

In conclusion, the described method, using Sr-Resin, presents simple a means for production of high pure ⁸⁵Sr and ⁸³Rb. Three attempts done show that a good repeatability of separation has been obtained. Moreover the procedure described above might be used also for producing ⁸²Sr by bombarding RbCl targets with 60 MeV protons.

The ⁸⁵Sr tracer was used in our strontium measurements [5] carried out with environmental samples within the Polish State Committee for Scientific Research (KBN) Project No. 6P04G 07520.

The carrier free ⁸³Rb tracer obtained as a by-product can be useful for radiochemical works on ⁸⁷Rb in the environment. The tracer is not commercially available to the best knowledge of the authors. Moreover, due to the chemical similarity of rubidium and caesium it might also be useful for studies on radiocaesium in environment, in particular in leaching or speciation experiments on ¹³⁷Cs, when two tracers have to be used. It is especially important in analysis of trace amounts of ¹³⁴Cs, which could be present in the environment (e.g. in the vicinity of nuclear power stations or fuel reprocessing plants). However such use of ⁸³Rb requires further studies on comparison of Rb and Cs behaviour during analytical procedures used by researches.

The results of this study demonstrate that the Sr-Resin is likely to find numerous applications beyond the separation of strontium from environmental samples.

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