Evaluation the reactions of production the radioactive Iodine-124

Evaluation the reactions of production the radioactive Iodine-124

Aeshah Ali Hussein[1], Naz T Jarallah[2], Bashair Mohamed Saied[3]

[1,2,3]Department of Physics, College of Education for Pure Sciences Ibn-AL Haitham, University of Baghdad, Baghdad, Iraq

Email: aisha.sataa.1983@gmail.com

Abstract. Nowadays, the field of radionuclide treatment is enjoying an exciting stage and preparing for further growth and progress in the future. For instance, in Asia, the large spread of liver and thyroid diseases has resulted in several new developments /clinical trials using molecular radiotherapy (i.e. targeted radionuclide therapy). Iodine-124 has unique physical properties including long half-life that adding an advantage for pharmacokinetics and radiopharmaceutical analysis. One of its applications in nuclear medicine is in Positron Emission Tomography (PET).The aim of the present work is to evaluate the production yield of ^{124}I via calculated the excitation functions /stopping power for the reactions^{nat}Te(P,X)^{124}I, ^{121}Sb(A,N)^{124}I,^{nat}Sb(A,X)^{124}I and ^{123}Sb(A,3N)^{124}I in the energy range (3.87-62.95)MeV by using Mat-lab and SRIM programs .

Keyword: Cross section, Excitation function, Iodine-124, Integral yield.

1. Introduction

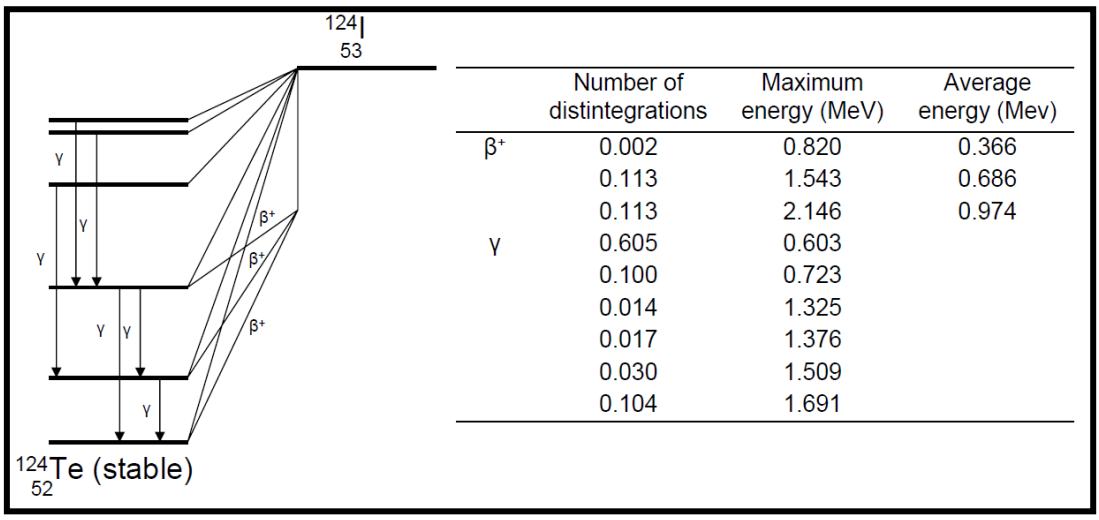
Generally, Radioisotopes are formed in reactors /cyclotrons to be utilized for diagnostic or treatment purposes in healthcare. The reaction is produced via the energy of the projectile particles to be utilized in the production of the target core and the radioisotopes. For situations where perform an experiment is expensive and hard, simulation studies are preferred to carry out owing to it is save both economy and time [1]. In nuclear medicine, iodine radionuclides are exceedingly utilized for labelling monoclonal antibodies, receptors, and other radiopharmaceuticals, specifically in the applications of diagnostic and therapeutic as quantitative imaging over an interval of a number of days is needful [2]. Unluckily, the nuclides most utilized are ^{131}I, ^{125}I and ^{123}I, and all have restrictions.^{131}I is considered the most commonly used among the three, and it has very high photon energy for optimal imaging. Moreover, rigorous attenuation correction does not allowed via Single-Photon Emission Computed Tomography imaging (SPECT), though a satisfactory empirical correction may sometimes be realized. ^{125}I has a photon energy which is very low for optimal imaging, particularly quantitative imaging, also it has an extremely long half-life (i.e. half-life is undesirably long). ^{123}I has a half-life which is very short. On the other hand, a positron-emitting nuclide is Iodine-124 with a half-life of 4.176 days, and this allows quantitative imaging over several days by Positron Emission Tomography PET (a powerful non-invasive technique for molecular imaging [2–4].

Among its clinical applications are Na, ^{124}I has been used to diagnose/dosimetry in the disease of thyroid and to estimate the springing up of metastatic thyroid carcinoma. Furthermore, [^{124}I]-iododeoxyuridine ([^{124}I]-IUdR) has been used in measuring the activity of proliferation of tumors in

patients suffer from tumors of brain, including gliomas and meningiomas. Besides,[^{124}I]-Miodobenzylguanidine ([^{124}I]-MIBG) has got massive potential for utilizing in cardiovascular imaging, diagnosis, and therapy of malignant diseases like carcinoids, neuroblastoma, pheochromocytoma and paraganglioma [3,5–8]. R. Lambrecht et al in (2010) produced Iodine-124 in great yield through >99.5% radionuclidic purity at (48 h) post irradiation and the ^{124}Te (d,2n)^{124}I reaction. Targetry methodology has been explained. The production yield was (0.55±0.06 mCi/μA.h) [9]. R. Weinreich and E. Knust in (1996) was produced ^{124}I (T = 4.15 d) using a compact cyclotron by the nuclear reaction ^{124}Te (d, 2n)^{124}I via irradiation of $^{124}TeO_2$ with deuterons of energies (14 MeV). And then, dry distillation of the radioactive iodine isotopes generated from irradiated target materials. By-products including (8.02d)^{131}I, (12.4h)^{130}I, (13.0d)^{126}I, (60d)^{125}I and (13.2d)^{123}I have been measured and collected in each charge. After (45 h) decay time, the data demonstration that the total of the activities of these nuclides is below 5% of the ^{124}I activity [10]. S. S. Salodkinand V. M. Golovkov in (2020) have been investigated the methods of production of Iodine-124 radionuclide in nuclear medicine. The effect of target material, the selected nuclear reactions, the irradiation conditions, and the ways of manufacturing / cooling of the irradiated targets on the Iodine-124 yield is considered [11]. In present work, the yield of ^{124}I has been calculated via weighted average cross sections and stopping power of ^{nat}Te(P,X)^{124}I , ^{121}Sb(A,N)^{124}I ,^{nat}Sb(A,X)^{124}I and ^{123}Sb(A,3N)^{124}I reactions by using sets of programs like Mat-lab and SRIM.

2. Decay Scheme of Iodine-124

Iodine-l24 has decay scheme as shown in Fig. (1). At minimum, there are 97 gamma-ray transitions, 25 electron capture transitions and 6 positron transitions. Within the limits, 23% of decays result in emission of positron. Furthermore, there is several high energy gamma rays, some in coincides with the emissions of the positron [2-4].



	Number of distintegrations	Maximum energy (MeV)	Average energy (Mev)
β+	0.002	0.820	0.366
	0.113	1.543	0.686
	0.113	2.146	0.974
γ	0.605	0.603	
	0.100	0.723	
	0.014	1.325	
	0.017	1.376	
	0.030	1.509	
	0.104	1.691	

Figure 1. Simplified decay scheme of ^{124}I

3. Theoretical background

For several decades, the possible use of radionuclides in treatment has been recognized. The most important advantage of radionuclide therapy is the ability to deliver a greatly concentrated absorbed dose to the tumor with preserving the surrounding natural tissue. Targeted radionuclide treatment has become one of the most favourite kinds of cancer treatment due to the administration of radionuclides is slightly invasive and the period of therapy is shorter than chemotherapy. A number of radionuclides including Iodine-124 have been successfully utilized for diagnostic and therapeutic [1,12]. There is a large number of reactions that is utilized for producing ^{124}I, relying on the particles, cyclotron and the

energies that are available in executing the irradiations [5]. In present work, ^{124}I has been produced by utilizing proton induced ^{nat}Te(P,X)^{124}I reaction and α induced ^{121}Sb(A,N)^{124}I ,^{nat}Sb(A,X)^{124}I , ^{123}Sb(A,3N)^{124}I reactions. The energy data for every reaction are obtained from the database of EXFOR and using Matlab programs language to calculate weighted average cross sections (W) for each reaction individually as the following:

$$W=\frac{(\sum wi\ yi)}{(\sum wi)} \quad (1)$$

$$w_i = {}^{1}/_{\mu_i^2} \quad (2)$$

where $\mu_i$is the standard deviation for sample i and yi is the cross section value for sample i [13].The cross section is one of the most significant physical quantity describing nuclear reactions. Experimentally, it can be calculated or measured utilizing varied technique (i.e. models) of nuclear reactions [14].

Furthermore, the integral yield (i.e. the overall number of the formed nuclei through an irradiation time) of these reactions is calculated according to the equation below [15]:

$$Y(t) = t \int_0^L I(x)\,\sigma(x)\,({}^{\rho}/_{ze})dx \quad (3)$$

$$= tI_0 \int_{E_L}^{E_0} (-\frac{1}{\rho}\frac{dE}{dX})^{-1}\left(\sigma(E)/_{ze}\right)dE \quad (4)$$

$$= t\,I_0 y \quad (5)$$

Here, ρ is the target isotope number density of sample material, t is the irradiation time, L is the thickness of sample material, $\sigma(x)$ is the cross section to produce the isotope at depth x in the sample, Z is the charge number, I_0 is the number of beam particles irradiating the sample per unit irradiation time, E_0 initial beam energy, E_L is the energy of the beam at the exit of the sample, $(-\frac{1}{\rho}\frac{dE}{dX})^{-1}$ is the stopping power and y is the thick target product yield (i.e. is the number of the formed nuclei per unit induced electric charge). It is used for both stable / radioactive products, however is frequently utilized for stable ones.

On the other hand, the number of the formed nuclei existing in the sample material (t) satisfies when the product is radioactive with decay constant(λ)according to the relation below:

$$\frac{dN(t)}{dt} = \frac{dY(t)}{dt} - \lambda\ N(t) = I_0\ y\ -\ \lambda\ N(t) \quad (6)$$

The solution is

$$N(t) = I_0\ y\ \frac{1-e^{-\lambda t}}{\lambda} \quad (7)$$

Then

$$\frac{\lambda N(t)}{I_0} = y\left(1-e^{-\lambda t}\right) = a(t) \quad (8)$$

where $\frac{\lambda N(t)}{I_0}$ represented the activity of the sample material per unit current at t and $a(t)$ is the decay rates of the product per unit current [15].

4. Result and discussion

The values of the cross section for ^{nat}Te(P,X)^{124}I , ^{121}Sb(A,N)^{124}I , ^{nat}Sb(A,X)^{124}I and ^{123}Sb(A,3N)^{124}I reactions are calculated from previous studies [16-29], in the range of energies (3.87-62.95MeV) with a step of (0.5 MeV).

The weighted average cross sections for the above mentioned reactions have been calculated using equations (1 and 2) and rely on the data of proton and α induced, energy and cross sections published via the (EXFOR) library affiliated to the International Atomic Energy Agency by using Mat-lab(8.3a 2014) program .

Moreover, the integral yield ^{nat}Te(P,X)^{124}I , ^{121}Sb(A,N)^{124}I , ^{nat}Sb(A,X)^{124}I and ^{123}Sb(A,3N)^{124}I reactions have been evaluated by using equations (3-8) depended on the values of stopping power that was calculated via Zeigler formula through (SRIM-2013) program. The results of the integral yields are given in table (1).

Table1. Thick target yield of reactions

$Nuclear\ reaction$	$Energy\ (MeV)$	$Thick\ target\ yield\ of\ ^{124}I(\frac{Bq}{\mu A}.h)$
$^{nat}Te(P,X)^{124}I$	$10-20$	$1400-1447$
$^{121}Sb(A,N)^{124}I$	$30-45$	$184.6-200$
$^{nat}Sb(A,X)^{124}I$	$26.72-47$	$446-575$
$^{123}Sb(A,3N)^{124}I$	$32-45$	$1942-2273$

The calculated results of excitation functions andintegral yields are presented in Figures. 2 – 9.

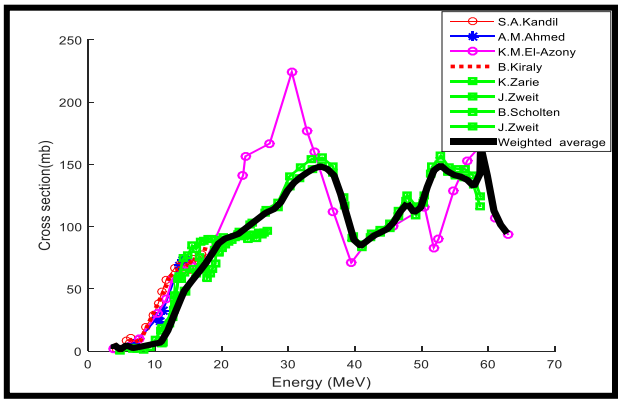


Figure 2. Excitation function of (_^nat)Te(P,X) (_^124)I

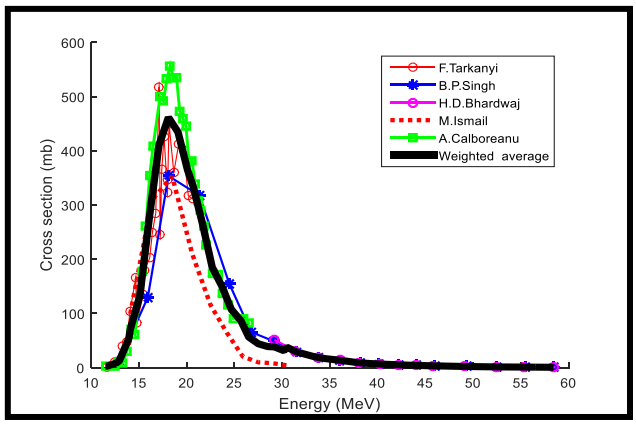


Figure 3. Excitation function of (_^121)Sb(A,N) (_^124)I

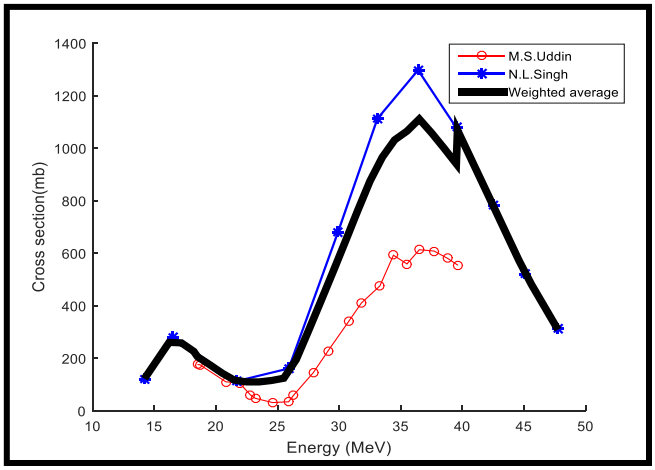


Figure 4. Excitation function of (_^nat)Sb(A,X) (_^124)I

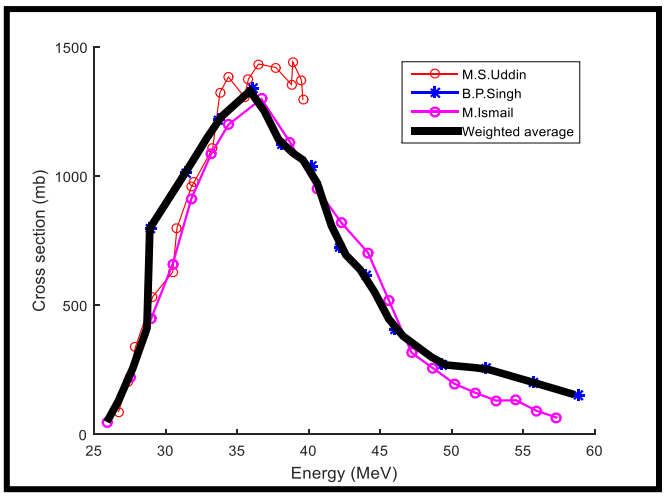


Figure 5. Excitation function of $^{123}Sb(A,3N)^{124}I$

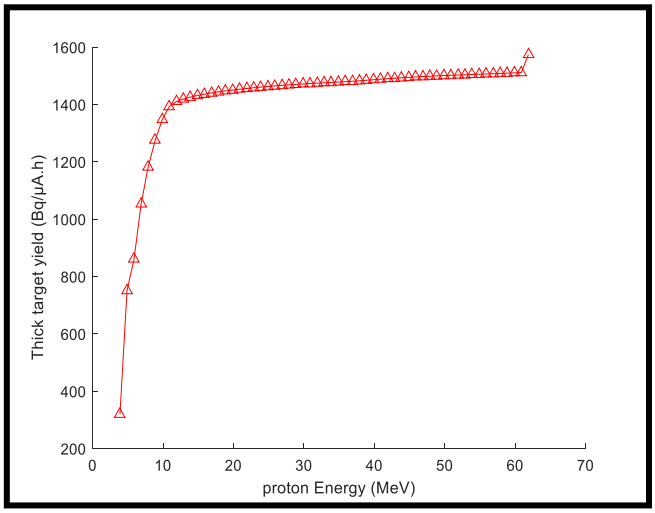


Figure 6. Production Yield for $^{nat}Te(P,X)^{124}I$ reaction

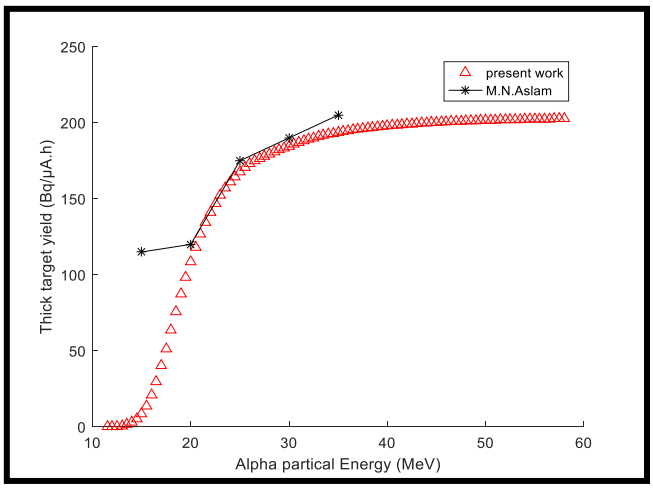


Figure 7. Production Yield for $^{121}Sb(A,N)^{124}I$ reaction.

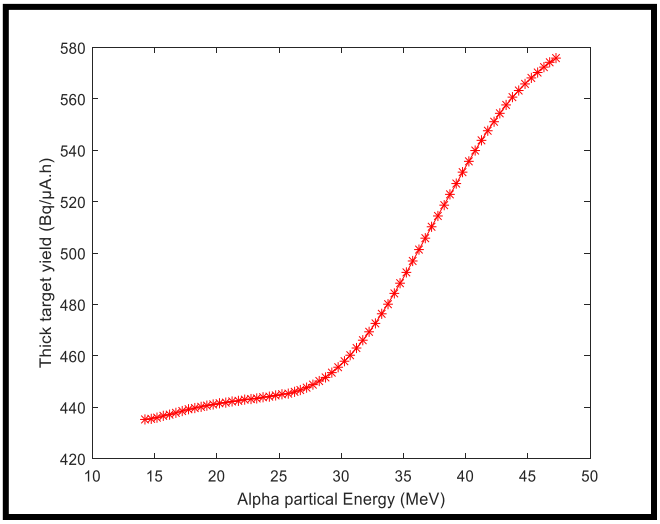


Figure 8. Production Yield for $^{nat}Sb(A,X)^{124}I$*reaction*

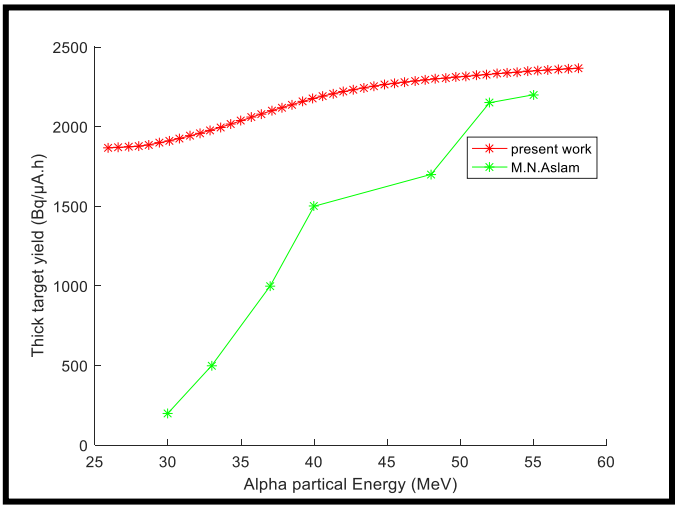


Figure 9. Production Yield for $^{123}Sb(A,3N)^{124}I$reaction.

We observed from figures (2-5) that the cross section of the ^{nat}Te(P,X)^{124}I ,^{121}Sb(A,N)^{124}I ,^{nat}Sb(A,X)^{124}I and ^{123}Sb(A,3N)^{124}I reactions is proportional to the energy of the incident particle.
$^{nat}Te(P,X)^{124}I$ *reaction*: It has two maximum cross-section values which are$(148.3824mb,164.9666mb)$ at energies $(35\ MeV, 59MeV)$ respectively.

$^{121}Sb(A,N)^{124}I$ *reaction*: it has one maximum cross-section value which is$(461.6968mb)$ at about18.1 MeV. After that, the cross-section values start decreasing until reaching to $(0.7882mb)$ at energy (58.42 MeV).

$^{nat}Sb(A,X)^{124}I$ *reaction* : It has two maximum cross-section values which are $(1.1107 \times 10^3 mb,$ $1.0681 \times 10^3 mb)$ at energies $(36.5\ MeV, 39MeV)$ respectively.

$^{123}Sb(A,3N)^{124}I$ *reaction*: It has one maximum cross-section value which is$(1.33 \times 10^3 mb)$ at energy35.5 MeV. After that, the cross-section values start decreasing until reaching to $(0.1579 \times 10^3 mb)$ at energy of58 MeV.
On the other hand, we noticed that after each maximum cross-section value, the cross-section values begin to decrease as the energy of the incident particle increases.
Furthermore, the integral yield of ^{nat}Te(P,X)^{124}I ,^{121}Sb(A,N)^{124}I ,^{nat}Sb(A,X)^{124}I and ^{123}Sb(A,3N)^{124}I reactions are directly proportional to the energy of the incident particle as shown in figures (6-9).
It is obvious that the values of the yield are different from one reaction to another owing to the difference in reaction cross section and energy of incident projectile. Choosing a suitable energy range for the incident particle improves the production process (i.e. Increases iodine yield and reduces impurities. The yields of reactions ^{121}Sb(A,N)^{124}I and ^{123}Sb(A,3N)^{124}I were in agreement with [30].
Generally, iodine-124 is formed good yield through the nuclear reaction $^{123}Sb(A,3N)^{124}I$.

5. Conclusions

The main objective of this work is the possibility of producing radioactive Iodine-124 to be used in nuclear medicine (i.e. diagnostic and therapeutic applications including Positron Emission Tomography (PET)), as it has physical and chemical properties that qualify it for that.
So, it was produced theoretically from different reactions by utilizing sets of programs like SRIM and Mat-lab. From the results that shown in table (1), we can conclude that the reactions

$^{123}Sb(A,3N)^{124}I$ are the best of other reactions to the production of iodine with the least possible energy (i.e. $^{123}Sb(A,3N)^{124}I$ reaction is ideally suited to produce Iodine-124.

6. References

[1] Rıdvan Ü and Akçaalan U 2020 The Reaction Cross Sections for 124,125 Te (p, xn) 123,124 I and 123, 124 Te (d, xn) 123, 124 I *Avrupa Bilim ve Teknol. Derg*. 958–963.
[2] Pentlow KS, Graham MC, Lambrecht RM, Daghighian F, Bacharach SL, Bendriem B, Finn RD, Jordan K, Kalaigian H, Karp JS, Robeson WR and Larson SM 1996 Quantitative imaging of iodine-124 with PET *J. Nucl. Med*. **37** 1557–1562.
[3] Lewis JS 2020 *Production , Use and Applications of 124 I*.
[4] Cascini GL, Niccoli Asabella A, Notaristefano A, Restuccia A, Ferrari C, Rubini D, Altini C and Rubini G 2014 124Iodine: A longer-life positron emitter isotope - New opportunities in molecular imaging *Biomed Res. Int.* **2014** https://doi.org/10.1155/2014/672094.
[5] Braghirolli AMS, Waissmann W, Da Silva JB and Dos Santos GR 2014 Production of iodine-124 and its applications in nuclear medicine *Appl. Radiat. Isot.* **90** 138–148. https://doi.org/10.1016/j.apradiso.2014.03.026.
[6] Guenther I, Wyer L, Knust EJ, Finn RD, Koziorowski J and Weinreich R 1998 Radiosynthesis and quality assurance of 5-[124I] iodo-2′-deoxyuridine for functional PET imaging of cell proliferation *Nucl. Med. Biol.* **25** 359–365.
[7] Ficola U, Quartuccio N, Paratore R, Treglia G, Piccardo A and Cistaro A 2013 *Utility of 124I-MIBG PET/CT in the follow-up of patients with advanced neuroblastoma*: first report of the AIMN PET-Pediatric Study InterGroup, in: Eur. J. Nucl. Med. Mol. Imaging, SPRINGER 233 SPRING ST, NEW YORK, NY 10013 USA S204–S205.
[8] Hartung-Knemeyer V, Rosenbaum-Krumme S, Buchbender C, Pöppel T, Brandau W, Jentzen W, Antoch G, Forsting M, Bockisch A and Kühl H 2012 Malignant pheochromocytoma imaging with [124I] mIBG PET/MR. *J. Clin. Endocrinol. Metab*. **97** 3833–3834.
[9] Lambrecht R, Sajjad M, Qureshi M and Al-Yanbawi S 1998 Production of iodine-124 *J. Radioanal. Nucl. Chem.* **127** 143–150.
[10] Weinreich R and Knus E 1996 Quality assurance of iodine-124 produced via the nuclear reaction124Te (d, 2n) 124I *J. Radioanal. Nucl. Chem*. **213** 253–261.
[11] Salodkin SS and Golovkov VM 2014 Cyclotron Production of Iodine-124, *Russ. Phys. J.* 1–7.
[12] Yeong CH, Cheng MH and Ng KH 2014 Therapeutic radionuclides in nuclear medicine: Current and future prospects, *J. Zhejiang Univ. Sci. B*. **15** 845–863. https://doi.org/10.1631/jzus.B1400131.
[13] Mohammed AL 2015 Production Yields of Ytterbium -169 Medically Relevant Radionuclide Within Medium Energy Range for the Protons, Deuterons and Alpha Particles *Int. J. Appl. Nat. Sci.* **4** 13–20. http://www.iaset.us/view_archives.php?year=2015&id=73&jtype=2&page=4.
[14] Nechifor C, Straticiuc M and Bercea M 2007 *Cross Sections and Protons Optimum Energy Ranges for Some Medical,* Cross Sect.
[15] Otuka N and Takács S 2015 Definitions of radioisotope thick target yields *Radiochim. Acta*. **103** 1–6. https://doi.org/10.1515/ract-2013-2234.
[16] Kandil SA and Al-Abyad M 2013 Cross section measurements and theoretical calculations of proton induced nuclear reactions on natural tellurium *Radiochim. Acta*. **101** 67–72.
[17] Ahmed AM, Hassan HE, Hassan KF, Khalaf AM and Saleh ZA 2011 Cross sections for the formation of radioiodines in proton bombardment of natural tellerium with particular reference to the validation of data for the production of 123I, *Radiochim. Acta Int. J. Chem. Asp. Nucl. Sci. Technol.* **99** 317–323.
[18] El-Azony K, Suzuki K, Fukumura T, Szelecsényi F and Kovács Z 2008 Proton induced reactions on natural tellurium up to 63 MeV: data validation and investigation of possibility of 124I production *Radiochim. Acta.* **96** 763–769.

[19] Király B, Tárkányi F, Takács S and Kovács Z 2006 Excitation functions of proton induced nuclear reactions on natural tellurium up to 18 MeV for validation of isotopic cross sections *J. Radioanal. Nucl. Chem.* **270** 369–378.
[20] Zarie K, AI-Hammad N and Azzam A 2006 Excitation functions of (p, xn) reactions on natural tellurium at low energy cyclotron: relevance to the production of medical radioisotope 123 I, *J. Nucl. Radiat. Phys.* **1** 93–105.
[21] Zweit J, Bakir MA, Ott RJ, Sharma HL, Cox M and Goodall R 1992 *Excitation functions of proton induced reactions in natural tellurium: production of no-carrier added iodine-124 for PET applications*.
[22] Scholten B, Qaim SM and Stöcklin G 1989 Excitation functions of proton induced nuclear reactions on natural tellurium and enriched 123Te: production of 123I via the 123Te (p, n) 123I-process at a low-energy cyclotron, Int. J. Radiat. Appl. Instrumentation. Part A. *Appl. Radiat. Isot.* **40** 127–132.
[23] Tárkányi F, Takács S, Király B, Szelecsényi F, Andó L, Bergman J, Heselius SJ, Solin O, Hermanne A and Shubin YN 2009 Excitation functions of 3He-and α-particle induced nuclear reactions on natSb for production of medically relevant 123I and 124I radioisotopes *Appl. Radiat. Isot.* **67** 1001–1006.
[24] Singh BP, Sharma MK, Musthafa MM, Bhardwaj HD and Prasad R 2006 A study of pre-equilibrium emission in some proton-and alpha-induced reactions *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip*. **562**, 717–720.
[25] Singh BP, Bhardwaj HD and Prasad R 1991 A study of pre-equilibrium emission in α-induced reactions on 121,123 Sb *Can. J. Phys.* **69** 1376–1382.
[26] Ismail M 1990 Measurement and analysis of the excitation function for alpha-induced reactions on Ga and Sb isotopes *Phys. Rev. C.* **41** 87.
[27] Calboreanu A, Pencea C and Salagean O 1982 The effect of gamma de-excitation competition on the (α, n) and (α, 2n) reactions on gold and antimony *Nucl. Physics, Sect. A.* **383** 251–263 https://doi.org/10.1016/0375-9474(82)90451-1.
[28] Uddin MS, Hermanne A and Sudar S 2011 Excitation functions of alpha-particle induced reactions on enriched Sb-123 and Sb –nat for production of I-124 *J. Appl. Radiat. Isot*. **69** 699–704.
[29] Singh NL, Shah DJ, Mukherjee S and Chintalapudi SN 1997 Excitation functions for alpha-particle-induced reactions with natural antimony *Nuovo Cim. Della Soc. Ital. Di Fis. A*. **110** 693–709.
[30] Aslam MN, Sudár S, Hussain M,. Malik AA and Qaim SM 2011 Evaluation of excitation functions of 3He-and α-particle induced reactions on antimony isotopes with special relevance to the production of iodine-124 *Appl. Radiat. Isot.* **69** 94–104.