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Possibility of production of ^{81}Rb via the ^{80}Kr(d,n) reaction at a small cyclotron†

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Summary. A new route has been investigated for the production of the ^{81}Rb(^{81m}Kr) generator parent radionuclide at low energy cyclotrons. The excitation function of the ^{80}Kr(d,n)^{81}Rb reaction was measured up to 14 MeV using natural Kr gas as target, the activation method and a stacked gas-cell irradiation technique. The saturation yield of ^{81}Rb over the energy range $E_d = 14 \rightarrow 6$ MeV amounts to 2.86 GBq/μA. A comparison of this production method with the commonly used ^{82}Kr(p,2n)^{81}Rb route is given.

Introduction

The ^{81}Rb(^{81m}Kr) generator system is widely used in nuclear medicine. Since the original reports on the medical application of 13.3 s ^{81m}Kr [1, 2] extensive medicinal studies have been performed in a large number of laboratories. The generator is used today both for ventilation and perfusion studies. Several routes have been suggested to produce the parent ^{81}Rb ($T_{1/2}$ = 4.58 h), including proton, deuteron, ^{3}He-and α-particle induced reactions (for reviews cf. [3, 4]). Out of those production routes, the proton induced reactions on Kr targets are the most widely used. The salient feature of all the earlier investigated reactions is that sufficient quantities of ^{81}Rb can be produced only at a medium-sized cyclotron. On the other hand, now a large number of low-energy cyclotrons are available, dedicated mainly to the production of positron emitters for Positron Emission Tomography (PET). The possible use of those cyclotrons to the production of other medically important radionuclides is of considerable current interest.

For the production of ^{81}Rb at a low energy cyclotron the ^{80}Kr(d,n)^{81}Rb reaction on an enriched ^{80}Kr gas target seems to be very promising. For this reaction, no report on production, yield or cross-section data was found in the literature. Concerning deuteron induced reactions in general, we found a single report by Gindler *et al.* [5] describing the experimental thick target yield of ^{81}Rb via the ^{82}Kr(d,3n)^{81}Rb reaction. We decided to measure the excitation function of the ^{80}Kr(d,n)^{81}Rb process from threshold up to 14 MeV deuteron energy to be able to investigate the feasibility of ^{81}Rb production at low energies and to compare this process with the proton route at an equivalent cyclotron.

Experimental

The excitation function was measured via activation and identification of the product, using the stacked gas-cell irradiation technique. The details related to targetry, irradiation, activity measurement and data evaluation were similar to those described in our several earlier works from Jülich and Debrecen [cf. 6−8]. Irradiations were done using the external beam of the Debrecen MGC 20E cyclotron at primary deuteron energies of 9.9, 9.7 and 7.4 MeV. Cross sections above 8 MeV were measured at the CV 28 cyclotron in Jülich (E_d = 14.2 MeV). Polyimide foil (15 μm thick, Kapton) windowed, cylindrical, stainless steel gas cells (20 mm $\phi \times 25$ mm), filled with natural krypton to a pressure of 1−2 bar were stacked together (3−5 cells) and irradiated with a low intensity (100 nA) deuteron beam. Thin Ti (21 μm) and Fe (12 μm) foils were inserted in front of the stacks and were used as beam intensity monitors. The number of incident particles was obtained from the charge collected in an isolated target holder (Faraday cup) as well as via the ^{nat}Ti(d,x)^{48}V monitor reaction [9] up to 10 MeV primary deuteron energy and ^{nat}Fe(d,x)^{56}Co monitor reaction [10] at 14 MeV incident deuteron energy. Cross sections [9, 10] multiplied by 0.82 (a normalisation factor) were used for monitoring. This normalisation factor was deduced from new experiments done in connection with the intercomparison of deuteron induced monitor reactions [11]. The results obtained from the monitor reactions and from the charge collection showed good agreement (5−10%).

The activity of each cell was determined without chemical separation via standard high-resolution gamma-ray spectroscopy. The sources were counted at large distances from the detector. On the basis of an independent calibration process it was found that, at the counting distance used, the middle plane of the cell can be regarded as a point source. The estimated relative uncertainty in the detector efficiency was 5%. The measurement was done repeatedly

† Dedicated to the memory of Professor Alfred P. Wolf.
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Table 1. Cross sections of ^{80}Kr(d,n)^{81}Rb nuclear reaction

Energy (MeV)	Cross section (mb)
4.1 ± 0.6	30.3 ± 3.9
5.2 ± 0.4	150 ± 19
5.8 ± 0.5	197 ± 24
6.5 ± 0.5	337 ± 40
6.6 ± 0.4	287 ± 33
7.3 ± 0.4	376 ± 50
7.4 ± 0.3	395 ± 48
8.0 ± 0.3	380 ± 48
8.3 ± 0.7	361 ± 43
9.6 ± 0.6	401 ± 51
10.9 ± 0.5	375 ± 46
12.0 ± 0.4	289 ± 38
13.1 ± 0.3	234 ± 33

for about 12 hours to follow the decay of the radioisotopes concerned and to allow the complete decay of the 30.6 min ^{81m}Rb (IT = 97.8%). The energy degradation along the target was calculated [cf. 12, 13]. The calculation of the cross section and the estimation of the uncertainties in the energy scale and the cross section were done the same way as described in more detail in our earlier publications [cf. 6–8]. The total uncertainty in the cross section amounted to about 13%. The estimated energy scale error was ±0.3 MeV at the first cell and ±0.7 MeV at the last cell in the stack. The decay data used for the investigated isotope ^{81}Rb (E_γ = 190.4 keV (64.3%); E_γ = 446.3 keV (23.3%)) as well as for the monitor isotopes ^{48}V and ^{56}Co were taken from Browne and Firestone [14].

Results and discussion

The measured cross sections and their uncertainties for the reaction ^{80}Kr(d,n)^{81}Rb are given in Table 1. The Q-values of the deuteron induced reactions on various isotopes of Kr allow to deduce the pure isotopic cross section for the ^{80}Kr(d,n)^{81}Rb reaction in spite of the low abundance of ^{80}Kr in natural Kr (2.25%). The threshold of the (d,3n) reaction on ^{82}Kr (having 11% isotopic abundance in natural Kr), which could possibly contribute to the formation of ^{81}Rb, is above the presently investigated energy region. The presented cross sections were calculated assuming a 100% enrichment of ^{80}Kr and represent cumulative cross sections, i.e. production cross sections of the ground state after complete decay of the isomeric state.

The measured cross sections are reproduced as a function of deuteron energy in Fig. 1. From the spline fitted excitation function curve, differential and integral yields of ^{81}Rb from the ^{80}Kr(d,n) reaction were calculated. The integral yield curve is shown in Fig. 2 and represents the saturation yield.

It is instructive to compare the production possibility of ^{81}Rb via the ^{80}Kr(d,n)^{81}Rb reaction investigated in this work with that via the commonly used ^{82}Kr(p,2n)^{81}Rb process described in the literature. The excitation function for the latter reaction was taken from Kovács *et al.* [4] and is shown in Fig. 1. The calculated yields are given in Fig. 2.

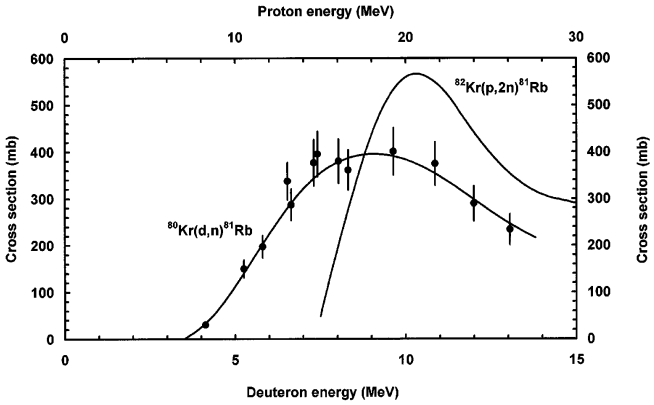

Fig. 1. Excitation function of ^{80}Kr(d,n)^{81}Rb reaction measured in this work. The curve through experimental points was obtained via the spline fitting method. For comparison the excitation function of the ^{82}Kr(p,2n)^{81}Rb reaction (taken from [4]) is also shown. Notice the different energy scales for deuterons and protons.

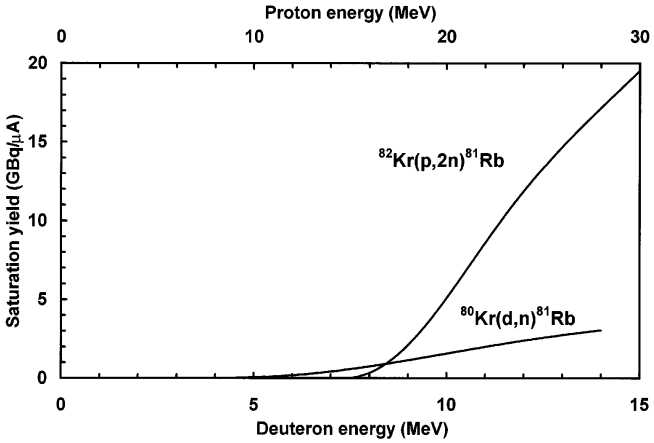

Fig. 2. Integrated saturation yields of ^{81}Rb via ^{80}Kr(d,n)^{81}Rb and ^{82}Kr(p,2n)^{81}Rb reactions calculated from the respective excitation functions. Notice the different energy scales for deuterons and protons.

Since in modern isochronous cyclotrons the maximum energy of the accelerated protons is twice the maximum energy of deuterons, the energy scale for the protons shown in Figs. 1 and 2 is twice that for the deuterons. A comparison of the two processes suggests:

- Over the optimum energy range E_d = 14 → 6 MeV the thick target saturation yield of ^{81}Rb amounts to 2.86 GBq (77 mCi)/μA.
- At commonly available low-energy two-particle cyclotrons the saturation yields of the (d,n) and (p,2n) processes are comparable, for example 1.14 GBq (30.9 mCi)/μA for 9 MeV deuterons and 1.15 GBq (31 mCi)/μA for 17 MeV protons. At $E_p \geq 17$ MeV, the yield via the (p,2n) process increases rapidly.
- At proton energies below 15 MeV the (p,2n) process is not applicable, but using the same cyclotron accelerating deuterons, appreciable quantities of ^{81}Rb could be produced (185 MBq (5 mCi)/μA for 6 MeV deuterons).
- Both processes require enriched targets to get significant yields in the investigated energy regions and to avoid generator shielding problems from high energy gamma rays of the possible impurity radioisotopes of Rb pro-

duced on other stable Kr isotopes. Considering the isotopic composition of natural krypton, the cost of the highly enriched ^{80}Kr is higher; consequently the (d,n) process requires higher initial investment.

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