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A NEW TYPE OF $^{81}\text{Rb}-^{81\text{m}}\text{Kr}$ GENERATOR FOR MEDICAL USE

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and

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SUMMARY

A new $^{81}\text{Rb}-^{81\text{m}}\text{Kr}$ generator is described. The new generator-type is suitable for ventilation and perfusion studies. By implantation of 60 KeV $^{81}\text{Rb}^+$ -ions into mylar or polyethylene foils, the pure $^{81\text{m}}\text{Kr}$ daughter can be eluted by an air-flow or by an isotonic NaCl solution.

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I. Introduction

The use of ^{81m}Kr for routine imaging of regional lung function, first proposed by Jones et al. (1) and Yano et al. (2), is one of the most important advances in radionuclide clinical investigation in the recent years (3).

Many efforts have been made to produce ^{81}Rb with different types of cyclotrons and to make useful generators. At present there are two principal routes to produce ^{81}Rb . Firstly, the $^{82}\text{Kr} (p, 2n) ^{81}\text{Rb}$ reaction, which requires proton-energies of $E_p > 20$ MeV and secondly the $^{79}\text{Br} (^4\text{He}, 2n) ^{81}\text{Rb}$ reaction, which requires a $^4\text{He}^{2+}$ -particle beam with $E > 28$ MeV. The produced ^{81}Rb is bound to either organic cationic exchange resin beads (applied in the Kr-gastarget route) or to zirconium phosphate (in the case of NaBr-targets). In order to reduce the dead volume the column, its size has to be minimized.

We propose a new type of ^{81}Rb - ^{81m}Kr generator as well as an additional way to produce ^{81}Rb .

The principle of the generator is based on the particular behaviour of the noble gases in the tape transport collection devices used for nuclear spectroscopy studies at the ISOLDE and JINR-Dubna isotope separators. It was observed that the noble gases escaped from the samples if they or their predecessors were implanted into plastic tapes such as mylar. Only by collection in metals or metallized foils could the noble gas be retained efficiently.

II. Experimental

In a preliminary experiment the ^{81}Rb was produced at the ISOLDE II isotope separator facility on-line with the CERN synchrocyclotron (SC) via high energy proton induced reaction ($E_p = 600$ MeV) with an uranium carbide target. The intensity of the proton beam was 2,5 μA , the UC-target density 14 g/cm^2 (4).

The Rb as well as other alkali elements were released from the hot target ($t = 2050^{\circ}\text{C}$) and ionized in a surface ionization ion source. The only ions with mass-number 81 produced under this condition corresponded to ^{81}Rb . The $^{81}\text{Rb}^{+}$ ions were implanted with 60 KeV into different backings:

- mylar foils of different thicknesses
- filter-paper
- polyethylen-foil
- aluminum-foil

A collection interval of 10-20 minutes gave ^{81}Rb samples of the order of 4 MBq (10 μCi). These samples were fixed in a small tube in front of a Ge(Li)-detector. Gamma ray spectra were recorded with and without passing an air stream through the tube. The procedure was repeated with water and isotonic NaCl-solution. Large volumes (150 ml) of NaCl-solution was used for elution in order to determine ^{81}Rb -break through.

III. Results and discussion

The results for gas-elution are shown in table 1. The ^{81}Rb activity remained constant while the $^{81\text{m}}\text{Kr}$ was eluted with high efficiency from the mylar and polyethylene foils independent of foil thickness. The elution yield from the paper-sample was surprisingly small. From the metallic sample practically no $^{81\text{m}}\text{Kr}$ was eluted. A very pure and quantitative $^{81\text{m}}\text{Kr}$ -elution was obtained by liquid elutions with water or 0.9 % NaCl solution as shown in table 2.

The ^{81}Rb break through of $3 \times 10^{-4} \text{ %/ml}$ eluted volume is small and of the same order, as measured for konventional ^{81}Rb - $^{81\text{m}}\text{Kr}$ generators. From the Al-backing practically no Kr was eluted. The ^{81}Rb contained in the filter-paper backing was washed out already with relatively small elution volumes.

The implantation of ^{81}Rb into plastic foils opens a new way to produce $^{81}\text{Rb}-^{81\text{m}}\text{Kr}$ generators. The Rb is not adsorbed at a surface but it is located relatively deep in a solid state material. The $^{81\text{m}}\text{Kr}$ is allowed to diffuse to the surface where it can be transferred efficiently to air or 0.9 % NaCl solution. A 1-2 GBq (25 - 50 mCi) generator requires a backing surface 25 - 50 mm² for an implantation density of 10¹⁴ cm⁻². The use of an isotope separator permits a simple and rapid production of the proposed implantation generator. In fact, Hansen and Feurer (5) proposed the connection of Kr-Gas target with a surface ionization ion source (6,7). The advantage of the implanted type of $^{81}\text{Rb}-^{81\text{m}}\text{Kr}$ generator is as follows:

- extremely small size which permits the injection of a highly active $^{81\text{m}}\text{Kr}$ bolus in a very small volume.
- the entire field of $^{81\text{m}}\text{Kr}$ application is covered.

The only disadvantage at present is the requirement of an isotope separator. The use of such an instrument seems to be very attractive for a continuous production of high purity radionuclides. In recent years a vast amount of new radiochemical techniques have been developed for the study of radioactive nuclei by means of isotope separators on-line to accelerators. Particular high production rates of a number of elements can be obtained by spallation reactions with high energy protons ($E_p \sim 500$ MeV). A review of the developments in target techniques for on-line isotope separator facilities has been given by Ravn (8). A variety of target materials like Nb-powder (9), Zr-foil (10), molten Y and La (11), Mo-foil (12) and Zr-carbide and oxide (13) have been investigated with the aim to determine production rates and release of spallation reaction products including Rb. As an example, the bombardment at ISOLDE of a 100 g/cm² Nb powder target (9) with the 4 μA SC proton beam, results in a ^{81}Rb collection rate of 10¹¹ atoms/s or 14 GBq/h. A corresponding production technology may also be applied to an off-line mode.

Table 1

Efficiency of ^{81m}Kr -elution with air from different ^{81}Rb implanted materials.

⁸¹ Rb sample implanted in	Peak area in cps				Efficiency of ^{81m} Kr- elution
	Airflow ON		Airflow OFF		
	191 keV ^{81m} Kr	446 keV ⁸¹ Rb	191 keV ^{81m} Kr	446 keV ⁸¹ Rb	
mylarfoil 20 μm	220	265	1625	264	86 %
mylarfoil 10 μm	207	275	1728	262	88 %
filter- paper	1440	367	2070	331	30 %
polyehti- lene foil 10 μm	128	158	994	156	87 %
Al-foil 10 μm	490	89	506	90	5 %

Table 2

Summary of liquid elution of implanted ^{81}Rb -samples

^{81}Rb sample implanted in	Efficiency of ^{81m}Kr -elution	Loss of ^{81}Rb in 30 ml elution volume
mylar foil and polyethylene foil	95 %	0.01 %
filter-paper	—	80 %
Al-foil	5 %	0.01 %

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