# EXCITATION FUNCTIONS FOR THE PRODUCTION OF POSITRON EMITTING RADIONUCLIDES $^{82}Sr(^{82}Rb)$ AND $^{73}Se$

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Abstract: Excitation functions were measured up to 33 MeV for the formation of $^{82}Sr$ in $^3He$-particle induced nuclear reactions on enriched $^{82}Kr$, enriched $^{83}Kr$ and natural Kr. The nuclear reaction of major interest is $^{82}Kr(^3He,3n)^{82}Sr$. This process is technically feasible for production at a medium-sized cyclotron but is uneconomical due to the long irradiations needed. In connection with the production of $^{73}Se$ absolute cross sections were determined for $^{75}As(p,3n)^{73}Se$, $^{75}As(d,4n)^{73}Se$ and $^{70}Ge(\alpha,n)^{73}Se$ processes. The (p,3n) reaction in the energy range of $E_p = 40 \rightarrow 30$ MeV is most suitable for production purposes.

($^{82}Sr(^{82}Rb)$ generator, $^{73}Se$, positron emission tomography (PET), stacked-foil technique, cross section, excitation function, isotopic enrichment, thick target yield, radioactive impurity)

## Introduction

With the increasing significance of positron emission tomography (PET) in diagnostic nuclear medicine, the demand for $\beta^+$ emitting radioisotopes, especially in the form of labelled biomolecules, has been increasing in recent years. The short-lived $\beta^+$ emitters $^{11}C(T\frac{1}{2} = 20$ min), $^{13}N(T\frac{1}{2} = 10$ min) and $^{15}O(T\frac{1}{2} = 2$ min) find application at the site of production while a few others with longer half-lives, e.g. $^{18}F(T\frac{1}{2} = 110$ min) and $^{75}Br(T\frac{1}{2} = 98$ min), are used in the vicinity of a cyclotron. A few short-lived $\beta^+$ emitters are available as radionuclide generator products, e.g. $^{82}Rb(T\frac{1}{2}$ 1.2 min) and $^{68}Ga(T\frac{1}{2} = 68$ min). Several other $\beta^+$ emitters are potentially important. We present cross-section data relevant to the production of $^{82}Sr(^{82}Rb)$ and $^{73}Se$.

### $^{82}Sr(T\frac{1}{2} = 25.3$ d)

This radioisotope is used for loading a generator column from which the $\beta^+$ emitting daughter $^{82}Rb$ ($T\frac{1}{2} = 1.2$ min) is milked off periodically. $^{82}Rb$ is a potassium analogue and is mainly used for myocardial blood flow studies [cf. 1].

$^{82}Sr$ is produced in quantities sufficient for making a generator system almost exclusively by spallation of Mo with 500-800 MeV protons [for a review cf. 1]. The cross section of this process is known [2]. At a cyclotron with a proton energy of about 70 MeV the $^{85}Rb(p,4n)^{82}Sr$ process appears to be very promising. Its excitation function has been measured [3] but large scale production has so far not been demonstrated. We thought it worthwhile to investigate the production of $^{82}Sr$ at a medium-sized cyclotron using ($^3He,xn$)-reactions on isotopes of krypton.

Cross sections were measured over the energy range of 10 to 33 MeV for the formation of $^{82}Sr$ in $^3He$-particle induced nuclear reactions on natural Kr, enriched $^{82}Kr$ and enriched $^{83}Kr$. The isotopic composition of each target gas used is given in Table 1. Gas cylinders having thin Ti-windows were irradiated in a series with a well collimated beam of 36 MeV $^3He$-particles. A sketch of the arrangement is given in Fig. 1. The profile of the beam broadens while traversing through the target gas but the whole beam is contained within the gas even in the last cylinder. After the end of irradiation the target gas was removed cryogenically, the walls of the cell were washed and the radiostrontium separated. The radioactivity of the product was determined via $\gamma$-ray spectrometry. The beam current was measured via the monitor reaction $^{nat}Ti(^3He,x)^{48}V$ as well as by collecting the charge in a Faraday cup. The experimental details have been described elsewhere [4,5].

Table 1. Isotopic Compositions of Target Gases

| Isotope | Mole % (= Vol %) | | |
|---|---|---|---|
| | $^{nat}Kr$ | enriched $^{82}Kr$* | enriched $^{83}Kr$* |
| $^{78}Kr$ | 0.35 | 0.8 | |
| $^{80}Kr$ | 2.25 | 9.5 | |
| $^{82}Kr$ | 11.6 | 72.9 | 22.0 |
| $^{83}Kr$ | 11.5 | 16.7 | 73.1 |
| $^{84}Kr$ | 57.0 | 0.1 | 4.9 |
| $^{86}Kr$ | 17.3 | | |

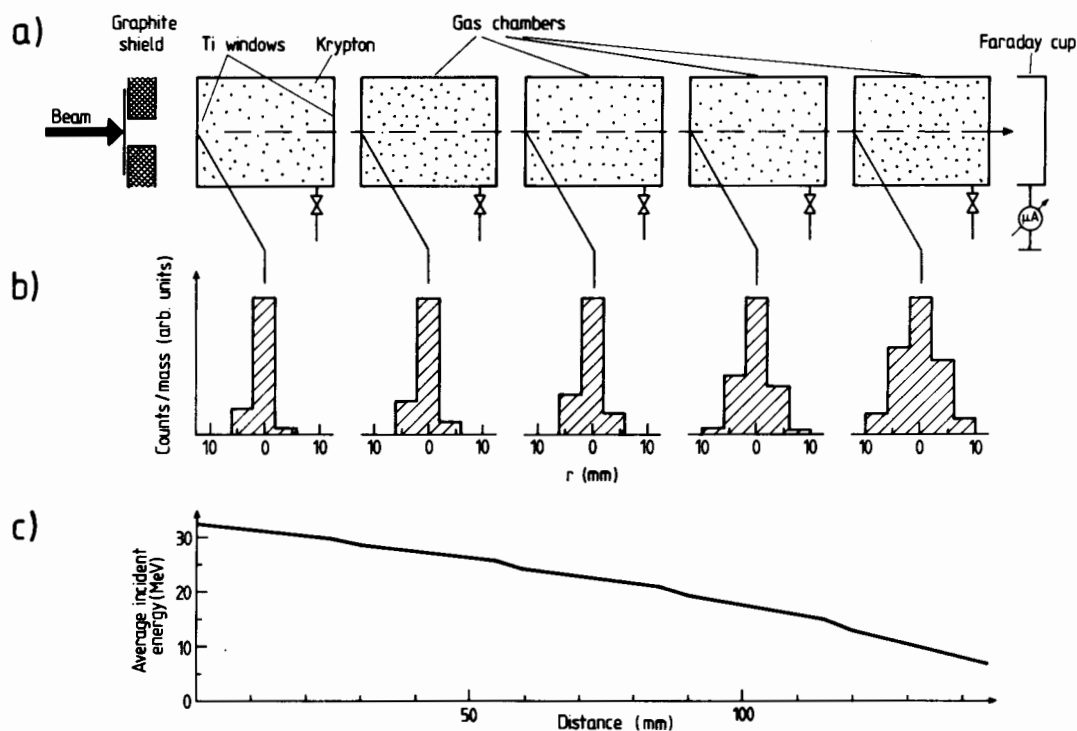*As given by the supplier Isotec Inc. Dayton, Ohio, USA.

Fig. 1. (a) Schematic arrangement for irradiation of gas target chambers. (b) Radial activity distribution in the front window of each gas cell. The distribution reflects the beam profile. (c) Typical calculated energy degradation along the successive gas chambers.
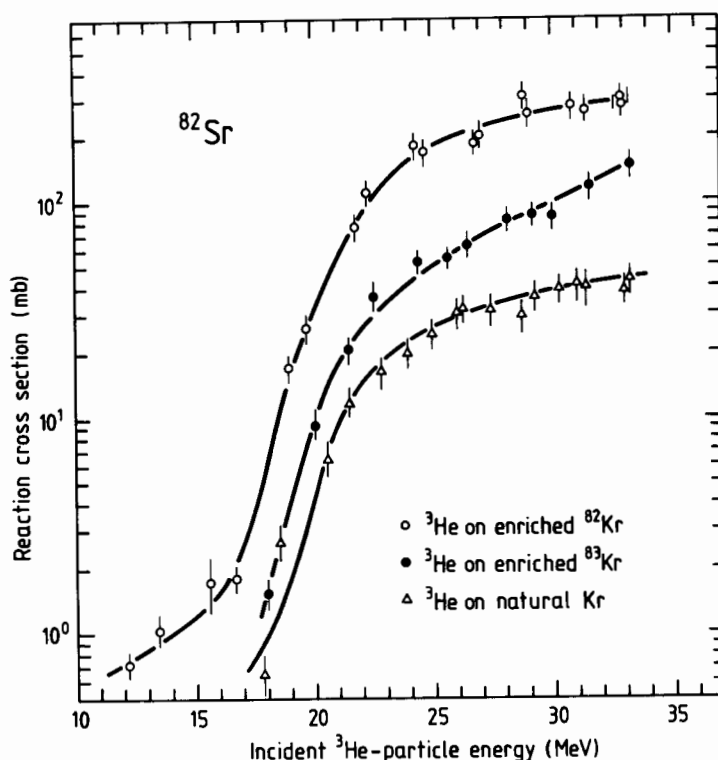


Fig. 2. Excitation functions for the formation of $^{82}$Sr in $^{3}$He-particle induced nuclear reactions on enriched $^{82}$Kr, enriched $^{83}$Kr and natural Kr. The major contributing process is the $^{82}$Kr($^{3}$He,3n)$^{82}$Sr reaction. The effective cross section for the formation of $^{82}$Sr from each target gas depends on the $^{82}$Kr content of each target gas: 72.9, 22.0 and 11.6 %, respectively, in enriched $^{82}$Kr, enriched $^{83}$Kr and $^{nat}$Kr.

The measured excitation functions are reproduced in Fig. 2. Over the investigated energy range only the $^{80}Kr(^3He,n)$-, $^{82}Kr(^3He,3n)$- and $^{83}Kr(^3He,4n)$-processes could possibly contribute to the formation of $^{82}Sr$. From systematics the $(^3He,n)$ cross section is expected to be negligibly small. Since the threshold of the $(^3He,4n)$-reaction was found to be at about 27 MeV [5], the major reaction of interest is $^{82}Kr(^3He,3n)^{82}Sr$. The effective cross section for the formation of $^{82}Sr$ from each target gas was different since the $^{82}Kr$-content in each case was different.

Measurements were also done on $^{83}Sr(T\frac{1}{2} = 32.4$ h), an impurity accompanying $^{82}Sr$. From the effective cross-section data the absolute excitation functions for the $^{82}Kr(^3He,2n)^{83}Sr$ and $^{82}Kr(^3He,3n)^{82}Sr$ processes were deduced. The results, with total errors of about 20 %, are shown as smooth curves in Fig. 3. Evidently the whole energy range of the $(^3He,3n)$ excitation function, i.e. $E_{^3He}$ = 33 → 18 MeV, can be used for the



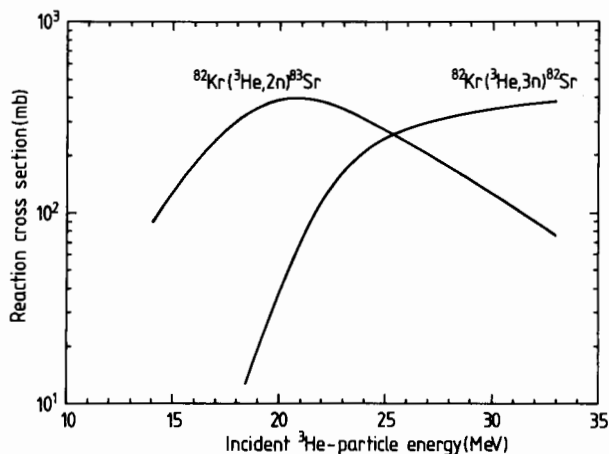Fig. 3. Absolute excitation functions of $^{82}Kr(^3He,2n)^{83}Sr$ and $^{82}Kr(^3He,3n)^{82}Sr$ reactions.

production of $^{82}Sr$. The calculated thick target yield of $^{82}Sr$ from 100 % enriched $^{82}Kr$ gas amounts to about 960 MBq ($\sim 26\mu Ci)/\mu Ah$. The $^{83}Sr$ impurity would also be formed but this should not be a serious problem since its half-life is much shorter. The $^{82}Kr(^3He,3n)^{82}Sr$ reaction is thus interesting for the production of $^{82}Sr$. The practical yield of $^{82}Sr$ achieved using a high current gas target [4] has confirmed the feasibility of the process for production purposes. However, the long irradiation times needed would make it uneconomical.

## $^{73}Se(T\frac{1}{2} = 7.1$ h)

This radioisotope is suited as a positron emitting analogue tracer for sulphur for labelling potentially useful selenoorganic compounds, such as seleno-amino acids, for PET studies. It can be produced via (p,3n) and (d,4n) reactions on arsenic or $(^3He,xn)$ and $(\alpha,xn)$ reactions on isotopes of germanium. We measured excitation functions of all the four processes using the common stacked-foil technique.

Thin samples of arsenic were prepared by electrolytic deposition on Al- or Cu-backing. Irradiations were done with 45 MeV protons and 56 MeV deuterons at low beam currents which were measured via $^{63}Cu(p,n)^{63}Zn$ and $^{27}Al(d,3p2n)^{24}Na$ reactions. The radioactivity of each investigated product was determined via $\gamma$-ray spectrometry. The details are given elsewhere [6]. Due to the more common availability of 40 MeV proton cyclotrons, the (p,3n) reaction is probably more relevant for $^{73}Se$ production than the (d,4n) reaction. We reproduce the data for proton induced reactions in Fig. 4. Our excitation function for the $^{75}As(p,3n)^{73}Se$ reaction is in good agreement with the few data points of Brodovitch et al [7] and the curve of Nozaki et al [8] in the energy range > 26 MeV. The optimum energy range for the production of $^{73}Se$ was found to be $E_p$ = 40 → 30 MeV.

Thin samples of germanium were also prepared by electrolytic deposition on Cu-backing. Irradiations were done with 36 MeV $^3He$-particles and 28 MeV $\alpha$-particles at low beam currents which were measured as described earlier [4]. Due to the use of $^{nat}Ge$ only effective cross sections for the formation of selenium radioisotopes were determined. Only for the $^{70}Ge(\alpha,n)^{73}Se$ reaction the absolute cross sections could be deduced. The excitation function for this process is given in Fig. 5. Our data agree with the curve of Nozaki et al [8] for energies > 18 MeV; in the lower energy region there is some disagreement. Good agreement is also observed with the data of Guillaume et al [9] for energies > 25 MeV; at lower energies there is a large discrepancy.

The calculated thick target yields of $^{73}Se$ and the levels of $^{72,75}Se$ impurities suggest that the $^{75}As(p,3n)^{73}Se$ reaction is the method of choice for production of $^{73}Se$ if a cyclotron with suitable energy is available. The $^{70}Ge(\alpha,n)^{73}Se$ reaction can be used at a compact cyclotron, provided a high-current target is developed.
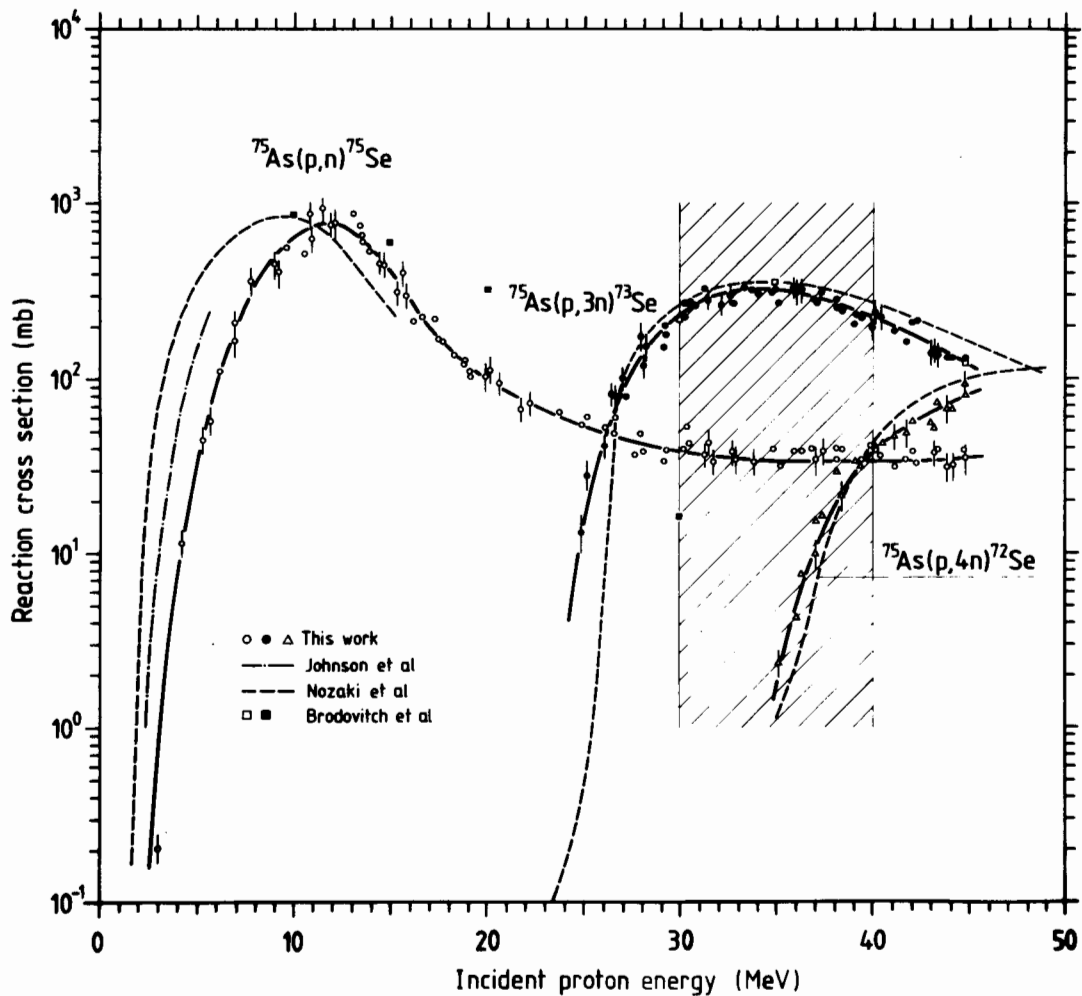
Fig. 4. Excitation functions of $^{75}As(p,xn)^{72,73,75}Se$ reactions. The optimum energy range for the production of $^{73}Se$ ($E_p = 40 \rightarrow 30$ MeV) is shown.
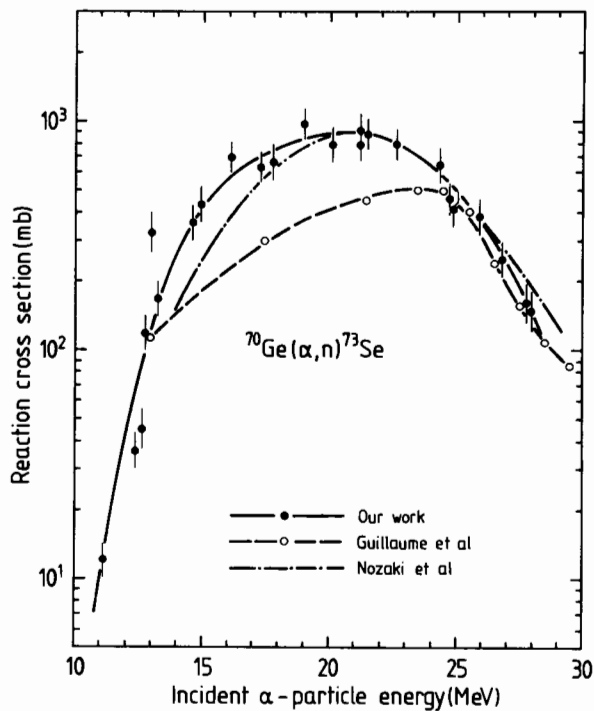


Fig. 5. Excitation function of $^{70}Ge(\alpha,n)^{73}Se$ reaction.

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