

Nuclear data for production of ^{88}Y , ^{140}Nd , ^{153}Sm and ^{169}Yb via novel routes

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Summary. The possibilities of production of the therapy-related radionuclides ^{140}Nd , ^{153}Sm and ^{169}Yb , and of ^{88}Y , in no-carrier-added form were investigated. For ^{88}Y production the nuclear processes $^{\text{nat}}\text{Sr}(p, xn)^{88}\text{Y}$ and $^{\text{nat}}\text{Rb}(\alpha, xn)^{88}\text{Y}$ over the energy ranges $E_p = 14 \rightarrow 9$ MeV and $E_\alpha = 18 \rightarrow 12$ MeV, respectively, can be utilised, although the former reaction is superior, both in terms of yield and radionuclidic purity. For ^{140}Nd production the reactions $^{141}\text{Pr}(p, 2n)^{140}\text{Nd}$ and $^{\text{nat}}\text{Ce}(^3\text{He}, xn)^{140}\text{Nd}$ were investigated in detail. The optimum energy ranges are $E_p = 30 \rightarrow 15$ MeV and $E_{^3\text{He}} = 35 \rightarrow 20$ MeV, respectively. Both processes yield high-purity ^{140}Nd . The former reaction, however, leads to a product yield about 17 times higher than the latter reaction, and is therefore to be preferred. In the case of ^{153}Sm , investigations were done on the $^{153}\text{Eu}(n, p)^{153}\text{Sm}$ reaction with 14 MeV d(Be) neutrons as well as on the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction. The yield of the (n, p) reaction is very low, but the (α, n) reaction over the energy range $E_\alpha = 25 \rightarrow 15$ MeV offers a good potential for production of no-carrier-added ^{153}Sm in quantities sufficient for therapeutic applications. Also for the radionuclide ^{169}Yb a novel route, viz. $^{169}\text{Tm}(p, n)^{169}\text{Yb}$ reaction, was investigated. A critical comparison of this route with the commonly used (n, γ) route, however, showed that, due to the much higher cost involved, the (p, n) process should be utilised only when there is a real need of the high specific radioactivity product.

1. Introduction

The trivalent metals in general, and the lanthanoids in particular, tend to accumulate in the bone marrow, especially in cancerous parts [cf. 1]. This property is being partly utilised in therapy through the use of suitable radionuclides of lanthanoids. In the framework of a COST Action of the EU, which has been running for the last 5 years, we investigated a few novel routes for production of the radionuclides ^{88}Y ($T_{1/2} = 106.65$ d), ^{140}Nd ($T_{1/2} = 3.37$ d), ^{153}Sm ($T_{1/2} = 1.95$ d) and ^{169}Yb ($T_{1/2} = 32.03$ d) at no-carrier-added level. Their major decay data are given in Table 1. Whereas ^{88}Y , ^{140}Nd and ^{169}Yb are almost pure Auger elec-

Table 1. Major decay data of the investigated radionuclides^a.

Radionuclide	Half-life	Mode of decay (%)	Radiation energy (keV)
^{88}Y	106.65 d	EC (99+) β^+ (0.2)	Auger electrons 761
^{140}Nd	3.37 d	EC (100)	Auger electrons
^{153}Sm	1.95 d	β^- (100)	705
^{169}Yb	32.03 d	EC (100)	Auger electrons

a: Data taken from [9].

tron emitters, ^{153}Sm emits low-energy β^- -particles of end-point energy around 705 keV. The radionuclides ^{140}Nd , ^{153}Sm and ^{169}Yb have found application as real therapeutic agents; the nuclide ^{88}Y , on the other hand, is used more as a tracer for studies with the therapeutic radionuclide ^{90}Y . The radionuclides ^{153}Sm and ^{169}Yb are hitherto not available in no-carrier-added form. For internal radiotherapy, however, it is strongly desired to produce those two radionuclides also with a high specific radioactivity. The emphasis in the present work was on nuclear data measurements and comparison of production possibilities under the criteria of yield and radionuclidic purity. Some of the work has already been published [2–4]. Here a brief overview is given of all the measurements done under the project while the new results on the production of ^{88}Y and ^{153}Sm are discussed in detail. The work related to ^{153}Sm production should be of considerable interest since this is the first attempt to obtain this important radionuclide in the no-carrier-added form.

2. Experimental

Nuclear reaction cross sections were determined via the activation technique. In studies on charged particle induced reactions thin samples were used. While investigating the neutron induced reaction for the formation of ^{153}Sm , however, a relatively thick sample was employed. The methodology used in the two types of studies is given below.

2.1 Charged particle induced reactions

Thin samples of high-purity ($> 99.99\%$) $^{\text{nat}}\text{SrCO}_3$, $^{\text{nat}}\text{CeO}_2$, Pr_2O_3 , $^{\text{nat}}\text{Nd}_2\text{O}_3$ and Tm_2O_3 were prepared by a special

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sedimentation technique [cf. 5]. A very fine suspension of the powder in water-free acetone was obtained by mixing and stirring. Additionally, in some cases cellulose nitrate was mixed in concentration of 3 to 5% with respect to the metal oxide (or compound). About 200 μL of the suspension was transferred to the sedimentation cell placed in a desiccator. The acetone evaporated slowly and the material was deposited on a 25 μm thick Cu 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 μm thick Al foil. The thus prepared sandwich (Al, target material, Cu) then served as a thin sample. While using $^{nat}\text{RbCl}$ as target (99.99% pure), instead of sedimentation, the powder was pressed on an Al foil of 10 μm thickness under a pressure of 6 ton/ cm^2 for 15 min. It was then covered with a 10 μm thick Al foil.

Irradiations were carried out using the conventional stacked-foil technique [cf. 2–4, 6]. Protons of energies 12, 16 and 20 MeV, ^3He -particles of energies 25 and 36 MeV, and α -particles of energy 26.5 MeV were extracted from the compact cyclotron CV 28 of the Forschungszentrum Jülich. Several stacks, each consisting of a few thin samples and thin Ti and Cu monitor foils, were irradiated at particle beam currents of about 100 nA. The particle flux on the target was measured by charge integration as well as *via* the monitor reactions $^{nat}\text{Cu}(p, xn)^{62,63}\text{Zn}$, $^{nat}\text{Ti}(^3\text{He}, x)^{48}\text{V}$, $^{nat}\text{Ti}(\alpha, x)^{51}\text{Cr}$ and $^{nat}\text{Cu}(\alpha, x)^{65}\text{Zn}$. The energy degradation in each stack was calculated using the computer program STACK, which is based on the formula for degradation of charged particle energy in matter [8].

The activity of each irradiated sample and monitor foil was measured non-destructively *via* γ -ray spectrometry. For this purpose several HPGe detectors were used and the peak area analysis was done using the software Gamma Vision 5.1, and lately 6.1 of EG&G ORTEC. In general, the samples were measured at a distance of at least 10 cm from the detector, and the monitor foils at distances between 10 and 50 cm. The detector counting efficiency at various distances was determined using standard sources supplied by the PTB Braunschweig or Amersham International. The count rate of each product nuclide was converted to decay rate by correcting for the γ -ray intensity [cf. 9] and the efficiency of the detector. The corrections due to random and true coincidences being very small were neglected.

From the experimentally determined beam flux and the activity of the product in a sample, the desired reaction cross section was calculated using the activation equation. The various individual uncertainties involved in these measurements were similar to those described earlier [cf. 2–4]. In general, the total uncertainty in each measured cross section amounted to 10%–20%. In the case of the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction, due to low count rates, some of the cross section values have uncertainties up to 50%.

2.2 Neutron induced reaction

For studying the $^{153}\text{Eu}(n, p)^{153}\text{Sm}$ reaction, 1.5 to 2.5 g $^{nat}\text{Eu}_2\text{O}_3$ (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. Al and Fe monitor foils were then attached on both sides of the pellet. The thus prepared sample was irradiated with fast neutrons produced *via* the breakup of 14 MeV deuterons on a thick Be-target [cf. 10–12] in the 0° direction relative to the deuteron beam. The distance between the sample and the neutron source was 1 cm. The deuteron beam current was kept constant at 5 μA and the irradiation time was 5 h. The shape of the neutron spectrum encountered in a 14 MeV d(Be) source has already been described [cf. 10, 12]. The neutron flux density was determined *via* the monitor reactions $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ and $^{56}\text{Fe}(n, p)^{56}\text{Mn}$ induced in the Al and Fe foils, respectively. Their excitation functions were taken from the literature [13]. The average neutron flux density effective in the pellet was $1.2 \times 10^{10} \text{ n cm}^{-2} \text{ s}^{-1}$.

From the irradiated Eu_2O_3 pellet, the desired (n, p) reaction product (^{153}Sm) was radiochemically separated. At first the KCl additive was dissolved in water and separated from the residue. Thereafter the residue was dissolved in HNO_3 and Sm separated *via* extraction chromatography using a silica gel column and 25% HDEHP in heptane. The ^{153}Sm activity was assayed *via* HPGe detector γ -ray spectrometry (see above) using the 103.2 keV γ -ray ($I_\gamma = 28.3\%$). The reaction cross section was then calculated using the activation equation, as mentioned above. The total uncertainty amounted to about 25%.

3. Results and discussion

The four radionuclides studied are considered below individually in some detail.

3.1 ^{88}Y ($T_{1/2} = 106.65 \text{ d}$)

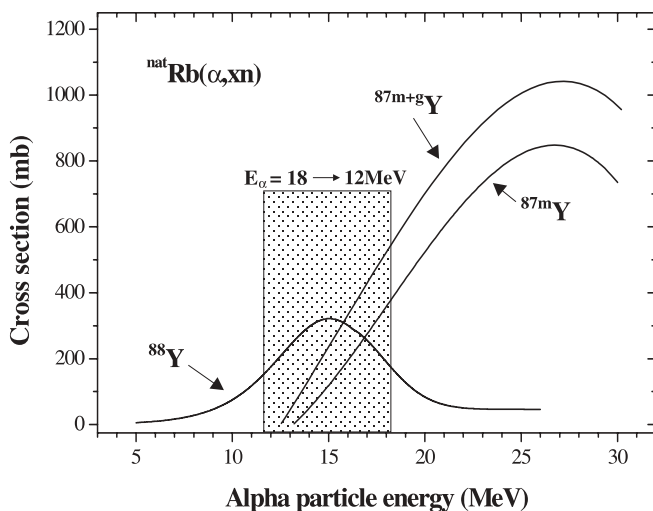
The radionuclide ^{88}Y itself is not of therapeutic interest since it is rather long-lived. However, it has attracted some attention with respect to the development of novel chemical syntheses for possible applications in ^{90}Y -therapy. At low and medium energy cyclotrons ^{88}Y can be produced *via* $^{nat}\text{Sr}(p, xn)^{88}\text{Y}$ and $^{nat}\text{Rb}(\alpha, xn)^{88}\text{Y}$ processes. The $^{nat}\text{Sr}(p, xn)^{88}\text{Y}$ process was recently investigated in our laboratory in detail [2]. Now a study on the $^{nat}\text{Rb}(\alpha, xn)^{88}\text{Y}$ process is described. The numerical data and a detailed discussion of the results are given elsewhere [14]. An overview of the measured excitation functions is given in Fig. 1. The suitable energy range for production of ^{88}Y appears to be $E_\alpha = 18 \rightarrow 12 \text{ MeV}$, where the yield of ^{88}Y amounts to 53 kBq/ $\mu\text{A h}$. The level of the $^{87\text{m}}\text{Y}$ impurity at EOB is very high but due to its short half-life it decays out within a few days. The level of the $^{87\text{m}+g}\text{Y}$ impurity is also high, but it would also decay out within about a month. The β^- -emitting radionuclide ^{90}Y ($T_{1/2} = 2.7 \text{ d}$) was not investigated. However, it should also not disturb after a decay time of this length. In fact if a decay time of one month is allowed, the whole α -particle energy range below 26 MeV could be used for production of ^{88}Y .

A comparison of the two investigated processes for the formation of ^{88}Y , *viz.* $^{nat}\text{Sr}(p, xn)$ and $^{nat}\text{Rb}(\alpha, xn)$, is given in Table 2. The yield of ^{88}Y *via* the (p, xn) route is about 35 times higher than *via* the (α, xn) process. Evidently, for production purposes the former route is to be preferred.

Table 2. Comparison of production routes of no-carrier-added ^{88}Y .

Nuclear process	Suitable energy range (MeV)	^{88}Y yield at EOB (MBq/ $\mu\text{A h}$)	Impurity at EOB (%)	
			$^{87\text{m}}\text{Y}^b$	$^{87\text{m}+g}\text{Y}^b$
$^{\text{nat}}\text{Sr}(p, xn)^a$	14 \rightarrow 9	1.75	4.0	6.0
$^{\text{nat}}\text{Rb}(\alpha, xn)$	18 \rightarrow 12	0.05	11110	3150

a: Data taken from [2];

b: The level of impurity at end of bombardment (EOB) is high. The impurities will, however, decay out completely after about 40 days; the ^{88}Y can then be conveniently used.**Fig. 1.** Trend curves for excitation functions of the $^{\text{nat}}\text{Rb}(\alpha, xn)^{88}\text{Y}$, $^{87\text{m}}\text{Y}$, $^{87\text{m}+g}\text{Y}$ reactions. The curves are based on a large number of data points measured in our laboratory and elsewhere (for details cf. [14]). The suitable energy range for the production of ^{88}Y is shown as a shaded area.

3.2 ^{140}Nd ($T_{1/2} = 3.37$ d)

This radionuclide emits Auger electrons with the estimated mean kinetic energy of about 3 keV [15]. Those Auger electrons are thus concentrated over a cellular dimension. The half-life of ^{140}Nd is also suitable for endoradiotherapy. Its daughter nuclide ^{140}Pr ($T_{1/2} = 3.4$ min, $EC = 49.2\%$, $I_{\beta^+} = 50.8\%$) brings the additional advantage of *in vivo* localisation via positron emission tomography (PET), thus making the radiation dosimetry more quantitative, without introducing any other β^+ -emitting isotopic nuclide, as it is done, for example, in the pair $^{86}\text{Y}/^{90}\text{Y}$.

For production of ^{140}Nd two routes, namely $^{\text{nat}}\text{Ce}(^3\text{He}, xn)^{140}\text{Nd}$ and $^{141}\text{Pr}(p, 2n)^{140}\text{Nd}$, were investigated. A detailed paper on cross section measurements and their theoretical validation has recently been published (cf. [3] and references cited therein). Here, simply a comparison of the two routes is given (cf. Table 3). Evidently, the

Table 3. Comparison of production routes of no-carrier-added ^{140}Nd ^a.

Production route	Suitable energy range (MeV)	^{140}Nd -yield ^b (MBq/ $\mu\text{A h}$)
$^{\text{nat}}\text{Ce}(^3\text{He}, xn)^{140}\text{Nd}$	35 \rightarrow 20	12
$^{141}\text{Pr}(p, 2n)^{140}\text{Nd}$	30 \rightarrow 15	210

a: For details [cf. 3];

b: No isotopic impurity observed at 30 h after EOB.

$^{141}\text{Pr}(p, 2n)^{140}\text{Nd}$ reaction is superior, its yield being about 17 times higher than the yield of the $^{\text{nat}}\text{Ce}(^3\text{He}, xn)^{140}\text{Nd}$ reaction, especially since in both processes no radionuclidic impurity was found.

3.3 ^{153}Sm ($T_{1/2} = 1.95$ d)

This radionuclide is finding increasing application in internal radiotherapy. It is mainly used as EDTMP (ethylenediamino-tetrakis-methylenediphosphonic acid) chelate for treatment of bone metastases, substituting ^{90}Y and ^{32}P . The radionuclide is hitherto produced in a nuclear reactor via the $^{152}\text{Sm}(n, \gamma)^{153}\text{Sm}$ reaction. Because of the relatively high cross section of the reaction, the amount of activity available is high but the specific radioactivity is rather low. Due to increasing demands for this radionuclide, co-ordinated attempts have been underway [cf. 16] 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 $^{153}\text{Eu}(n, p)^{153}\text{Sm}$ and $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ were investigated, to ascertain their potential for the production of high specific activity ^{153}Sm .

The 14 MeV d(Be) neutron spectrum averaged cross section for the $^{153}\text{Eu}(n, p)^{153}\text{Sm}$ reaction was measured as 0.26 ± 0.06 mb [12]. This value is by a factor of about 17 higher than the estimated value of 0.015 mb for the same reaction with fission spectrum neutrons [17]. Thus 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, provided the neutron fluxes would be comparable. In absolute terms, however, the expected ^{153}Sm yield via the (n, p) reaction would be much smaller than via the commonly used (n, γ) reaction, and thus the produced quantity would not be sufficient for medical application.

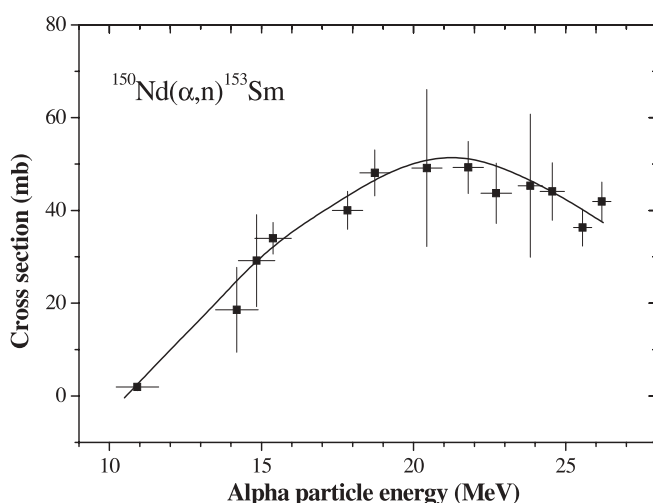
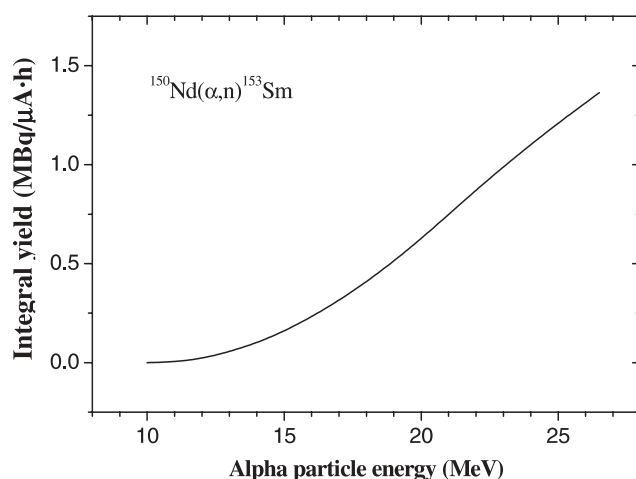
The measured cross sections of the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction are given in Table 4. Some preliminary data were given earlier for $^{\text{nat}}\text{Nd}$ [18]. The measurements were done using $^{\text{nat}}\text{Nd}_2\text{O}_3$ as target material but since ^{153}Sm can be formed only via the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction, the data were corrected for the low abundance of ^{150}Nd (5.64%). The cross sections thus refer to 100% abundance/enrichment of ^{150}Nd . These data have been measured for the first time. The results are reproduced in Fig. 2 as a function of the α -particle energy. The maximum cross section of about 45 mb occurs at about 20 MeV. Since no other Sm radionuclide can be formed from ^{150}Nd over the α -particle energy range investigated in this work, it is possible to produce high-purity ^{153}Sm via this route. The expected yield, however, needs to be considered in detail.

From the measured excitation function of the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction (Fig. 2) differential and integral

Table 4. Measured cross sections of the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction^a.

α -particle energy (MeV)	Cross section (mb)
10.9 ± 0.8	2 ± 0.3
14.2 ± 0.7	19 ± 9
14.8 ± 0.7	29 ± 10
15.4 ± 0.6	34 ± 3
17.8 ± 0.6	40 ± 2
18.7 ± 0.5	48 ± 5
20.4 ± 0.5	49 ± 17
21.8 ± 0.5	49 ± 6
22.7 ± 0.4	44 ± 4
23.8 ± 0.4	45 ± 15
24.6 ± 0.4	44 ± 6
25.6 ± 0.3	36 ± 4
26.2 ± 0.3	42 ± 4

a: Assuming 100% abundance of ^{150}Nd .

**Fig. 2.** Excitation function of the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction. The curve gives an eye guide through the data points.**Fig. 3.** Integral yield of ^{153}Sm calculated from the excitation function given in Fig. 2 (assuming a 100% enriched ^{150}Nd metal target).

yields of ^{153}Sm were calculated assuming a 100% enriched ^{150}Nd metal target, a beam current of 1 μA and an irradiation time of 1 h. The results on integral yields are shown in Fig. 3. Over the suitable energy range of $E_\alpha = 25 \rightarrow 15$ MeV the ^{153}Sm yield amounts to 1.1 MBq/ μA h.

Table 5. Comparison of possible production routes for ^{153}Sm in no-carrier-added form.

Nuclear reaction	Energy range (MeV)	Calculated yield (MBq)
$^{153}\text{Eu}(n, p)^{153}\text{Sm}$	Fission neutrons ^a 14 MeV d(Be) neutrons ^b	10.3 22×10^{-3}
$^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$	Spallation neutrons ^c $25 \rightarrow 15$ ^d	180 2.1×10^3

Assuming

a: 5 g Eu_2O_3 , $\sigma_{(n,p)} = 0.015$ mb, $\Phi = 10^{14}$ n cm^{-2} s⁻¹, irradiation time = 100 h;

b: 5 g Eu_2O_3 , $\sigma_{(n,p)} = 0.26$ mb, $\Phi = 1.2 \times 10^{10}$ n cm^{-2} s⁻¹, irradiation time = 100 h;

c: 5 g Eu_2O_3 , $\sigma_{(n,p)} = 0.26$ mb, $\Phi = 10^{14}$ n cm^{-2} s⁻¹, irradiation time = 100 h;

d: 100% enriched $^{150}\text{Nd}_2\text{O}_3$, $I_\alpha = 100$ μA , irradiation time = 30 h.

A comparison of production yields of no-carrier-added ^{153}Sm via the investigated (n, p) and (α, n) reactions under realistic production conditions is given in Table 5. The expected yield in a nuclear reactor amounts to about 10 MBq. In spite of the much higher (n, p) cross section with 14 MeV d(Be) neutrons the expected yield of ^{153}Sm amounts to only about 22 kBq because the presently available neutron fluxes are low. On the other hand, with future spallation neutron sources, with fluxes $> 10^{14}$ n cm^{-2} s⁻¹, the yield of ^{153}Sm may reach up to 180 MBq. In contrast, the batch yield via the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ reaction may amount to 2.1 GBq. This reaction is then certainly interesting. Although the total yield is still much lower than via the (n, γ) reaction and the cost would be much higher, the advantage of no-carrier-added form may justify the costs. It appears therefore attractive to carry out further development work on the $^{150}\text{Nd}(\alpha, n)^{153}\text{Sm}$ process.

3.4 ^{169}Yb ($T_{1/2} = 32.03$ d)

This radionuclide is gaining importance in endoradiotherapy, because it is almost a pure Auger electron and X-ray emitter. However, since hitherto it has been produced only in a nuclear reactor via the $^{168}\text{Yb}(n, \gamma)^{169}\text{Yb}$ reaction, the specific radioactivity of the product is rather low. We investigated an alternative reaction, viz. $^{169}\text{Tm}(p, n)^{169}\text{Yb}$, at a cyclotron. The details have already been published (cf. [4] and references cited therein). A critical comparison of the two routes showed that the batch yield of ^{169}Yb achieved in the reactor irradiation is much higher than via the cyclotron irradiation. The cyclotron route, however, would lead to a no-carrier-added product in quantities sufficient for medical application and would not rely on the availability of enriched ^{168}Yb . The cyclotron method would involve higher costs and is recommended when really high specific radioactivity is needed.

4. Conclusions

Production of all the four investigated radionuclides, viz. ^{88}Y , ^{140}Nd , ^{153}Sm and ^{169}Yb , in no-carrier-added form is possible utilising charged particle induced reactions at a cyclo-

tron. The production of ^{88}Y and ^{140}Nd in quantities sufficient for applications has been practically demonstrated. For ^{169}Yb production the $^{169}\text{Tm}(p, n)$ reaction is suggested when really high specific radioactivity is needed. In the case of ^{153}Sm , the $^{150}\text{Nd}(\alpha, n)$ 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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