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A $^{81}\text{Rb}/^{81\text{m}}\text{Kr}$ GENERATOR MADE BY ION IMPLANTATION

G.J. Beyer

Radioisotope Dept., CINR Rossendorf, Dresden, German Democratic Republic

H.L. Ravn and the ISOLDE Collaboration

CERN, Geneva, Switzerland

ABSTRACT

Isotopically pure ^{81}Rb was implanted in plastic foils, and the daughter- $^{81\text{m}}\text{Kr}$ was eluted quantitatively by air or by a 0.9% NaCl solution. The elution yield, the purity of the $^{81\text{m}}\text{Kr}$, and the washing-out losses of ^{81}Rb were studied as functions of the implantation density. It was seen that the densities below 5×10^{12} Rb atoms per square centimetre provide a high yield for elution both by liquid and air. The first 100 MBq implantation-type generator was tested by making a lung-ventilation study in man.

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1. INTRODUCTION

In a previous paper [1] we proposed a new approach for a $^{81}\text{Rb}/^{81\text{m}}\text{Kr}$ generator to be used in nuclear medicine. In this type of generator the ^{81}Rb is implanted in plastic foils [Mylar and polyethylene (PE)]. The elution of the $^{81\text{m}}\text{Kr}$ is performed by flowing air (for ventilation studies) or even a 0.9% NaCl solution (for infusion) over the surface of the plastic foil. Since, in our first experiment [1], only the effect of high elution yield and high purity of the eluted $^{81\text{m}}\text{Kr}$ could be demonstrated at a low-activity level, we report here on the preliminary results of experiments carried out at a high ^{81}Rb activity level.

2. EXPERIMENT

2.1 Production route

The ^{81}Rb was produced at the on-line mass-separator facility ISOLDE-2 at CERN [2]. A Nb-foil target [3] was irradiated with 600 MeV protons from the CERN Synchrocyclotron. The proton beam intensity was $1\ \mu\text{A}$. The Rb atoms formed in the spallation reaction evaporate from the target material and are ionized by means of surface ionization [4]. After passing the magnet of the isotope separator, a pure ion beam of $^{81}\text{Rb}^+$ can be obtained with an intensity of $> 3 \times 10^{10}$ atoms per second.

2.2 Collection

The ^{81}Rb ion beam was collected on foils of different composition (Mylar, PE, Al) during a collection time that was varied from 10 seconds up to several hours. The implantation angle was 90° and the area was of the order of $0.5\ \text{cm}^2$. The number of implanted ^{81}Rb ions was determined from the measured beam intensity as well as from the measured ^{81}Rb activity of each sample.

2.3 Measurements

Two different methods of measuring the elution yield were applied. The ^{81}Rb sample was positioned inside a glass tube, 3 m away from a Ge(Li) detector. An airstream ($> 11/\text{min}$) or a 0.9% NaCl solution (10 to 150 ml/min) was used for the $^{81\text{m}}\text{Kr}$ elution. The detector could see either the 'generator column' or the eluted $^{81\text{m}}\text{Kr}$. The other source was protected by a 5 cm thick lead shielding.

3. RESULTS AND DISCUSSION

The experimental results are illustrated in fig. 1.

3.1 Air elution

At low implantation densities ($< 10^{13}\ \text{cm}^{-2}$), high $^{81\text{m}}\text{Kr}$ elution yields were observed. When the implantation density was increased to $10^{14}\ \text{cm}^{-2}$, the elution yield dropped to only several per cent.

3.2 Liquid elution

In the case of liquid elution, a behaviour similar to that of air elution was observed. High $^{81\text{m}}\text{Kr}$ elution yields have been measured for implantation densities of $< 10^{13}\ \text{cm}^{-2}$. An elution yield of only a few per cent was obtained at implantation densities of $> 10^{14}\ \text{cm}^{-2}$. Between 10^{13} and $10^{14}\ \text{cm}^{-2}$, the elution yield decreased.

3.3 ^{81}Rb washout

The ^{81}Rb washout was measured for liquid elution. At low implantation densities of $< 5 \times 10^{12} \text{ cm}^{-2}$, nearly no losses of ^{81}Rb were observed. In these cases the ^{81}Rb implantation spot was invisible on the foils. An increased implantation density led to increased losses of ^{81}Rb when washing was done with the NaCl solution. The 'soluble' ^{81}Rb was washed out by the first few millilitres of eluant. After this procedure, the further eluted $^{81\text{m}}\text{Kr}$ was clean.

It is interesting to note the obvious change in the behaviour at an implantation density of $5 \times 10^{13} \text{ cm}^{-2}$. Starting from this value, virtually no further ^{81}Rb washout was observed. Simultaneously, the elution yield for both air and liquids dropped to very low values and a visible implantation spot occurred.

The behaviour observed may be explained by surface destruction effects. Note that as long as the implantation damage-channels are well separated, normal generator behaviour can be observed: high elution yield, high purity, no washout losses. Increased implantation density leads to partial destruction of the implantation channels. Consequently, only part of the ^{81}Rb could be reached by the liquids when washing the foil, and growing washout losses were observed. The remaining ^{81}Rb , located in well-isolated channels, showed a normal elution behaviour. High implantation densities ($> 5 \times 10^{13} \text{ cm}^{-2}$) led to surface melting, which closed the implantation channels. Thus the $^{81\text{m}}\text{Kr}$ could no longer reach the surface.

No differences in the behaviour of the Mylar and the PE foils could be found. However, when implantation was made in Al foil, no $^{81\text{m}}\text{Kr}$ elution and no ^{81}Rb washout could be measured. The implantation channels are closed during implantation.

A first real $^{81}\text{Rb}/^{81\text{m}}\text{Kr}$ generator was produced in the following way. Two samples of 2×10^{13} ^{81}Rb atoms were collected during three minutes' collection time each. The implantation density was 10^{13} cm^{-2} ^{81}Rb atoms, and the total ^{81}Rb activity was 0.84 GBq (22.7 mCi) at collection time. Both Mylar foils were introduced into a small PE tube connected with a capillary-tube system. The standard lead-shielding configuration of a commercial ROTOP $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generator*) was used to make a convenient shielding. A first lung-ventilation study was performed at the Cantonal Hospital of Geneva.

4. CONCLUSION

The ion-beam implantation technique is suitable for the production of medical $^{81}\text{Rb}/^{81\text{m}}\text{Kr}$ generators. A high-energy proton beam of 1 μA bombarding a Nb target can produce ^{81}Rb activities for one generator within a few minutes. Only a rather simple low-resolution mass separator is needed for this purpose.

5. OUTLOOK

Further studies are required in order to optimize the conditions for an infusion generator. A lowering of the implantation density and a more homogeneous implantation are obviously the steps to take. It seems to be possible to produce an *in vivo* implantation-type generator. The ^{81}Rb could be implanted in the surface of a plastic tube, which may be used as a catheter to be placed at any required position *in vivo*.

*) Rossendorf $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ Sterile Generator, CINR obtainable from ISOCOMMERZ, Rossendorf, Dresden.

Human blood is then used as the eluting agent that transports the ^{81m}Kr to the place where perfusion has to be studied. Note that an area corresponding to 2 cm (length) of a 3 mm diameter tube can carry approximately 40 MBq (10 mCi) of ^{81}Rb . The experiments are continuing.

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Fig. 1: Performance of the $^{81}\text{Rb}/^{81m}\text{Kr}$ generator (elution yield and washout losses) as a function of the implantation density (for details see text): (●) elution with air; (+) elution with NaCl; (*) ^{81}Rb washout.