International Conference on Nuclear Data for Science and Technology 2007
DOI: 10.1051/ndata:07238

Comparative studies on the production of the medically important radionuclide ^{124}I via p-, d-, ^{3}He- and α-particle induced reactions

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Abstract. Excitation functions of the reactions $^{\mathrm{nat}}$Sb(α,xn)^{123,124,125,126}I, ^{121}Sb(α,xn)^{123,124}I, $^{\mathrm{nat}}$Sb(^{3}He,xn)^{121,123,124} and ^{126}Te(p,xn)^{123,124,125,126}I were measured from the respective threshold up to 22 MeV α-, 36 MeV ^{3}He- and 100 MeV p-energy, with particular emphasis on data for the production of the medically important radionuclide ^{124}I. The conventional stacked-foil technique was used. The optimum energy range and yield for the production of ^{124}I were determined. The isotopic impurities in the produced ^{124}I are discussed. A comparison of all production routes of ^{124}I is given, including other (p,xn) and (d,xn) nuclear reactions already known. The α- and ^{3}He-induced reactions on antimony are not suitable for the production of ^{124}I due to low batch yield and high impurity level. The commonly used production route ^{124}Te(p,n)^{124}I gives the purest form of ^{124}I and the production can be done at a small-sized cyclotron. Large scale production via ^{125}Te(p,2n) or ^{126}Te(p,3n) reaction is possible, but with a higher impurity level. Cyclotrons with higher proton energy are needed (up to 40 MeV). The final choice will depend upon the tolerable level of impurities.

1 Introduction

The radionuclide ^{124}I ($T_{1/2}$ = 4.18 days; I_{β^+} = 22%; E_{β^+} = 2.13 MeV) is the only long-lived β^+ emitting radioisotope of iodine and finds application both in diagnosis via PET and radiotherapy. It is generally produced using the ^{124}Te(p,n)^{124}I [1] or ^{124}Te(d,2n)^{124}I [2] reaction at low energies. However, other possible production routes deserved detailed investigations to find a high yield, high purity route. The investigation of the ^{3}He- and α-particle induced reactions on $^{\mathrm{nat}}$Sb and the α-particle induced reactions on highly enriched ^{121}Sb should provide a reliable database and clear the discrepancies in old data measurements. Several (p,xn) reaction cross sections on highly enriched ^{126}Te should also be measured for the first time. A comparison of all the ^{124}I production reactions appeared worthwhile.

2 Experimental

The excitation functions of various nuclear reactions on tellurium and antimony were measured via the stacked-foil technique. This involved the preparation of thin layers of $^{\mathrm{nat}}$Sb and ^{121}Sb (99.45% enrichment) by a sedimentation process (for details see [3,4]) and of enriched ^{126}Te (98.69% and 99.8%) by electroplating on Ti foils (for details see [5]). The uniformity of the deposit was checked using a microscope. Thereafter the deposit was covered by a 10 μm thick Al foil. The targets were irradiated together with beam monitors (for details see [3,4, 6]) in the stacked-foil arrangement at low beam currents of about 100 nA to avoid loss of iodine during the irradiation. The irradiations with 36 MeV ^{3}He- and 28 MeV α-particles were performed at the compact cyclotron CV 28. To cover the whole energy range from 5 to 100 MeV with protons, irradiations were done at two cyclotrons at the Forschungszentrum Jülich, Germany, namely the compact cyclotron CV 28 (5–20 MeV) and the injector cyclotron of COSY (20–45 MeV), as well as at the Separate Sector Cyclotron (SSC) (40–100 MeV) of the iThemba LABS, Somerset West, South Africa.

The radioactivity of each thin sample and monitor foil was determined non-destructively, using high-resolution HPGe detectors. The low energy γ-ray (35.5 keV) of ^{125}I demanded a suitably calibrated low-energy detector. All measurements were done repeatedly to follow the decay of the nuclides, and the contributions of different nuclides to the same γ-ray were separated, e.g., 159 keV γ-ray: ^{123}I (13.2 h), $^{123\mathrm{m}}$Te (119.7 d), ^{47}Sc(3.35 d). Cross sections were calculated using the well-known activation equation. Individual uncertainties are given in [3,4] or were similar to [6]. In general, the total uncertainty amounted to between 9 and 23%. In the case of ^{126}Te(p,2n)^{125}I reaction the errors are about 45%. The integral yields were calculated and the impurity levels in all the investigated reactions determined in order to find the optimum conditions for the production of ^{124}I.

3 Results and discussion

For the first time a reliable set of excitation functions for the reactions $^{\mathrm{nat}}$Sb(α,xn)^{123,124,125,126}I has been achieved from the respective threshold of each reaction up to 26 MeV, and discrepancies in older measurements have been removed. Figure 1 shows the excitation function of the $^{\mathrm{nat}}$Sb(α,xn)^{124}I process. The data from the literature are given for comparison. The calculated yield of ^{124}I (1.02 MBq/μAh) in the energy range E_α = 22 → 13 MeV is very low, and the amount of coproduced ^{125}I (13%) and ^{126}I (16%) intolerably high. The $^{\mathrm{nat}}$Sb(α,xn)^{124}I reaction is therefore not suitable for the production of ^{124}I. The long-lived impurities are produced only from ^{123}Sb, therefore we also investigated the (α,xn)

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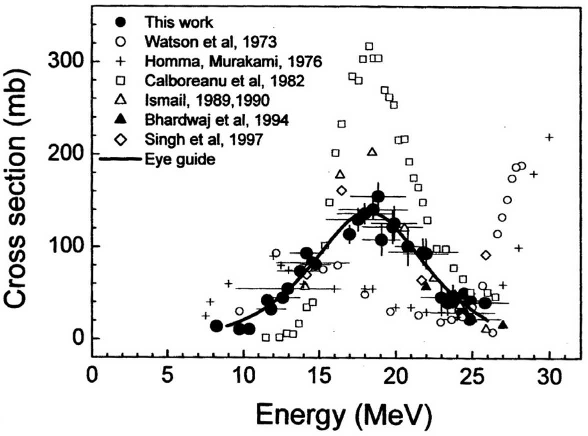

Fig. 1. Cross sections of the ${}^{nat}Sb(\alpha,xn){}^{124}I$ reaction. The eye guide curve is through our data points. For details see [3].

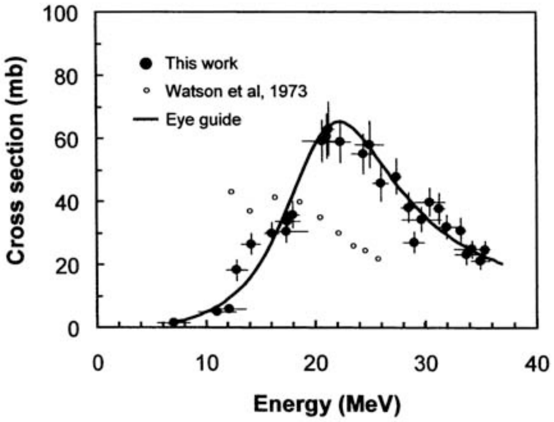

Fig. 2. Cross sections of the ${}^{nat}Sb({}^{3}He,xn){}^{124}I$ reaction. The eye guide curve is through our data points. For details see [3].

reactions on enriched ${}^{121}Sb$. Our new data set for the reactions ${}^{121}Sb(\alpha,xn){}^{123,124}I$ cleared discrepancies in older measurements. As expected, the yield of ${}^{124}I$ is almost doubled and the ${}^{125,126}I$ impurities could not be detected. Nevertheless the ${}^{124}I$ yield is still low and the ${}^{123}I$ impurity (895%) too high. Thus the use of enriched ${}^{121}Sb$ also does not make the α-induced reaction on antimony suitable for the production of ${}^{124}I$.

The excitation functions of the reactions ${}^{nat}Sb({}^{3}He,xn){}^{121,123,124}I$ were also measured thoroughly from the respective threshold up to 36 MeV. Figure 2 shows the excitation function of the ${}^{nat}Sb({}^{3}He,xn){}^{124}I$ reaction.

The calculated yield of ${}^{124}I$ (0.95 MBq/μAh) from ${}^{3}He$-particles on ${}^{nat}Sb$ in the energy range $E_{{}^{3}He} = 35 \rightarrow 13$ MeV is very low. The amount of coproduced ${}^{125}I$ and ${}^{126}I$ is estimated to be only 0.6%, but the level of ${}^{123}I$ impurity is intolerably high. The ${}^{nat}Sb({}^{3}He,xn)$ reaction is therefore also not suitable for the production of ${}^{124}I$.

The above results show that ${}^{3}He$- and α-particle induced reactions on antimony isotopes are inferior to proton and deuteron induced reactions on tellurium, as far as the production of ${}^{124}I$ is concerned.

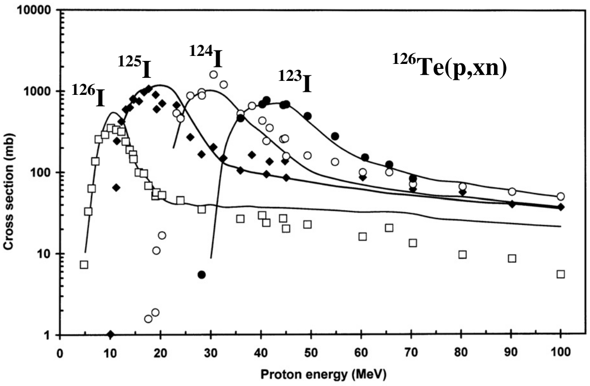

Fig. 3. Cross sections of the ${}^{126}Te(p,xn){}^{126,125,124,123}I$ reactions. The curves show the theoretical calculations using ALICE-IPPE.

It should be mentioned that already in 1995 we showed that the ${}^{124}Te(p,n)$ reaction is ideally suited for the production of ${}^{124}I$ at small cyclotrons with energies below 16 MeV and that it is superior to the earlier used ${}^{124}Te(d,2n)$ reaction due to lower impurity levels. In the energy range $E_p = 12 \rightarrow 8$ MeV the yield is 16 MBq/μAh, with very small ${}^{123}I$ impurity and extremely low ${}^{125}I$ content. This production route gives the purest ${}^{124}I$ and today it is worldwide the preferred production method. Recently Nye et al. [7] have shown that even at lower energies ${}^{124}I$ can be produced for local use. Nevertheless, the yield of the process is limited, therefore we investigated proton induced reactions at intermediate energies which promised higher yields.

Hohn et al. [6] measured in 2001 the excitation functions of the ${}^{125}Te(p,xn){}^{123,124,125}I$ reactions from threshold up to 100 MeV. The optimum energy range was found to be $E_p = 21 \rightarrow 15$ MeV to produce ${}^{124}I$ via the ${}^{125}Te(p,2n)$ reaction. The yield is five times higher than the yield of the (p,n) reaction. The coproduced ${}^{123}I$ (7.4%) decays out within a half-life of ${}^{124}I$. A disadvantage of the process is the somewhat higher ${}^{125}I$ impurity level (0.9%), but, after four days cooling time, it is similar to the impurity level for the ${}^{124}Te(d,2n)$ reaction (1.7% ${}^{125}I$ at EOB), which is still in use for production of ${}^{124}I$ at some places.

We determined now the excitation functions of the nuclear reactions ${}^{126}Te(p,xn){}^{123,124,125,126}I$, and the results are shown in figure 3. The higher energy range was investigated earlier. Here in particular the range below 40 MeV was thoroughly studied. The experimental cross sections agree fairly well with the theoretical calculation using the code ALICE-IPPE, shown as curves in figure 3. With ALICE-IPPE calculations good predictions can be made for (p,xn) reactions for energies up to 30 MeV above the threshold of the reactions. Beyond that, however, the model overestimates the cross sections.

For ${}^{124}I$ production the optimum energy range was found to be $E_p = 38 \rightarrow 28$ MeV with a yield of about 222 MBq/μAh. At lower energy large amount of ${}^{125}I$ is coproduced; at higher energy the amount of ${}^{123}I$ increases to an intolerable level. The impurities at EOB in the considered energy range are 148% for ${}^{123}I$ and about 1% for each ${}^{125}I$ and ${}^{126}I$. The ${}^{123}I$ impurity needs to decay to a tolerable level till the time of application. Within about 6 days it decreases to <1%, while the level of

Table 1. Comparison of production routes of ^{124}I (all values calculated from excitation functions measured at Jülich).

Nuclear reaction	Energy range [MeV]	Thick target yield of ^{124}I [MBq/μA · h]	Impurity [%]		
			^{123}I	^{125}I	^{126}I
^{124}Te(d,2n)	14 → 10	17.5	-	1.7	-
^{124}Te(p,n)	12 → 8	16	1.0	<0.1	-
^{125}Te(p,2n)	21 → 15	81	7.4	0.9	-
^{126}Te(p,3n)	38 → 28	222	148	1.0	1.0
^{nat}Sb(α,xn)	22 → 13	1.02	890	13	16
^{121}Sb(α,n)	22 → 13	2.1	895	<0.2	<0.2
^{nat}Sb(^{3}He,xn)	35 → 13	0.95	3877	0.6	0.6

^{125}I and ^{126}I impurity will increase to 2–2.5% each. The ^{124}I yield is then reduced to about one third of its value at EOB.

Table 1 shows the optimum energy ranges, the production yields and the impurities in various routes for the production of ^{124}I. As discussed above, the ^{3}He- and α-particle induced reactions on antimony show insufficient production yields and high impurity levels.

From the four production routes based on tellurium the ^{126}Te(p,3n) reaction shows much higher yield, but also gives the most impure ^{124}I. The ^{124}Te(p,n) reaction leads to the purest product and its production can be performed at small cyclotrons. For large scale production the proton induced reaction on ^{125}Te or ^{126}Te appears to be mandatory, but their use will depend upon the tolerable levels of the impurities in ^{124}I. For therapeutic use a higher impurity level will be more tolerable than for diagnostic purposes.

Part of this work was done under a German-Egyptian bilateral agreement. We thank the concerned authorities.

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