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Direct Production of Radioiodine-123 from Tellurium with Lowest Level of Iodine Impurity-124

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ABSTRACT

Radioiodine is one of the most important radioisotopes uses in the therapeutic and diagnosis of some diseases. The radioactivity emitting from the radioactive isotopes such as Auger electrons, positrons and gamma rays play an important role in medical applications. The half-life of radioactive nuclei I-123 with 13.2h is appropriate for treatment and diagnostic. The main production routes can be obtained using ${}_{52}Te$ target. Excitations functions of (p,n) and $(p,2n)$ reactions for the production of I-123 and I-124 have been evaluated using experimental data of incident protons, energy and cross sections, published by the International Atomic Energy Agency (IAEA), especially (EXFOR) library. These data are for different authors and several years prior to the present. The best reaction for the production of Iodine-123 is ${}^{123}_{52}Te_{71}(p,n)$ reaction, while the best reaction for the production of Iodine-124 is ${}^{124}_{52}Te_{72}(p,n)$. The analysis of a complete energy range has been done for each reaction. The cross sections are reproduced in fine steps of incident proton in 0.01MeV intervals with their corresponding error. Impurity levels that appear in the production processes, which have the highest medical applications, have been estimated. The best useful energy range has been chosen by using cyclotron to reduce the appearance of nuclear impurities.

Keywords: Radioisotope, I–123, I–124, Cross Section, Iodine Production, Impurity

I. INTRODUCTION

Radionuclides have the same chemical properties. Their existence is measured in half-lives of their isotopes. Nuclear reactions provide different therapeutic radioisotopes. Radioisotopes which emit gamma rays in coincidences formed in the annihilation of a positron are used in disease treatment, especially cancer, treating aids and other diseases. Therapeutic radioisotopes are also used for medical diagnostics of many organs functioning [1].

Two imaging techniques involve the detection to give information about the organs and tissues are the Positron Emission Tomography (PET) which is a very good field for clinical diagnosis. A positron emission tomography (PET) imaging scan is a radioactive trace to detect the organs and tissues functions in 3-D pictures [2]. The second technique is the Single Photon Emission Computed Tomography (SPECT). It is subsequently processes them into 3-D images representing the distribution of nuclear activity and enables the physician to view the activity distribution in cross sections of the human body [3].

Iodine-123 isotope is typically suitable for (SPECT) imaging since it has a half-life of few hours that the radiation exposure of the patient is not too high, and now I-124 isotope is used for (PET) imaging [3,4]. One of the most promising uses of I-123 is in the imaging of monoclinical antibodies to localize and visualize tumours [5]. It is also used as nuclear imaging tracers to evaluate the anatomic and physiologic function of the thyroid [6]. It gives a much lower radiation dose to the patient, and the gamma ray energy of 159keV is ideally suited for using in a gamma camera. The gamma ray will penetrate tissue very effectively without an excessive radiation dose. For this reason, it has in many instances replaced reactor produced I-131 [5].

Iodine-124 is a proton rich isotope with a half-life of 4.18d. The long lived positron emitter I-124 is long

enough to be used for localization with monoclonal antibodies and in the field of organic chemistry and molecular imaging, the 23% positron decay allows imaging with (PET) scan [7]. I-124 as Iodide salt can be used to image the thyroid using positron emission tomography (PET) [5]. Although I-124 has been considered as an impurity in preparations of I-123 it does have good property for use in (PET) [8, 9].

II. METHODS AND MATERIAL

A. Production Yield

From the point of view of radioisotope production we only interested in the total activity of the product nuclei that can be produced during a given irradiation. The disintegration equation can be written as [10]:

$$A_t = A_{0\, e^{-\lambda t}} \quad (1)$$

Curie activities A_0 and A_t at time zero and time t, respectively. For any A we have:

$$\frac{dN}{dt} = A \times 3.7 \times 10^{10} = \lambda N \quad (2)$$

So that $A = \frac{\lambda N}{3.7\times10^{10}}$ (3)

Hence, $A_t = A_0\, e^{-\lambda t}$ follows from $N_t = N_0 e^{-\lambda t}$ (4) where λ is the decay constant. If the mass of a radioactive material consisting of a nuclide of atomic mass M, then the number of nuclei present is given by:

$$N = Avogadro's\ number \times \frac{m}{M} = 6.02 \times 10^{23} \times \frac{m}{M}\ nuclei \quad (5)$$

and the activity will be $A = \frac{\lambda}{3.7\times10^{10}}\ 6.02 \times 10^{23} \times \frac{m}{M} = 1.63 \times 10^{13} \times \frac{\lambda m}{M}\ curies$ (6)

The Yield of a nuclear reaction is defined as a ratio of the number of the nucleus formed in a nuclear reaction to the number of the bombarding particles hitting the target. For any energy, E the activity Yield in Bq of product nuclei can be expressed as the function of the cross section as [11]:

$$Yield = I(\varphi\, n)\, H\left(1 - e^{-\lambda t}\right) \int_{E_{out}}^{E_{in}} \sigma\,(E)\,(-\frac{dE}{dx})^{-1} \quad (7)$$

Where I is the current of projectile in μA, φ is the particle beam flux in s^{-1} that pass through the target nucleus material. The beam intensity is measured as $\frac{particles}{cm^2.\ sec}$ and is called flux, φ. For a nuclear reactor the flux is $\sim10^{12}$ to $10^{14} n/cm^2.sec$. Charged particle flux is commonly measured as a beam current μA, which can be converted to particles/sec. n is number of atoms per unit volume $(\frac{N}{A})$. N is Avogadro's number, A is the mass number of the target in (amu), H is the isotopic abundance (or enrichment) of the target, λ is the decay constant of the product $(\frac{0.693}{t_{1/2}}\ in\ h^{-1})$, t is time of irradiation in (h), $-\frac{dE}{dx}$ is the stopping power, $\sigma(E)$ is the cross section at energy E in (mb).

B. Impurity Level

The impurity level can be calculated at any energy as long as the relevant cross sections are known (Barrall et al. 1981) [12]. The percentage of I-A2 in I-A1 is given by:

$$I - A2 = \frac{\sigma^{**}\frac{N^*}{N'} + \sigma^*}{\frac{S}{S'}\left(\sigma\frac{N}{N'} + \sigma'\right) + \left(\sigma^{**}\frac{N^*}{N'} + \sigma^*\right)} \times 100\% \quad (8)$$

Where $\sigma, \sigma', \sigma^*, \sigma^{**}$ are cross sections for different reactions of the composite target induced by the same element with different isotopes, N, N', N^* are the abundance of the target with different isotopes. $S = (1 - e^{-\lambda_1 t})$ and $S' = (1 - e^{-\lambda_2 t})$ with λ_1 and λ_2 are the decay constant for $I - A1$ and $I - A2$ respectively and t is the time of irradiation.

III. RESULTS AND DISCUSSION

The radioisotope I-123 with a half life 13.2h is commonly used halogen nuclide for labelling bimolecular for diagnostic studies using single photon emission computed tomography (SPECT). It can be produced via various direct routes ${}^{123}_{52}Te_{71}(p,n){}^{123}_{53}I_{70}$, ${}^{124}_{52}Te_{72}(p,2n){}^{123}_{53}I_{70}$, ${}^{124}_{52}Te_{72}(p,n){}^{124}_{53}I_{71}$, and ${}^{125}_{52}Te_{73}(p,2n){}^{124}_{53}I_{71}$. Among these reactions its production via, the ${}^{124}_{52}Te_{72}(p,2n){}^{123}_{53}I_{70}$ reaction, is very important in nuclear data for its high cross sections. In order to decrease the level of isotopic impurities in the ${}^{123}_{53}I_{70}$ produced, it is essential to use highly enriched ${}^{124}_{52}Te_{72}$ (91.86%, 91.87%, 99.51%) which is given by

Acerbi et al. (1975) [13], Kondo et al. (1977) [14] and Scholten et al. (1995) [15] respectively, as a target material. However, due to the competing $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$ reaction with a highly enriched $^{124}_{52}Te_{72}$ (99.87%), it is not possible to eliminate the I-124 (with a half-life 4.18d) impurity completely, even if $^{124}_{52}Te_{72}$ is nearly to 100% enriched. As can be seen from **figure 1 and 2**, it is not possible to eliminate the I–124 impurity from the I-123, because the I–124 is being made at the same energy. In **figure 4**, the $^{123}_{52}Te_{71}(p,n)^{123}_{53}I_{70}$ reaction with energy range 4.6–17MeV has been dominated, since abundance is 0.89%, by degrading the incident energy only to 18.0MeV. All that can be done to minimize the I–124 impurity by choosing energy where the production of I–124 is near a minimum. In this case proton energy, higher than about 25MeV, will give a minimum of I–124 impurity.

The most common reaction for the production of I–123 has been concluded to be $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$ reaction on highly enriched Te–124. The high enrichment is necessary since there is a second source of I–124 contamination and this comes from $^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$ nuclear reaction on Te–125 which may be present in the target material with 7% abundance, as shown in **figure 1, 2 and 3**.

After high enrichment for $^{124}_{52}Te_{72}$, **figure 3** shows the remaining two reactions $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$ and $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$ both with abundance of target 4.6% [16]. This leads to one of the basic facts of life in radioisotope production. It is not always possible to eliminate the radionuclide impurities even with the highest isotopic enrichment and the widest energy range selection. An experiment results of the product I–123 with a minimum of I–124 is given by Lambrecht and Wolf (1973) [17]; Guillaume et al. (1975) [18]; Qaim and Stockin (1983) [19] and Clem and Lambrecht (1991) [20], with enriched Te–124(99.87%), and a proton energy range (15–20MeV). Also Scholten et al. (1989) [21] measured the optimum energy range for the production of I–123 via $^{123}_{52}Te_{71}(p,n)^{123}_{53}I_{70}$ reaction $(E_p = 14.5 \rightarrow 11MeV)$ if highly enriched $^{123}_{52}Te_{71}$ (>91.0%) is used, and the theoretical thick target yield 148.14*MBq* (4*mCi*)/*μAh*. While Scholten et al. (1995) [15] measured the excitation functions of $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$ and $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$ reactions on highly enriched Te–124 in the proton energy range (6→31*MeV*). Measurement, which shows that the thick target yield of I-124, is fairly high and amounting to 20*MBq*(0.54*mCi*)/*μAh* over the optimum energy range $(E_p = 13 \rightarrow 9MeV)$, for $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$ reaction.

Bastian et al. (2001) [22] measured the excitation functions of $^{125}_{52}Te_{73}(p,n)^{125}_{53}I_{72}$ and $^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$ with (99.8%) enriched Te–124. The yield of I-124 is four times higher than that in $^{124}_{52}Te_{72}(d,n)^{125}_{53}I_{72}$ and $^{124}_{52}Te_{72}(d,2n)^{124}_{53}I_{71}$ reactions, but the level of 0.9% I-125 impurity is relatively high.

Hohn et al. (2001) [23] measured the cross section for $^{125}_{52}Te_{73}(p,n)^{125}_{53}I_{72}$ and $^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$ reactions with (98.3%) enriched Te–125. The energy range $(E_p = 15 \rightarrow 21MeV)$ appears to be very suitable for the production of I-124. The thick target yield of I-124 amounts to 81*MBq* (*2.19mCi*)/*μAh* and the level of I-125 impurity to 0.9%. However, the calculated results with high enriched Te-124 (99.87%), and a proton energy range $(E_p = 18 \rightarrow 25MeV)$ have a good agreement in comparison with these studies under optimum conditions for each reaction.

Figure 4 shows the recommended excitation function based on the measurements data described in references [13,14,15] that the ideal proton energy range for the production of I-123 is $(E_p = 18 \rightarrow 25MeV)$, i.e., the energy of the incident protons should be selected as 25MeV and the thickness of the tellurium target should degrade the incident energy only to 18MeV.

Under these conditions **figure 5** shows the level of I-124 impurity in I-123 at the end of bombardment (EOB) amounts to about 3.1553%. Evidently, it is necessary to know the excitation functions of the various competing reactions accurately.

Table 1. The major reactions router for the production of Iodine-123

Nuclear Reaction	Natural Abundance of target % [25]	Energy Range (MeV)	Enrichment of target %	References
$^{123}_{52}Te_{71}(p,n)^{123}_{53}I_{70}$	0.89	5.9−19.6 4.6−17.8 4.6−19.6	85.4 95.6	Scholten et al. (1989)[21] Mahunka et al. (1996) [26] Recommended value (PW)
$^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$	4.6	12.33−29.27 12.45−28.19 12.2−29.18 11.9−31.2 11.9−31.2	91.86 99.87 91.87 99.51	Acerbi et al. (1975a)[13] Kondo et al. (1977a)[14] Kondo et al. (1977b) [14] Scholten et al. (1995a) [15] Recommended value (PW)
$^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$	4.6	12.41−29.29 12.45−28.19 12.2−29.18 5.8−31.2 5.8−31.2	91.86 99.87 99.87 99.51	Acerbi et al. (1975b) [13] Kondo et al. (1977c) [14] Kondo et al. (1977d) [14] Scholten et al. (1995b)[15] Recommended value (PW)
$^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$	7.0	10.6−100.5 10.6−100.5	98.3	Hohn et al. (2001g) [23] Recommended value (PW)

Figure 6 shows the range of incident protons penetrating the Tellurium target using SRIM 2003 [24], compared with the calculated adopted results and proved to be in agreement with no relative difference. It is clear from **figure 6** that 1.55mm layer (both calculated by SRIM and in the present work) of Te-124 is required to reduce the proton beam energy from 25 to 18MeV.

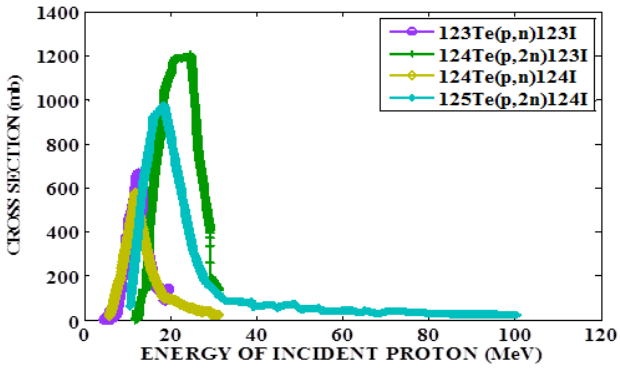

Figure 1: Recommended cross sections of the $^{123}_{52}Te_{71}(p,n)^{123}_{53}I_{70}$, $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$, $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$, and $^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$ reactions versus the incident proton energy up to 100MeV.

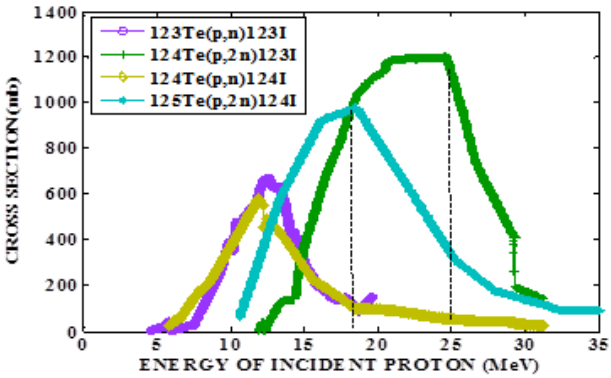

Figure 2: Recommended cross sections of the $^{123}_{52}Te_{71}(p,n)^{123}_{53}I_{70}$, $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$, $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$, and $^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$ reactions versus the incident proton energy up to 35MeV.

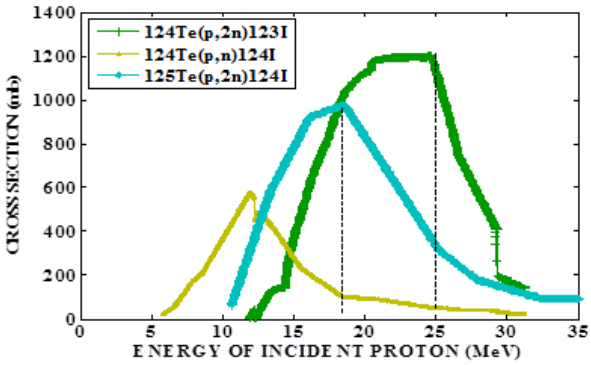

Figure 3: The recommended cross sections of the $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$, $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$ and $^{125}_{52}Te_{73}(p,2n)^{124}_{53}I_{71}$ reaction versus the incident proton energy.

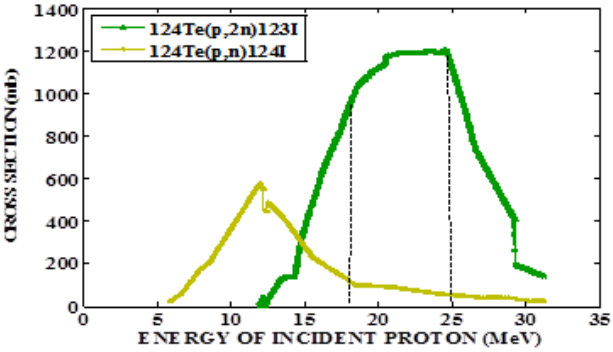

Figure 4: The recommended cross sections of the $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$ and $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$ reaction versus the incident proton energy.

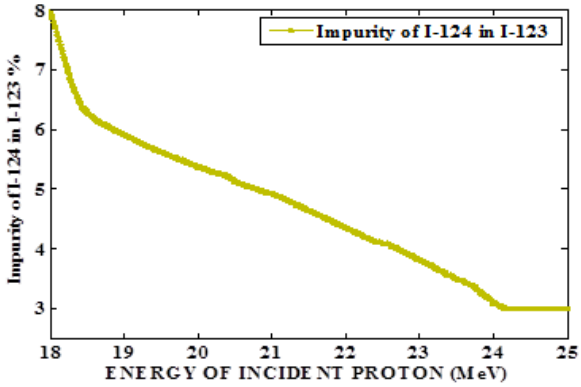

Figure 5: Impurity percentage of the I-124 in I-123 from the reaction $^{124}_{52}Te_{72}(p,n)^{124}_{53}I_{71}$, and $^{124}_{52}Te_{72}(p,2n)^{123}_{53}I_{70}$.

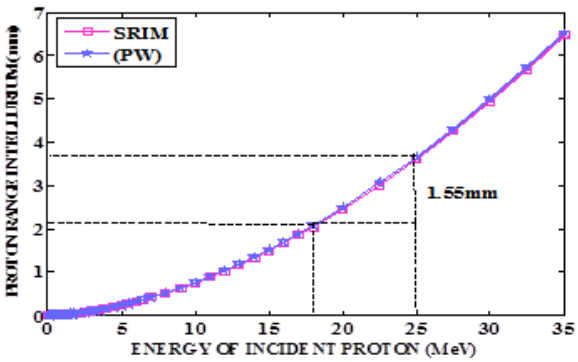

Figure 6: Incident proton energy range penetrating the Tellurium target

IV. CONCLUSION

Higher enrichment of the targets ${}_{52}Te$ has been used, for the production of Iodine isotopes, to reduce the appearance of impurities. The characteristic of the diagnosis and treatment radioisotopes is to ensure the access of radiation to the organ for diagnostic or treated without moving to the nearby tissues by selecting a useful energy. We found the best reactions for the production of I-123 are ${}^{123}_{52}Te_{71}(p,n)$ reaction without any impurity and useful energy range 5→15MeV. The calculated yield is 49.06*MBq(1.33mCi)/μAh* has a discrepancy with the value obtained by Milton K. et al. (1977) [27] of *266.2MBq(7.2mCi)/μAh*, for the same useful energy range. For higher useful energy range 18→25MeV, the ${}^{124}_{52}Te_{72}(p,2n)$ reaction is used with calculated impurity 21.8% of Iodine-124 and yield 47.1*MBq(1.27mCi)/μAh*, which can be compared with the results obtained by (IAEA) (2009) [28] for the same useful energy range and Uddin U. S. et al. (2011) [29] of impurity 3.21%, of Iodine-124 and yield 19.98*MBq(0.54mCi)/μAh*.

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