

A MONTE CARLO APPROACH TO CALCULATE THE PRODUCTION PREREQUISITES OF ^{124}I RADIOISOTOPE TOWARDS THE ACTIVITY ESTIMATION

by

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The Monte Carlo simulation code MCNPX has been used to simulate the production of ^{124}I by $^{124,125}\text{Te}(p, xn)$ and $^{123,124}\text{Te}(d, xn)$ reactions to form high activity ^{124}I . For this reason, the TALYS-1.8 and ALICE/ASH codes were used to calculate the reaction cross-section. The optimal energy range of projectile is selected for this production by identifying the maximum cross-section and the minimum impurity due to other emission channels. Target geometry is designed by SRIM code based on stopping power calculations with identical dimensions as the experimental data. The thick target yield of reactions is predicted because of the excitation functions and stopping power. All of the prerequisites obtained from the above interfaces are adjusted in MCNPX code and the production process is simulated according to benchmark experiments. Thereafter, the energy distribution of proton in targets, the amount of residual nuclei during irradiation, were calculated. The results are in good agreement with the reported data, thus confirming the usefulness and accuracy of MCNPX as a tool for the optimization of other radionuclides production. Based on the results, the $^{124}\text{Te}(p, n)^{124}\text{I}$ process seems to be the most likely candidate to produce the ^{124}I in low-energy cyclotrons.

Key words: ^{124}I , production yield, cyclotron, MCNPX, TALYS-1.8, ALICE/ASH

INTRODUCTION

Numerous experiments are being performed every year on the cross-section of reaction and its yield, in the field of nuclear physics. Nevertheless, the results of these experiments were compared with some similar studies; it is also of high importance to take into account the prediction ability of introduced models. But the fact to be considered is narrowing the models on a specific physical phenomenon, *e. g.* into a nuclear reaction, to achieve a specific simulator package capable of reaching the final response by receiving the partial models.

Both modeling and simulation have enabled the scholars to attain optimal results, so that they can be utilized as a roadmap, especially for high-cost experiments, as a result of their fast and reasonable solutions, and they can also be used as the benchmark data. In other words, the modeling provides more information which leads to a better understanding of the physical phenomena and its results can act as a guideline in the

setup and the design of the physical experiments. Thus, theoretical models used in nuclear processes have a great role in all the steps of nuclear data evaluation for both understanding of the physical phenomena and to estimations of the required cross-sections where data are not fully available. On the other hand, Monte Carlo (MC) approaches can provide a realistic view of the physical phenomena and simulate a the same reality situation in evaluation of a nuclear reaction. Moreover, it seems that a wide international cooperation between theoretical and experimental teams is important for fostering the collection of more detailed and accurate data on nuclear reactions relevant to radioisotope production and improving their theoretical description [1].

To delve more into the problem, applying the modeled approach, from the production prerequisites to the yield estimation, this work tends to monitor the production of ^{124}I ($T_{1/2} = 4.17$ d, $E_{\beta^+} = 2.13$ MeV, $I_{\beta^+} = 22\%$) and its radioisotopic impurities by charged particle irradiation. It is worth mentioning that ^{124}I is a suitable long-lived radionuclide for both therapeutic and diagnostic application in nuclear medicine. The relatively long half-life of ^{124}I (4.17 d) is appropriate

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for long-time *in vivo* studies of heavy molecular compounds absorption, such as positron emission tomography (PET) imaging [2].

The ^{124}I can be produced at cyclotron utilizing enriched tellurium-124 through deuteron- and proton-induced reactions, with respect to the type of available cyclotron, particle, and energies. At first, ^{124}I was mainly produced by $^{124}\text{Te}(d, 2n)^{124}\text{I}$ reaction [3-6]. Nevertheless, in the recent years, by increasing the number of low-energy cyclotrons, $^{124}\text{Te}(p, n)^{124}\text{I}$ reaction has attained popularity in practice [7-10]. In 2003, Qaim *et al.* [10] showed that high purity ^{124}I can be produced on a medium scale employing low-energy (p, n) reactions on high enriched ^{124}Te , at a small-sized cyclotron. The evaluation of possible processes for the production of ^{124}I from enriched Te and antimony targets were presented by Aslam *et al.* [11, 12] according to the TALYS, STAPRE and EMPIRE codes and variation of optical parameters. However, Braghirolli *et al.* [13] have summarized recent advances in ^{124}I radionuclide production and its medical applications, with mainly discussion in targetry, target processing, and PET imaging application. Recently, the excitation functions of all the possible reactions based on different nuclear codes for the production of ^{124}I and also the ^{123}I , ^{125}I , and ^{126}I radionuclides were evaluated, which may be co-produced with ^{124}I as radionuclidic impurities [14]. Therefore, this literature will certainly provide results with a meaningful comparison.

The modeling and simulation have more significance in high-cost experiments such as cyclotron production of radioisotopes, to explore the best condition of the production because of their fast, reasonable and low-cost solutions. Therefore, in the ^{124}I production procedure, because of its relatively high cost of production and the need of applying benchmark data, utilization of a Monte Carlo (MC) code for optimization of production process, *i. e.* using an appropriate reaction and optimum range of projectile energy for production of ^{124}I nuclide, assumes a very important role in assessing the yield of ^{124}I , as well as contaminants inherent in the production of ^{124}I nuclide. On the other hand, since the accuracy of the calculations depends on the correctness of the computational models, implemented in the used MC code, the comparison of the results of different codes, with different implementations of physics and tracking, as well as experimental data, have a special importance for selection of an appropriate code with the least discrepancies with the experimental data.

In this study, Monte Carlo code, MCNPX 2.6 [15], was utilized to calculate the production prerequisites of ^{124}I and its co-produced impurities (^{123}I and ^{125}I) through $^{124}\text{Te}(p, n)$, $^{125}\text{Te}(p, 2n)$, $^{123}\text{Te}(d, n)$, and $^{124}\text{Te}(d, 2n)$, reactions. In each process, the calculated cross sections in our previous work [14] were utilized to estimate the desired radionuclide production yield. Stopping power and range of particles in target matter

were obtained utilizing the SRIM code [16]. The target system was modeled with identical dimensions as the experimental data [17, 18]. In each reaction, the available experimental values were used as the benchmark data. At the end of the study, the appropriate reaction and optimum energy range of projectile for the production of ^{124}I , as well as the usefulness of MCNPX code for optimizing the radionuclide production, were considered.

MATERIALS AND METHODS

MCNPX 2.6 code

MCNPX is a general-purpose Monte Carlo radiation transport code developed by the Los Alamos National Laboratory (LANL). MCNPX was planned to follow various particle types over a wide energy range and to analyze the transport of protons, neutrons, electrons, gamma rays and other particles. MCNPX 2.6 has many new abilities, particularly in the areas of transmutation, burn-up and delayed particle production. Moreover, many new tally sources and variance-reduction options have been expanded [15].

The MCNPX input file contains information about a desired problem; such as geometry characteristics, definition of materials, description and location of sources, type of tallies, and variance-reduction methods utilized to enhance the performance of a program [19].

TALYS-1.8 and ALICE/ASH codes

TALYS-1.8 and ALICE/ASH are nuclear codes used to predict and analyze nuclear reactions or produce nuclear data.

TALYS is utilized to simulate nuclear reactions involving protons, neutrons, deuterons, photons, tritons, ^3He - and α -particles, in the 1 keV to 200 MeV energy range and covers the target nuclides of mass 12 and heavier. By default, TALYS utilizes phenomenological optical model parameterizations for neutrons and protons on a nucleus-by-nucleus basis to obtain the transmission coefficients and reaction cross-sections. If such a potential is not available, TALYS automatically utilizes a global optical mode. For deuterons and other complex particles, the nucleon potentials, either local or global, are taken as the basis of these complex particle potentials [20, 21].

ALICE/ASH code is an improved version of the ALICE code. This code was written to investigate the interaction of intermediate energy nucleons with target nuclei. This code calculates energy and angular distributions of particles emitted in nuclear reactions, residual nuclear yields and total non-elastic cross-sections for nuclear reactions induced by particles with

energy up to 300 MeV. Models utilized in this code explain the pre-compound composite particle emission and fast γ -emission and contain different procedures for nuclear level density and fission fragment yield calculations. The geometry dependent hybrid model (GDH) is utilized to describe pre-equilibrium particle emission from nuclei [22].

Target geometry and beam configuration

In the fabrication of target material for cyclotron bombardment, the tellurium target and its 3 mm backing plate along with the 10 μ m thick aluminum-foil cover were represented by elliptical and rectangular macro-bodies. The incident beam passes through a 28 mm in diameter, 50 μ m thick titanium foil, that separates the vacuum region from the target assembly (fig. 1). Moreover, a set of vacuum beam line and a graphite collimator have been located at the 6° beam-target angle. Figure 1 shows the modeled target system, as designed for input file of the MCNPX. The configuration was specified by the source definition (SDEF) card. The SDEF card has many parameters that are used to define approximately all the characteristics of all the sources. The projectile beam was designed to reach to the target with 6° target-beam angle. The target thickness is decreased and optimized by 90 % as a result of the angular shifting. The monoisotopic tellurium targets were irradiated by 15.2 MeV and 21.9 MeV protons and 13.3 MeV and 16.9 MeV deuterons. These irradiation energies were calculated in such a way that according to the SRIM results, the incident particles, after losing a part of their energy when passing through the 50 μ m titanium and 10 μ m inclined aluminum foils, enter the tellurium target layer by desired effective energies of 14 and 21 MeV for protons and 11 and 15 MeV for deuterons, respectively. So, the effect of the Ti vacuum and Al cover foils was taken into account in the calculations as a reducing agent of energy beams while passing through it. The optimum thickness of the target layers were procured for 90° beam-target angle geometry employing the SRIM code. The beam, that was specified by SDEF card, has a Gaussian distribution in energy and in divergence with the mean full width at half maximum (FWHM) values of about 300 keV and 17.4 mrad, respectively, and a circular shape in the plane perpendicular to the incident direction of the

beam, with a Gaussian spatial distribution and FWHM value of 4 mm.

Simulation yield calculation

Regarding the simulation-based yields, incident particle flux (proton and deuteron) and energy distributions were computed utilizing MCNPX version 2.6. The projectile energy bin width was set to 0.5 MeV and energy range extended to 30 MeV. Repeated simulations were run with a history of 10^7 projectiles as a variance reduction method. To estimate radionuclide activity, the normalized particle energy distribution function $P(E)$ in the tellurium target body was computed from the "f4/e4" tally output (flux) for each reaction. The results are shown in fig. 2. As can be seen, energy distributions of incident particles in the tellurium target layers cover the desired irradiation energy ranges, *i. e.* 14 to 7 MeV for protons in ^{124}Te , 21 to 15 MeV for protons in ^{125}Te , 11 MeV to 6 MeV for deuterons in ^{123}Te , and 15 MeV to 10 MeV for deuterons in ^{124}Te target.

Hence, the efficacious distribution function $P(E)\sigma(E)$ was computed from the model based cross-sections from TALYS-1.8 code. By integrating the following differential equation for a radioactive product nuclide, the activity of the generated radionuclides can be determined [23]

$$A(t) = \frac{I\rho dN_A}{M} (1 - e^{-\lambda t}) \int_{E_{out}}^{E_{in}} P(E)\sigma(E)dE \quad (1)$$

where $A(t)$ is the product nuclide activity (in Bq), N_A – the Avogadro number, I – the projectile beam current, λ – the radionuclide decay constant, t – the time of irradiation, E_{in} and E_{out} are the initial and outgoing energies of the beam. M , ρ , and d are the molar mass, density and thickness of the target material, respectively. The tellurium target was irradiated for 60 min with 1 A beam current.

SRIM code calculations

There are several computational codes for calculating the stopping power and the range of incident particles and also the required thickness of target matter. SRIM code is a group of programs which compute the stopping and range of ions (up to 2 GeV/amu) in the target matter utilizing a quantum mechanical treat-

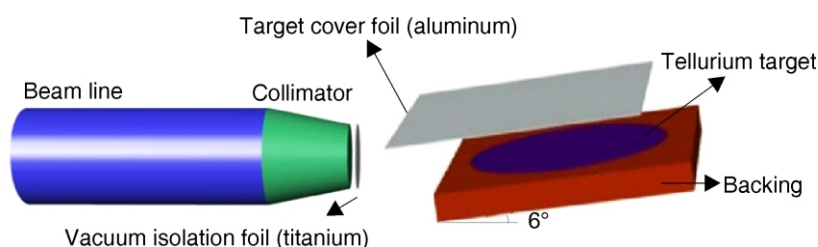


Figure 1. Simulation setup of target geometry and source configuration designed by visual editor MCNPX

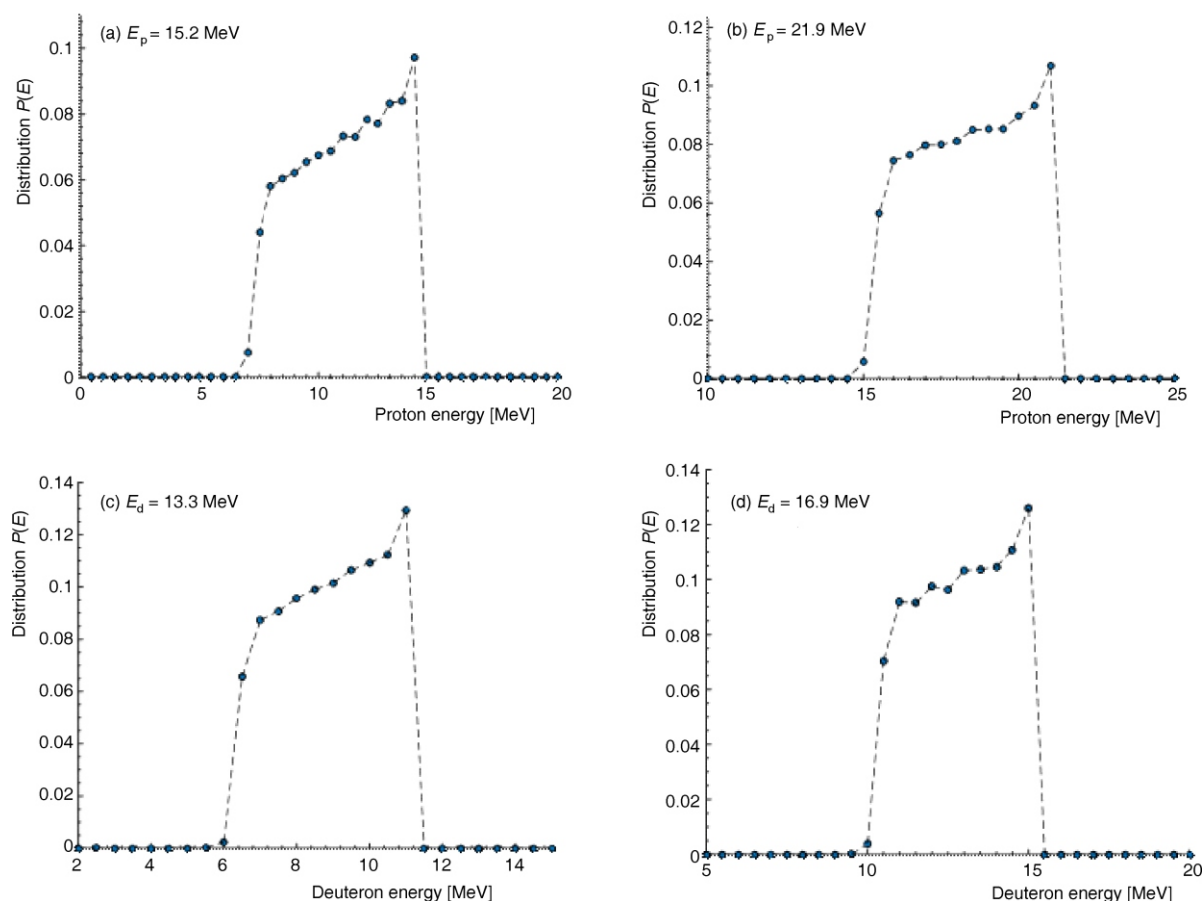


Figure 2. The normalized energy distribution function (MCNPX tally) for: (a) protons in ^{124}Te target, (b) protons in ^{125}Te , (c) deuterons in ^{123}Te , and (d) deuterons in ^{124}Te target. The irradiation energies were indicated in each figure

ment of ion-atom collisions. Furthermore, this code is used to calculate the thick blocks of materials employed to lower the energy of a beam of particles. This is mostly carried out for light ions to rapidly obtain lower energies. The stopping power and range of incident particles (proton and deuteron) in the target matter (tellurium isotopes) and also the physical thickness of target block were calculated for the given beam-target angle geometry (6°) in optimum energy range using the SRIM-2013 code [16, 24-27].

Theoretical yield calculation

To calculate the theoretical yield, the cross-sectional data and the stopping power of the projectile in the target, were acquired by employing the ALICE/ASH, TALYS-1.8 and SRIM codes, respectively. The thick-target integral yields can be calculated employing the following equation [14]

$$A(t) = \frac{IN_A}{M} (1 - e^{-\lambda t}) \int_{E_{out}}^{E_{in}} \frac{dE}{d(\rho x)} \sigma(E) dE \quad (2)$$

where $dE/d(\rho x)$ is the stopping power of incident particle in target matter. Other parameters are the same as given in eq. (1). The Simpson numerical integral method was used to solve the integral.

RESULTS AND DISCUSSION

Monte Carlo calculations

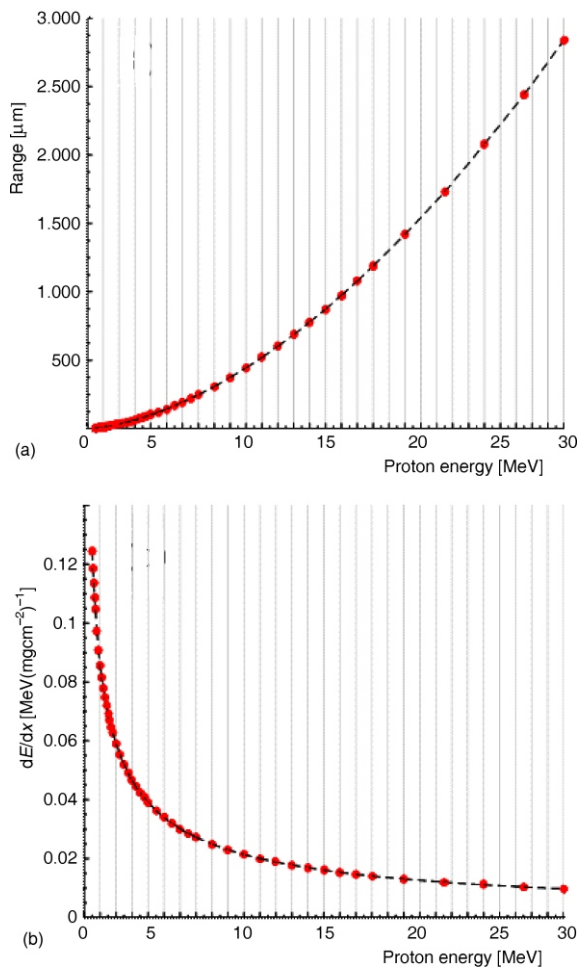
The production of ^{124}I was predicted through $^{124}\text{Te}(p, n)$, $^{125}\text{Te}(p, 2n)$, $^{123}\text{Te}(d, n)$ and $^{124}\text{Te}(d, 2n)$ reactions with different energies. Tellurium targets covered by Al-foil (fig. 1) were irradiated through 4 different energy levels, for 1 hour. The simulation yields of ^{124}I production based on the TALYS-1.8 code were obtained through modeled proton and deuteron energy distribution functions in the targets body (fig. 2). Furthermore, the simulation yield of undesired nuclides, ^{123}I and ^{125}I , were obtained to determine the best energy range to decrease the production of undesired radionuclide impurities. The results are shown and compared with the available experimental values in tab. 1.

According to SRIM code calculation plotted in fig. 3, a 540 μm thickness is required for the ^{124}Te target irradiated in proton energy range of 14-7 MeV; the thickness was chosen in such a way that the outgoing particle energy should be 7 MeV. The corresponding values are 681, 212, and 272 μm for ^{125}Te , ^{123}Te and ^{124}Te targets, respectively, according to the incident particle energy. In the investigated energy regions of various ^{124}I production channels, two other radionuclides, ^{123}I (13.2 h) and ^{125}I (59.4 d), may be

Table 1. Comparison of ^{124}I simulated yields with the theoretical and experimental data in the optimum energy range of projectiles

Reactions	Effective energy range [MeV]	Optimum target thickness [mm]	Theoretical yield (^{124}I) [MBq(Ah) $^{-1}$]		Simulation yield* [MBq(Ah) $^{-1}$]			Experimental yield (^{124}I) [MBq(Ah) $^{-1}$]
			TALYS-1.8	ALICE/ASH	^{123}I (13.2 h)	^{124}I (4.17 d)	^{125}I (59.3 d)	
$^{124}\text{Te}(p, n)^{124}\text{I}$	14 7	0.540	27.54	29.18	41.92	25.76	0.0027	21.1 [26]
$^{125}\text{Te}(p, 2n)^{124}\text{I}$	21 15	0.681	78.58	86.37	50.22	70.83	0.36	81 [9]
$^{123}\text{Te}(d, n)^{124}\text{I}$	11 6	0.212	2.54	2.84	60.06	2.4	1.0E-5	2.8 [27]
$^{124}\text{Te}(d, 2n)^{124}\text{I}$	15 10	0.272	20.40	20.79	0.67	18.88	0.17	20.7 [3]

*All values calculated from the TALYS acquired excitation functions

**Figure 3. Range (a) and stopping power of protons (b) in the monoisotopic ^{124}Te target**

formed as impurities. The ^{123}I , because of its short half-life, is not a major concern and can be drastically reduced by allowing to decay a few days after the end of bombardment. However, ^{125}I , due to its rather long half-life, is the major concern in the ^{124}I production procedure and makes its removal from product nuclides difficult. To resolve this, it is necessary to strategically select the irradiation energy region in each channel to maximize the ^{124}I production yield and minimize the produced impurities, especially the ^{125}I nuclide. The selection of this energy range leads to a disagreement between purity and yield.

According to results (tab. 1), the proton induced reaction on ^{124}Te target is a method of choice because

of its rather low amount of ^{125}I impurity. The high amount of ^{123}I product can be minimized by allowing it to decay a few days after irradiation. The $^{125}\text{Te}(p, 2n)^{124}\text{I}$ reaction can be utilized in the medium-cyclotrons, nevertheless, the high level of ^{125}I produced in this route is the major problem. The production yield of ^{124}I in the $^{123}\text{Te}(d, n)^{124}\text{I}$ reaction is very low and by taking into account the high cost of target material ^{123}Te , this is insignificant for the ^{124}I production. In the $^{124}\text{Te}(d, 2n)^{124}\text{I}$ reaction, the level of ^{125}I impurity is relatively high, therefore this reaction, when compared to the $^{124}\text{Te}(p, n)^{124}\text{I}$ reaction, is recessive.

On the other hand, the results show the 8.79-22.08 % relative discrepancies between the MC code and corresponding experimental data and also the 5.51-9.86 % between MC code and statistical codes. As is well known, the experimental yield is influenced by several factors, such as inhomogeneity in the target layer, beam profile and intensity, radiation damage effects, and chemical separation yield. On the other hand, in this simulation the targets were considered 100 % pure isotopes, whereas in the experimental works, the targets have a slight contaminant. Therefore, it can be said that the computed results are in acceptable agreement with the experimental values. Hence, the MCNPX 2.6 code is a good tool for the prerequisite calculation and yield estimation of ^{124}I production with the reasonable uncertainties.

Theoretical calculations

Natural tellurium consists of 8 stable isotopes: ^{120}Te 0.09 %, ^{122}Te 2.55 %, ^{123}Te 0.89 %, ^{124}Te 4.74 %, ^{125}Te 7.07 %, ^{126}Te 18.84 %, ^{128}Te 31.74 %, and ^{130}Te 34.08 % [14]. The theoretical yield of ^{124}I production based on TALYS-1.8 and ALICE/ASH codes were calculated for monoisotopic ^{123}Te , ^{124}Te , and ^{125}Te targets in the mentioned reactions. The calculation results were utilized as a benchmark data for simulation yields to determine the accuracy of the MC code. The calculation results are presented in tab. 1. Furthermore, as an application of SRIM code, the stopping power and range of protons in the ^{124}Te target were obtained in a wide energy range (fig. 3).

The results of these figures have been utilized to determine the optimum thickness of the target body, to achieve the required energy range of incident particle in that region (as mentioned above).

CONCLUSION

In this study, the ability of analytical and Monte Carlo models was used to predict nuclear parameters.

The simulation production yields, over a wide energy range, were predicted for $^{124}\text{Te}(p, n)^{124}\text{I}$, $^{125}\text{Te}(p, 2n)^{124}\text{I}$, $^{123}\text{Te}(d, n)^{124}\text{I}$, and $^{124}\text{Te}(d, 2n)^{124}\text{I}$ reactions through MCNPX and SRIM codes. The result of calculations was compared with the TALYS-1.8 and ALICE/ASH results, as well as experimental data, for the same reactions. Eventually, the reasonable agreement between the theoretical and simulation production yields, as well as experimental data, was achieved in optimum energy range. According to the results, the $^{124}\text{Te}(p, n)^{124}\text{I}$ process appears to be the most likely candidate to produce the ^{124}I in low-energy cyclotrons, because of the sufficient amounts of ^{124}I production, with relatively low levels of ^{125}I impurity as the major concern in the ^{124}I production due to its rather long half-life of 59.4 days.

AUTHORS' CONTRIBUTIONS

The Monte Carlo and theoretical calculations were undertaken by H. Azizakram. M. Sadeghi supervised the presented research and development that resulted in this paper. P. Ashtari prepared the cyclotron target geometry. F. Zolfagharpour was the advisor of this work and provided theoretical concepts of research. All authors were included into result analysis and discussion, while manuscript and figures' preparation was done by H. Azizakram.

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МОНТЕ КАРЛО ПРИСТУП ПРОРАЧУНУ ПРЕДУСЛОВА ПРОИЗВОДЊЕ РАДИОИЗОТОПА ^{124}I РАДИ ПРОЦЕНЕ АКТИВНОСТИ

Применом програмског пакета MCNPX симулирана је производња ^{124}I путем реакција $^{124,125}\text{Te}(p, xn)$ и $^{123,124}\text{Te}(d, xn)$ ради добијања ^{124}I високе активности. Ефикасни пресеци за ове реакције израчунати су помоћу програмских пакета TALYS-1.8 и ALICE/ASH. Оптимални енергетски опсег пројектила за ову производњу изабран је одређивањем максимума ефикасног пресека и минимума нечистоћа услед емисије осталих канала. Геометрија мете дизајнирана је на основу прорачуна зауставне моћи програмским пакетом SRIM при идентичним димензијама као и у експерименталним подацима. Претпостављен је допринос услед реакција на дебелој мети због функција ексцитације и зауставне моћи. Сви предуслови добијени претходним прорачунима прилагођени су MCNPX програмском пакету и производни процес симулиран је према бенчмарк експериментима. Потом је израчуната енергетска расподела протона у метама и количина преосталих језгара после озрачивања. Резултати су у доброј сагласности са објављеним подацима чиме се потврђује погодност и тачност програмског пакета MCNPX као средства за оптимизацију производње других радионуклида. На основу резултата, реакција $^{124}\text{Te}(p, n)^{124}\text{I}$ чини се да је највероватнији кандидат за производњу ^{124}I у нискоенергетским циклотронима.

Кључне речи: ^{124}I , допринос производње, циклојтрон, MCNPX, TALYS-1.8, ALICE/ASH