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# New nuclear data for production of $^{73}$As, $^{88}$Y and $^{153}$Sm: important radionuclides for environmental and medical applications

I. Spahn[1,a], G.F. Steyn[2], S.A. Kandil[1,3], H.H. Coenen[1], and S.M. Qaim[1]

[1] Institut für Nuklearchemie, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany
[2] Radionuclide Production, iThemba LABS, Somerset West 7129, South Africa
[3] Cyclotron Facility, Nuclear Research Centre, Atomic Energy Authority, Cairo 13759, Egypt

**Abstract.** The radionuclides $^{73}$As, $^{88}$Y and $^{153}$Sm are of considerable significance in environmental and biological research as well as in internal radiotherapy. Cross section measurements related to the n.c.a. production of these radionuclides were carried out for the nuclear reactions $^{nat}$Ge(p,xn)$^{73}$As, $^{nat}$Rb($\alpha$,xn)$^{88}$Y and $^{150}$Nd($\alpha$,n)$^{153}$Sm, using protons and alpha particles of energies up to 100 MeV and 26.5 MeV, respectively. The excitation function of the $^{150}$Nd($\alpha$,n)$^{153}$Sm reaction has been measured for the first time. The existing nuclear database for the other two nuclides could be expanded and strengthened. In the case of $^{153}$Sm additionally the $^{153}$Eu(n,p)$^{153}$Sm reaction cross section was measured averaged over a 14 MeV d(Be) neutron spectrum. The calculation of thick target yields was done and the production possibilities of the three radionuclides were evaluated.

## 1 Introduction

The three radionuclides $^{73}$AS ($T_{1/2}$ = 80.3 d), $^{88}$Y ($T_{1/2}$ = 106.6 d) and $^{153}$Sm($T_{1/2}$ = 1.93 d) are of considerable interest in different fields of sciences for various applications. Arsenic is an element of relevance to environmental and toxicological investigations [1]. Due to its relatively long half-life and soft $\gamma$-ray radiation, $^{73}$As is especially interesting for studying environmental processes. Its production has hitherto not been properly studied. The $\gamma$-ray emitter $^{88}$Y, on the other hand, is a good tracer for studies on biological behaviour of trivalent metals. It has also gained some attention with respect to the development of novel chemical synthesis as well as for investigation of slow metabolic processes. The application of reactor produced $^{153}$Sm is established in internal radiotherapy. The production of all three radionuclides in *no-carrier-added* (n.c.a.) form is of great interest, but has not been established so far in the case of $^{73}$As and $^{153}$Sm. The large scale production of the radionuclide $^{88}$Y is mostly done via the spallation of Mo with 800 MeV protons thereby requiring large irradiation facilities and elaborate chemical processing. An alternative production route involving the $^{nat}$Sr(p,xn)$^{88}$Y reaction has been reported by Kettern et al. [2]; but it leads to the formation of relatively small quantities of the n.c.a. product.

In this work some novel routes leading to the formation of n.c.a. products were investigated. The existing nuclear databases concerning the production of $^{73}$As and $^{88}$Y via proton induced reactions up to 100 MeV on the one hand, and $\alpha$-particle induced reactions up to 26.5 MeV on the other, have been expanded and strengthened. Concerning the formation of $^{153}$Sm two different production routes were investigated, one using fast 14 MeV d(Be)-breakup neutrons and the other employing $\alpha$-particles. Based on the measured cross section data the integral yields of all three radionuclides were calculated and the production possibilities were evaluated.

[a] Present author, e-mail: i.spahn@fz-juelich.de

## 2 Experimental

Nuclear reaction cross sections were determined via the activation technique. In studies on charged particle induced reactions thin samples were prepared, using $^{nat}$Ge, $^{nat}Ge_2O_3$, $^{nat}$RbCI and $Nd_2O_3$ powders as target materials. While investigating the neutron induced reaction for the formation of $^{153}$Sm, however, a relatively thick sample was employed. The methodology used is given below.

### 2.1 Sample preparation

In the measurements of the $^{nat}$Rb($\alpha$,xn)$^{88}$Y and $^{nat}$Nd($\alpha$,n)$^{153}$Sm reaction cross sections, thin samples were prepared using two different techniques. While using $^{nat}$RbCI as target (99.99% pure), the powder was pressed on an Al foil of 10 $\mu$m thickness under a pressure of 6 ton/cm$^2$. It was then covered with a 10 $\mu$m thick Al foil. High-purity $^{nat}Nd_2O_3$ samples, on the other hand, were prepared by a special sedimentation technique [3]. A very fine suspension of the powder (99.999% pure) in water and ethanol was obtained by mixing and stirring. About 200 $\mu$L of the suspension was transferred to the sedimentation cell placed in a desiccator. The liquid evaporated slowly and the material was deposited on a 50 to 100 $\mu$m thick Al foil (99.9% purity) with a diameter of 13 mm. The resulting layer was 10 mm in diameter. The uniformity of the deposit was checked using a microscope. Thereafter the deposit was covered with a 10 $\mu$m thick Al foil. The experimental studies on the $^{nst}$Ge(p,xn)$^{73}$As reaction were carried out using two different types of targets. At iThemba LABS Ge metal foils (99.999% pure, Koch Chemicals Ltd.) of 250 $\mu$m thickness were irradiated. At Forschungszentrum Jülich, $GeO_2$ of natural isotopic composition (99.999% pure, ChemPur) was used and in order to obtain thin targets for irradiation the sedimentation technique described above was applied. This way layers could be obtained with $^{nat}$Ge contents of 0.015 to 0.035 g/cm$^2$. For studying the $^{153}$Eu(n,p)$^{153}$Sm reaction, 1.5 to 2.5 g $^{nat}Eu_2O_3$ (99.9%, Alfa Products) mixed

with about 2.5 g KCl was pressed at 10 ton/cm$^2$ to a pellet of 2.0 cm diameter and 0.3 mm thickness. The pellet was put into an aluminium capsule for irradiation.

### 2.2 Irradiations

Irradiations with protons were done at the compact cyclotron CV 28 and the injector cyclotron of COSY at Forschungszentrum Jülich, Germany, and at the Separate Sector Cyclotron (SSC) at iThemba LABS, Somerset West, South Africa. The incident proton energies were 99, 66, 45, 40, 21 and 18 MeV. All $\alpha$-particle irradiations were done at the CV 28 in Jülich, using an incident projectile energy of 26.5 MeV. The particle flux on the targets was measured by charge integration as well as via the monitor reactions $^{nat}$Cu(p,xn)$^{62,63}$Zn, $^{nat}$Ti($\alpha$,x)$^{51}$Cr and $^{nat}$Cu($\alpha$,X)$^{65}$Zn, the cross sections of which were taken from the literature [4]. All irradiations with protons and $\alpha$-particles were done in the stacked-sample geometry. Several stacks, each consisting of a few thin samples and thin Ti and/or Cu monitor foils, were utilised at particle beam currents of about 100 nA. The energy degradation in each stack was calculated using the computer program STACK, which is based on the Bethe formalism and the tables of Williamson et al. [5].

For studying the $^{153}$Eu(n,p)$^{153}$Sm reaction the samples were irradiated with fast neutrons produced via the breakup of 14 MeV deuterons on a thick Be-target [6, 7] in the 0° direction relative to the deuteron beam. The distance between the sample and the neutron source was 1 cm and the deuteron beam current was kept constant at 5 $\mu$A. The neutron flux density was determined via the monitor reactions $^{27}$Al(n,$\alpha$)$^{24}$Na and $^{56}$Fe(n,p)$^{56}$Mn induced in the Al and Fe foils, respectively. The average neutron flux density effective in the pellet was $1.2 \times 10^{10}$ n cm$^{-2}$ s$^{-1}$.

### 2.3 Measurement of radioactivity

The radioactivity of each thin sample and monitor foil was determined non-destructively, using high-resolution HPGe detectors. The peak area analysis was done using either the software Gamma Vision 6.01 (EG&G Ortec) or Emca 2000 2.03.0 (Silena Int. spa). The distance of each sample to the detector was between 10 and 50 cm, so that the dead time was always below 5%. The detector efficiency was determined using calibrated standard sources from PTB Braunschweig and Amersham International. The corrections due to random and true coincidences being very small were neglected. All investigated radionuclides possess clear and relatively intense $\gamma$-lines. However, the low energy $\gamma$-ray (53.4 keV) of $^{73}$As demanded a suitably calibrated low-energy detector. For the measurement of the $^{153}$Sm radioactivity produced in the thick $Eu_2O_3$ sample, irradiated with neutrons, the 103.18 keV $\gamma$-ray of 31.4% intensity was chosen for analysis. In this investigation, the radioactive product $^{153}$Sm was separated radiochemically from the $^{154}$Eu matrix activity. Details on the separation procedure are given elsewhere [8].

### 2.4 Calculation of reaction cross sections

Cross sections were calculated using the well-known activation equation. The results for $^{73}$As and $^{153}$Sm were obtained from the $\gamma$-rays mentioned above. In case of $^{88}$Y, where two clear $\gamma$-lines of the same nuclide (898 keV and 1836 keV) could be analysed, the resulting activities were averaged for the cross section calculation.

The various individual uncertainties involved in these measurements were similar to those described earlier [2, 9]. The total uncertainty was calculated with error propagation according to Gauss. In general, it amounted to between 8 and 15%. The uncertainties in the primary projectile energy are estimated to be about ±0.2 MeV in case of proton bombardments and about ±0.5 MeV for $\alpha$-particle irradiations. In the case of the $^{150}$Nd($\alpha$,n)$^{153}$Sm reaction, due to low count rates, some of the cross section values have uncertainties up to 50%.

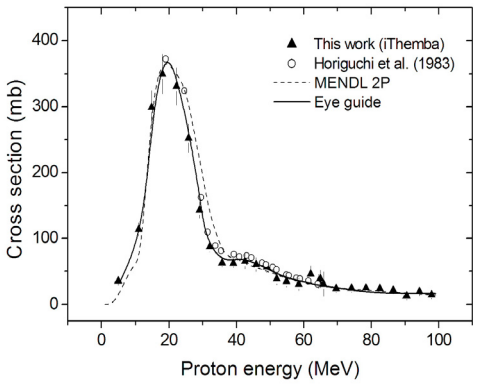


**Fig. 1.** Excitation function of the $^{nat}$Ge(p,xn)$^{73}$As reaction. The dashed curve gives the MENDL-2P results, which are based on the ALICE-IPPE code, while the solid curve gives an eye guide through the data points. For more details see [14].

## 3 Results and discussion

The measured cross section data of the nuclear processes under investigation are shown in figures 1 to 3. If available, data from the literature are shown as well for comparison. The excitation functions of the reactions are discussed below individually in some detail.

### 3.1 Excitation functions

The excitation function of the $^{nat}$Ge(p,xn)$^{73}$As reaction is given in figure 1. A major peak due to the $^{74}$Ge(p,2n)$^{73}$As reaction at about 20 MeV is observed. The peak is, however, rather broad due to some possible contribution from the low-abundance $^{73}$Ge (7.73%) via the $^{73}$Ge(p,4n)$^{73}$As process. A small bump due to the $^{76}$Ge(p,4n)$^{73}$As reaction at about 44 MeV is also visible. Due to the low abundance of the $^{76}$Ge (7.44%) the contribution of this nuclear process is small. In the literature only the experimental data of Horiguchi et al. [10] are available for comparison. Those data from 20 to 65 MeV as well as the results of ALICE-IPPE calculations agree well with our data. The database for the production of $^{73}$As is thus strengthened.

The results for the $^{nat}$Rb($\alpha$,xn)$^{88}$Y process (fig. 2) show that the basic contributing reaction is $^{85}$Rb($\alpha$,n)$^{88}$Y. Our data are in agreement with those of Levkovskij [11] and Iwata [12] within the energy range 17 to 25 MeV but show some discrepancy over the energy range of 13 to 17 MeV. The maximum of the excitation function is shifted to higher energy by about 2 MeV.

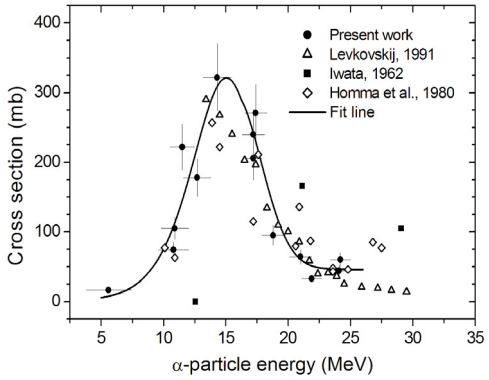


**Fig. 2.** Excitation function of the $^{nat}$Rb($\alpha$,xn)$^{88}$Y reaction. The data of Levkovskij were normalised to refer to $^{nat}$Rb. The curve is a polynomial fit through our data points. For more details see [15].

The data of Homma et al. [13] are too sparse over the investigated energy range.

It should also be pointed out that below 13 MeV only two cross section values were published before. Our data thus describe the threshold energy region in more detail. At $\alpha$-particle energies higher than 25 MeV, the Iwata data show a slight increase, possibly due to the onset of the $^{87}$Rb($\alpha$,3n)$^{88}$Y reaction, which is not seen in the ($\alpha$,n) reaction on enriched $^{85}$Rb measured by Levkovskij.

Due to increasing demands for the radionuclide $^{153}$Sm, co-ordinated attempts have been underway to optimise its production but, so far, it has not been possible to obtain the no-carrier-added product. In this work, therefore, the reactions $^{153}$Eu(n,p)$^{153}$Sm and $^{150}$Nd($\alpha$, n)$^{153}$Sm were investigated.

The 14 MeV d(Be) neutron spectrum averaged cross section for the $^{153}$Eu(n,p)$^{153}$Sm reaction was measured as 0.26 ± 0.06 mb [16]. In figure 3 the measured cross section data of the $^{150}$Nd($\alpha$,n)$^{153}$Sm reaction are given. They refer to 100% abundance of $^{150}$Nd in the target. The maximum cross section of about 45 mb occurs at about 20 MeV.

### 3.2 Production yields

From the excitation functions measured in this work, the optimum energy ranges for the production of the three radionuclides, namely $^{73}$As, $^{88}$Y and $^{153}$Sm were deduced. The differential and integral yields were calculated assuming an irradiation time of 1 h and the proton or $\alpha$-particle beam current as 1 $\mu$A. In table 1 the integral yields of the products via the nuclear reactions under consideration are given. In the case of $^{153}$Sm the value refers to 100% abundance of $^{150}$Nd in the target.

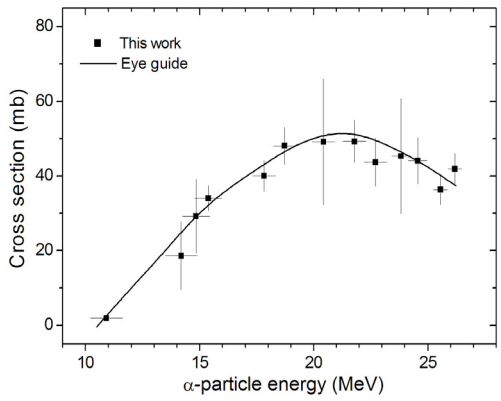


**Fig. 3.** Excitation function of the $^{150}$Na($\alpha$, n)$^{150}$Sm reaction. The curve gives an eye guide through the data points. For more details see [8].

**Table 1.** Integral yields of the products and radioactive isotopic impurities in the investigated nuclear reactions over the optimum energy range.

| Nuclear reaction | Energy range (MeV) | Product yield (MBq·$\mu$A$^{-1}$ · h$^{-1}$) | Impurity (%) |
|---|---|---|---|
| $^{nat}$Ge(p, xn)$^{73}$As | 30 → 18 | 2.4* | $^{74}$As(11.1) |
| $^{nat}$Rb($\alpha$, xn)$^{88}$Y | 26 → 5 | 0.05* | $^{87}$Y(< 0.1) |
| $^{150}$($\alpha$, n)$^{153}$Sm | 25 → 15 | 1.1** | — |

∗ 60 days after EOB
∗∗ at EOB

### 3.3 Evaluation of production possibilities

The long-lived radionuclide $^{73}$As could possibly be produced in a relatively pure form if a decay time of about 60 days is allowed. The optimum energy range for production is $E_p$ = 30 → 18 MeV leading to a possible yield of 4 MBq/$\mu$A·h. The level of radionuclidic impurities at EOB is rather high but within a decay time of about 60 days, it will decrease drastically, the only impurity then being $^{74}$As. Assuming that a $^{nat}$Ge target can withstand 30 MeV proton beam currents of about 100 $\mu$A and that the irradiation time is 30 h, about 7 GBq of $^{73}$As containing 11% $^{74}$As could be obtained at 60 d after EOB. This quantity should be sufficient for environmental studies. If the application would take place after a further time lapse of about 20 days, the level of the hard $\gamma$-ray emitting $^{74}$As impurity would be reduced to less than 5%.

The suitable energy range for production of $^{88}$Y appears to be $E_\alpha$ = 18 → 12 MeV, where the yield of $^{88}$Y amounts to 53 kBq/$\mu$A·h. The level of the $^{87m+g}$Y impurity is high, but it would decay out within about a month after irradiation. In fact if a decay time of this length is allowed, the whole $\alpha$-particle energy range below 26 MeV could be used for production of $^{88}$Y. However, the radionuclide yield via the (p,xn) route reported by Kettern et al. [2] is about 15 times higher than via the ($\alpha$,xn) process. Thus the production route investigated in this work appears to be inadequate for production of $^{88}$Y for application purposes.

Regarding the production of $^{153}$Sm, n.c.a. product could be obtained via both routes studied. The expected yield of $^{153}$Sm

via the (n,p) reaction on $^{153}$Eu using a fast neutron source would be much higher than that using a fission reactor. However, in absolute terms the possible production yield would be very small, due to the considerably lower neutron flux and the relatively small cross section of the reaction. The investigation of the $^{150}$Nd($\alpha$,n)$^{153}$Sm reaction leads to a possible production yield of about 1.1 MBq/$\mu$A·h over the suitable energy range of $E_\alpha = 25 \rightarrow 15$ MeV. The possible batch yield using this nuclear reaction may lead to GBq amount of $^{153}$Sm, provided that an enriched high-current target is available for irradiation. Although this total yield is still much lower than via the (n,$\gamma$) reaction and the cost would be much higher, the advantage of no-carrier-added form may justify the costs.

## 4 Conclusion

The spectrum averaged cross section of the $^{nat}$Eu(n,p)$^{153}$Sm process and the excitation functions of the $^{nat}$Ge(p, xn)$^{73}$As, $^{nat}$Rb($\alpha$, $xn$)$^{88}$Y and $^{150}$Nd($\alpha$, n)$^{153}$Sm reactions were determined, and from them the integral yields of the products were calculated. The results show that production of all the three investigated radionuclides, viz. $^{73}$As, $^{88}$Y and $^{153}$Sm at a cyclotron in no-carrier-added form is possible. The production of $^{73}$As in quantities sufficient for application has been elucidated. The possible yields for the formation of $^{88}$Y and $^{153}$Sm via the $^{nat}$Rb($\alpha$,xn) and $^{nat}$Eu(n,p) processes, respectively, proved to be too low for practical utilisation. The latter process, however, may find an application in case of high-intensity fast neutron sources. In the case of $^{153}$Sm the $^{150}$Nd($\alpha$,n) reaction would lead to sufficient yield of the no-carrier-added product, provided a highly enriched target is used. In view of the increasing significance of $^{153}$Sm, further development of this new route appears worthwhile.

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