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OPTIMIZING IRRADIATION PARAMETERS OF CYCLOTRON-PRODUCED RADIONUCLIDES Cu-64, I-123 and I-124

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ABSTRACT

OPTIMIZING IRRADIATION PARAMETERS FOR CYCLOTRON-PRODUCED RADIONUCLIDES Cu-64, I-123 AND I-124. *Successful production of cyclotron-based radionuclides depends on various irradiation parameters including the type, energy and beam current of incident particle, target thickness and geometry as well as irradiation time. This paper presents theoretical calculations for optimization of future Cu-64, I-123 and I-124 radionuclide production using BATAN's CS-30 cyclotron from $^{64}Ni(p,n)^{64}Cu$, $^{123}Te(p,n)^{123}I$, $^{124}Te(p,n)^{124}I$ and $^{124}Te(p,2n)^{123}I$ nuclear reactions. Optimum target thickness and proton incidence angle, proton energy and beam current which result in optimum End Of Bombardment (EOB) yields are highlighted. A well-developed and widely available software called Stopping and Range of Ion in Matter (SRIM) version 2013 are employed to determine the optimum Ni and Te target thickness for several irradiation requirements and then followed by calculations of the EOB yields. In addition, the calculated EOB yields are then compared with previous experimental results obtained elsewhere. For production of Cu-64, the optimum Ni target thickness when a 26.5 MeV proton beam is incident at 0° angle relative to the target normal should be nearly 1.5 mm which yields up to 560 mCi/µA.hr at the end of the bombardment. At proton beam energy of 12 MeV, 12 MeV and 22 MeV for production of I-123 from Te-123 target, I-124 and I-123 from Te-124 targets respectively, the associated optimum target thickness are 0.64 mm, 0.65 mm and 1.8 mm whereas their EOB yields are predicted to be 30.6 mCi/µA.hr, 4.2 mCi/µA.hr and 159.3 mCi/µA.hr respectively. For all Cu-64, I-123 and I-124 radionuclide production, comparisons with some selected experimental data indicate that the much-lower-than-expected EOB yields are mainly due to incorrect target thickness prepared for the irradiation.*

Key words *: cyclotron, proton, Ni and Te targets, Cu-64, I-123 and ^{124}I-124, EOB yield*

ABSTRAK

OPTIMASI PARAMETER IRADIASI UNTUK RADIONUKLIDA Cu-64, I-123 DAN I-124 YANG DIPRODUKSI MENGGUNAKAN SIKLOTRON. *Keberhasilan produksi radionuklida berbasis siklotron tergantung pada berbagai parameter iradiasi, termasuk jenis, energi dan arus berkas partikel penembak, tebal dan geometri target dan juga waktu iradiasi. Makalah ini menyampaikan perhitungan secara teori untuk optimasi produksi radionuklida Cu-64, I-123 dan I-124 dimasa yang akan datang dari reaksi nuklir $^{64}Ni(p,n)^{64}Cu$, $^{123}Te(p,n)^{123}I$, $^{124}Te(p,n)^{124}I$ dan $^{124}Te(p,2n)^{123}I$ menggunakan siklotron CS-30 yang dimiliki oleh BATAN. Ketebalan target, sudut penembakan, energi proton dan arus proton yang menghasilkan optimum End Of Bombardment (EOB) yields dibahas dalam makalah ini. Dalam penelitian ini, software Stopping and Range of Ion in Matter (SRIM) versi 2013 yang tersedia secara gratis dan online digunakan untuk menentukan ketebalan optimum target Ni dan Te untuk beberapa kondisi iradiasi yang kemudian dilanjutkan dengan perhitungan EOB yield. Lebih jauh lagi, hasil perhitungan EOB yield selanjutnya dibandingkan dengan data eksperimen yang sebelumnya telah dipublikasikan. Untuk produksi Cu-64, ketebalan optimum target Ni ketika berkas proton berenergy 26,5 MeV ditembakkan tegak lurus terhadap permukaan target adalah sekitar 1,5 mm dengan hasil EOB yield sebesar 560 mCi/µA.jam. Dengan energi proton masing-masing sebesar 12 MeV, 12 MeV dan 22 MeV untuk produksi I-123 dari target Te-123, I-124 dan I-123 dari target Te-124, target Te hendaknya dibuat setebal 0,64 mm, 0,65 mm dan 1,8 mm untuk mendapatkan EOB yield masing-masing sebesar 30.6 mCi/µA.jam, 4.2 mCi/µA.jam and 159.3 mCi/µA.jam. Untuk produksi ketiga radionuklida Cu-64, I-123 dan I-124 tersebut, data eksperimen menunjukkan hasil EOB yang jauh lebih kecil dari perhitungan teoritis karena adanya kesalahan penentuan tebal target.*

Kata kunci *: siklotron, proton, target Ni dan Te, Cu-64, I-123, I-124, EOB yield*

INTRODUCTION

The CS-30 cyclotron owned by the Indonesian National Nuclear Energy Agency (BATAN) in Serpong is currently under maintenance and is scheduled to be employed for production of short-lived radionuclides for medical purposes in the near future. The H^{+}-accelerating cyclotron is capable of producing proton beam of up to 26.5 MeV at variable external beam current of up to 30 μA. Depending on the target of interest, the proton energy can be reduced using aluminum degraders.

Medical radionuclides such as ^{64}Cu, ^{123}I and ^{124}I produced by proton irradiation have been widely used and developed overseas for pre-therapeutic dosimetric studies [1 – 4]. ^{64}Cu is produced via nuclear reaction $^{64}Ni(p,n)^{64}Cu$ and has been of great interest due to its potential applications in medical field, particularly for cancer diagnosis. The β^{+} emitting ^{64}Cu whose half-life is 12.7 hours is used for Positron Emission Tomography (PET). The threshold energy for $^{64}Ni(p,n)^{64}Cu$ is nearly 2.5 MeV and the maximum cross-section is approximately 765 mbarn which occurs at nearly 10 MeV based on TALYS-calculated data [5]. Enriched nickel targets (^{64}Ni) in the form of electroplated targets have been widely suggested as the best target for ^{64}Cu production [1,2], though natural Ni target has also been of interest elsewhere [6].

Radionuclide ^{123}I decays by electron capture, which is immediately followed by emission of gamma ray with a predominant energy of 159 keV at a half life of 13.22 hours. The gamma ray is primarily used for imaging by means of Single Photon Emission Computed Tomography (SPECT). In contrast, radionuclide ^{124}I is a β^{+} emitter with a half life of 4.18 days, which is useful for Positron Emission Tomography (PET).

Both medical radioactive iodine ^{123}I and ^{124}I can be produced by either direct or indirect methods. Direct method of producing ^{123}I and ^{124}I uses a relatively low energy (8 – 22 MeV) cyclotron [7] as a proton accelerator in which the proton beam is then irradiated into enriched or natural tellurium (Te) targets. However producing the radioisotopes from natural Te targets requires relatively higher proton energy, yet it results in much lower radioactivity than those produced from enriched Te targets because of their low cross-sections [8].

Successful production of the PET and SPECT radionuclides requires thorough understanding of the irradiation parameters, including energy of proton as an incident particle, incidence angle, target preparation and thickness, proton beam current as well as irradiation time. Knowledge about optimum proton energy is essential since it corresponds to the threshold energy and cross-section/excitation function of a particular target when the incident proton is bombarded into the target surface.

Target preparation is also one of the crucial factors to consider prior to the target irradiation. Careful studies of the types of targets (i.e. electroplated targets, foil targets or mixed targets) should be carried out to minimize failures associated with the target handling before, during and after irradiation as well as optimum radioactivity yields.

Another important parameter relevant to the ^{64}Cu production is the target thickness as it corresponds to the radioactivity yield. Knowledge about proton distributions in the Ni and Te targets is, therefore, paramount to successfully determine the correct target thickness prior to proton irradiation. The proton distributions in Ni and Te targets can be examined from the particle's stopping power/energy loss and range, which can be calculated using the Stopping and Range of Ion in Matter (SRIM) package [9]. In the SRIM codes, stopping power is defined as the energy required to slow down the incident particle during its interaction with matter over a certain distance, whereas the distance over which the ion totally stops is called the range. Mathematical equations correspond to the stopping and range of ion in matter have been described elsewhere [10], and that the SRIM-calculated data agree with experimental results within 10% accuracy or less [11].

Since the threshold energy for $^{64}Ni(p,n)^{64}Cu$ nuclear reaction is nearly 2.5 MeV, any proton irradiation over 2.5 MeV will result in some radioactive yields during and at the end of the bombardment. For $^{123}Te(p,n)^{123}I$, $^{124}Te(p,n)^{124}I$ and $^{124}Te(p,2n)^{123}I$ the threshold energy is 5 MeV, 5 MeV and 10 MeV respectively. The End-Of-Bombardment (EOB) yield (Y) for any nuclear particle-produced radioisotope is not only dependent on the nuclear cross-section at a particular energy, $\sigma(E)$, but also on the stopping power, $d(E)/dx$, and some other parameters as described by [9]:

$$Y = \emptyset(1-e^{-\lambda t})\frac{N_A}{M}\int_{E_i}^{E_{th}}\left[\frac{\sigma(E)}{\frac{1}{\rho}\frac{d(E)}{dx}}\right]dE \quad \text{...... (1)}$$

Where Φ is the number of charged particles per unit of time, λ is the decay constant of the resulting radioisotope, t is the duration of irradiation, N_A is the Avogadro number, ρ and M are the mass density and atomic mass of the target respectively, E_i is the initial energy of the incident particle, and E_{th} is the threshold energy.

This paper reports on the use of the SRIM codes to discuss the range and dissipated energy of energetic protons in Ni and Te targets relevant to Cu-64, I-123 and I-124 production. The EOB yields associated with the proton-irradiated Ni and Te targets are also discussed for several irradiation parameters, including target thickness, proton beam current and irradiation time. The predicted results are also compared with the experimental and calculated data available elsewhere.

METHODOLOGY

SRIM Calculations

The SRIM package employed in the simulations was the SRIM 2013 version, in which proton beams in the energy range between 5 MeV and 50 MeV were incident in ^{64}Ni, ^{123}Te and ^{124}Te targets, initially normal to the targets surfaces. In order to study the dependence of the proton beam range on the incidence angle, the targets were theoretically irradiated at several incidence angles ranging from 0° to 70° relative to the targets surfaces as depicted in Fig. 1, with nearly 100,000 protons simulated in the calculations.

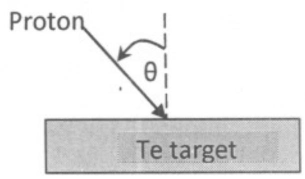

Fig. 1 Proton beam and Te target set-up in the SRIM calculations.

The proton energy of 12 MeV and 22 MeV were chosen for the angle variation study in ^{123}Te and ^{124}Te targets respectively whereas proton energy of 10 MeV were employed for ^{64}Ni target.

Calculations of End-Of-Bombardment (EOB) Yields

The EOB Yields were theoretically calculated using equation (1) for optimum ^{64}Cu, ^{123}I and ^{124}I yields from proton-irradiated ^{64}Ni, ^{123}Te and ^{124}Te targets at a number of proton beam current of up to 30 μA (equal to the maximum possible current the BATAN's CS-30 cyclotron could generate). The first term (Φ) in equation (1), for a proton beam as the incoming particle, can be expressed as [12]:

$$\emptyset = \frac{6.25x10^{18}}{Z} I \qquad (2)$$

Where Z is the charge of proton, and I is the beam current.

The irradiation parameters used for the calculations are given in Table 1 while the excitation functions for the particular nuclear reactions were based on the TALYS-calculated data found in reference [5] and are shown in Fig. 2. The procedures for calculating the EOB yield have been described elsewhere for ^{18}F production [12].

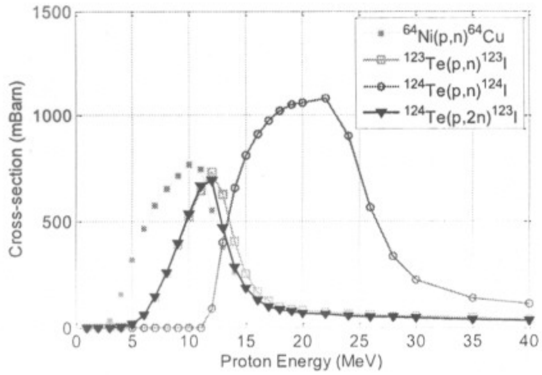

Fig. 2 TALYS-Calculated excitation function of $^{64}Ni(p,n)^{64}Cu$, $^{123}Te(p,n)^{123}I$, $^{124}Te(p,n)^{124}I$ and $^{124}Te(p,2n)^{123}I$ nuclear reactions [5].

RESULTS AND DISCUSSION

Calculated Results for Cu-64

The behavior of the proton beam distributions in the energy range between 5 MeV and 30 MeV is relatively similar which can be inferred from the shape of their energy loss/stopping power plots (Fig. 3). In general, for any proton energy, the stopping power increases with increasing distance of travel until it peaks at a certain value (called Bragg peak) and then drops dramatically following the loss of the proton energy.

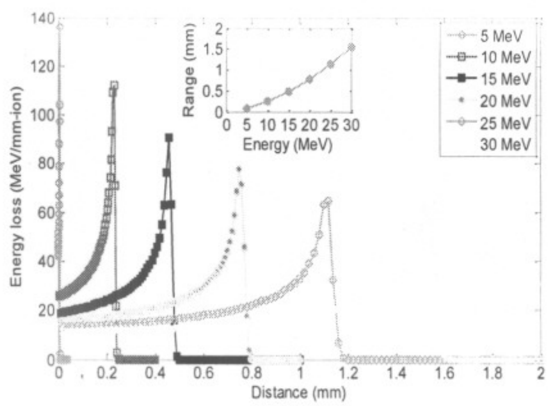

Fig. 3 Energy loss of several energetic proton beams ranging from 5 MeV to 30 MeV in nickel target, calculated using the SRIM 2013 version package [10]. The corresponding ranges are shown in the inset

In contrast to the general trend of the energy loss, in which it decreases with increasing proton energy, the range increases with increasing proton energy as shown in the inset of Fig. 3. The range goes up quite steeply from 73.8 μm at proton energy of 5 MeV to 154 μm for the 30-MeV proton beam, whereas there are 47 target atoms displaced by the incoming 5 MeV proton beam compared to 137 vacancies as a result of the 30-MeV proton irradiation.

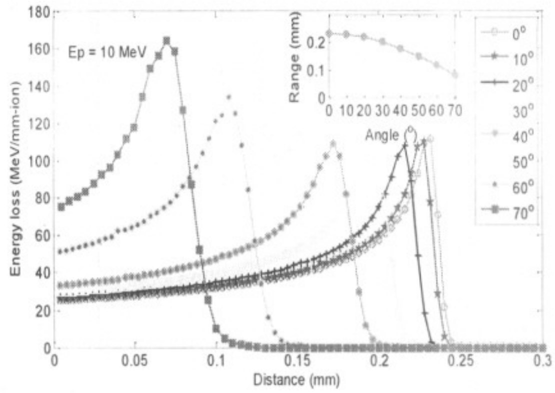

Fig. 4 Stopping power and range (inset) of a 10-MeV proton beam in Ni target at various angles of incidence

The dependence of the proton range on the incidence angle for proton energy of 10 MeV is plotted in Fig. 4. For a beam of 10-MeV protons, the larger the incidence angle the shorter the distance it travels, which is due to higher stopping power as depicted in Fig. 4. In other words, the range of the proton is shorter as the incidence angle increases (inset, Fig. 4). It is also clear that the distribution of the energy loss broadens with increasing incidence angle.

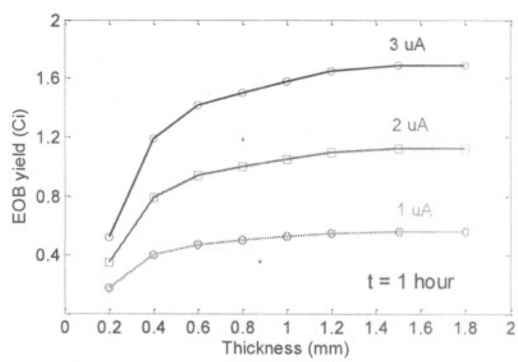

Fig. 5 EOB yields as a function of Ni target thickness at different proton beam current ranging from 1μA to 3 μA and fixed energy of 26.5 MeV for irradiation time of 1 hour

Using equation (1) and (2), as stated earlier in the calculation section, the EOB yields of a 26.5-MeV proton beam at different current between 1µA and 3µA were calculated as a function of ^{64}Ni target thickness depicted in Fig. 5 which indicate similar behavior for irradiation time of 1 hour. The rapid increase in the ^{64}Cu yields is evident when the 26.5-MeV proton beam is irradiated into a less-than-0.5-mm Ni target, though the EOB yields rise further at a slower rate before they eventually level off when the target is over 1.2-mm thick. In theory, there will be no added radioactivity yield should the Ni target thickness is increased further to greater than 1.5 mm thick. For an hour irradiation time, the maximum EOB yield is expected to be approximately 0.56 Ci for proton beam current of 1 µA.

In order to further study the influence of irradiation time and Ni target thickness over the EOB yields, a range of yield calculations were carried out with 10-minute increments, again at fixed proton energy of 26.5 MeV, and the results are shown in Fig. 6 for a proton beam of 1 µA. The dramatic surge in the EOB yields can be clearly seen in the figure for all investigated Ni target thickness ranging from 0.2 nm to 1.5 nm. EOB yields of up to 1.44 Ci is expected to be produced following the irradiation of a 1.5-mm thick Ni target over a period of 180 minutes (3 hours).

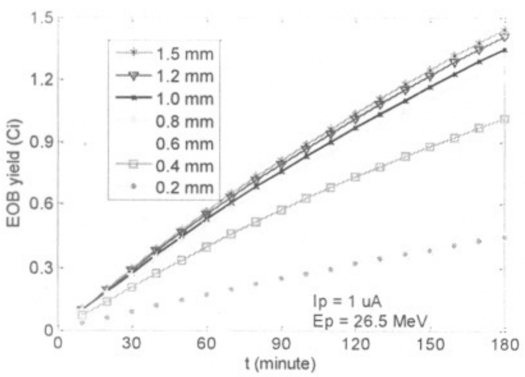

Fig. 6 EOB yields as a function of irradiation time at different Ni target thickness and fixed energy of 26.5 MeV for proton beam current of 1 µA

To sum up an optimum EOB yield of 560 mCi/µA.hr (0.56 Ci/µA.hr) is expected to be achieved when a 1.5-mm enriched Ni target is irradiated using the BATAN's 26.5-MeV proton cyclotron. However when the target thickness is less than the optimum thickness, the EOB yield would be down to approximately 173 mCi/µA.hr for a 200-µm Ni target.

A range of experimental data were collected from several references to verify the calculated EOB yields as listed in Table 1 (for Ep = 12 – 15.5 MeV) . Using a 12-MeV proton beam, Obata *et al* irradiated enriched Ni targets at a constant beam current of 50 µA. At the end of the bombardment, they obtained ^{64}Cu radioactivity yields of 3.079 mCi/µA.hr, 3.734 mCi/µA.hr, and 6.565 mCi/µA.hr for target thicknesses of 127.45 µm, 144.16 µm and 277.28 µm respectively. These experimental results are, however, much lower than the predicted results calculated in this report as well as those obtained elsewhere [13].

Based on the SRIM-calculated data, a 12-MeV proton beam is able to penetrate relatively deep into a Ni target and pass the target at an average range of 377.2 µm (Fig. 4). Therefore, the optimum yield of around 6.89 mCi/µA.hr at this particular proton energy would only be obtained if the Ni target thickness was around 377.2 µm. However in the case of Obata, *et al* investigation [14], they employed up to 277.28-µm thick Ni targets to produce ^{64}Cu, which are too thin to totally stop the incoming 12-MeV proton beam. At a distance of 277.28 µm from the Ni surface, the protons would lose nearly 10.58 MeV of their total energy; hence, a vast number of protons would pass through the thin Ni target and deposit only some fraction of their total energy. This explanation also applies to the other thinner Ni targets. For this reason, the proton-bombarded Ni targets in their experiments resulted in much lower-than expected EOB yields.

Calculated Results for I-123 and I-124

The energy loss as a function of the total distance traveled by 5 – 50 MeV proton beams in ^{123}Te and ^{124}Te targets (calculated using SRIM 2013 codes; equation (1) and (2)) is shown in Fig. 7 and Fig. 8, in which they exhibit a very similar behavior over the energy range. In general, the energy loss

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decreases with increasing proton energy, and that the particle distribution in each target broadens at higher energy. For the same proton incident energy, the energy loss of the nuclear particle is slightly higher in ^{124}Te compared with that of in ^{123}Te, though the difference stands at less than 3%.

The projected range is plotted as a function of the proton incident energy for each elemental target (inset, Fig. 7 and 8) which conforms that proton penetrates deeper into the material target as the energy is increased, and that the range is inversely proportional to the stopping power. A slight difference in the projected range of the same incident energy is noticeable, even though it is less than 1%. For example, for proton energy of 50 MeV, the projected range of the incoming proton beam in ^{123}Te and ^{124}Te targets are 6.62 mm and 6.67 mm respectively.

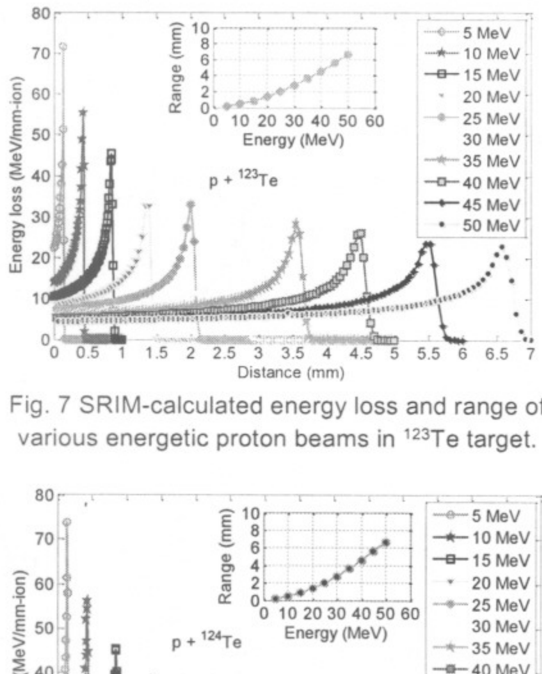

Fig. 7 SRIM-calculated energy loss and range of various energetic proton beams in ^{123}Te target.

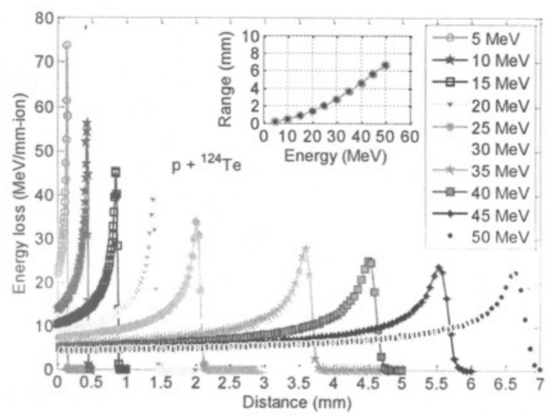

Fig. 8 SRIM-calculated energy loss and range of various energetic proton beams in ^{124}Te target.

The dependence of proton range on the incidence angle in both elemental targets is shown in Fig. 9 and 10. The range of a 12-MeV proton beam was evaluated in ^{123}Te target for proton incidence angle ranging from 0° to 70° with respect to the target normal (Fig. 9), whereas the same range of incidence angle was simulated for a 22-MeV proton beam in ^{124}Te target (Fig. 10). For both energetic proton beams investigated in this report, the larger the incidence angle, the broader the ion distribution in the target and the shallower the penetration. Another interestingly similar behavior is that the energy loss drops with increasing incidence angle of up to 30°, but then it relatively levels off up to 45° followed by a sudden increase as the incidence angle goes up further. As well, the projected range of both the 12-MeV and 22-MeV proton beams in Te targets decreases very quickly as the incidence angle increases, however their nominal projected range is very different (inset, Fig. 9 and 10). For instance, at 0°-incidence angle, the projected range for the 12-MeV protons is 585 μm in ^{123}Te target, while the projected range of the 22-MeV protons at the same angle is 1.64 mm.

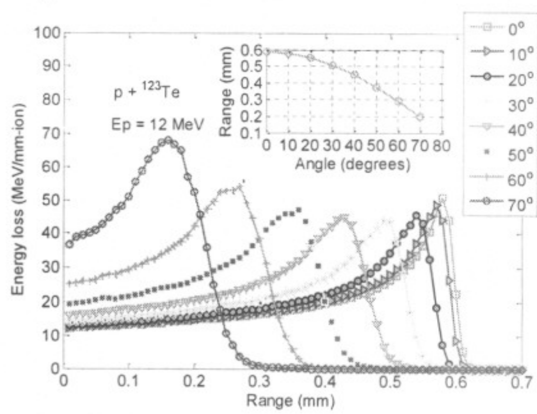

Fig. 9 SRIM-calculated energy loss and range of a 12-MeV proton beam in ^{123}Te target at varied incidence angles.

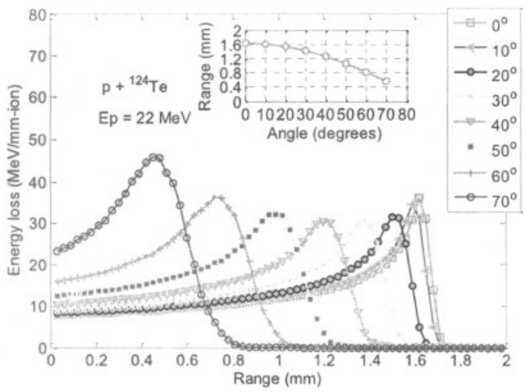

Fig. 10 SRIM-calculated energy loss and range of a 22-MeV proton beam in ^{124}Te target at varied incidence angles.

The recommended Tellurium target thickness may be determined from the projected range of the particular proton beam in which it completely dissipates its energy into the target surface and then added by a 10% of its projected range to compensate with the standard error since the accuracy of the calculated range and stopping power is within 5 – 10% [9]. When the Te target is irradiated at 0° incidence angle with respect to target normal, the optimum target thickness for producing ^{123}I is 0.64 mm and 1.8 mm from ^{123}Te(p,n)^{123}I and ^{124}Te(p,2n)^{123}I nuclear reactions respectively, whereas a target thickness of 0.65 mm is required for generating ^{123}I from ^{124}Te(p,n)^{124}I reaction. For the three investigated nuclear reactions, the targets should be made thinner with larger incidence angles.

It is widely known that for proton-produced radionuclides, the radioactivity yield depends on the irradiation time and beam current as discussed by Suryanto, *et al* [12] for ^{18}F production from ^{18}O(p,n)^{18}F nuclear reaction. In the case of ^{123}I and ^{124}I production, the predicted EOB yields have been theoretically calculated using equation (1) for ^{123}Te(p,n)^{123}I, ^{124}Te(p,n)^{124}I and ^{124}Te(p,2n)^{123}I nuclear reactions as can be seen in Fig. 11, in which the proton energy was set to be 12 MeV for ^{123}Te(p,n)^{123}I and ^{124}Te(p,n)^{124}I reactions, and 24 MeV for ^{124}Te(p,2n)^{123}I reaction. As expected, in general, the radioactivity yield increases with increasing duration of irradiation and beam current with ^{123}I from ^{124}Te(p,2n)^{123}I reaction yields the highest radioactivity among the three since it has the highest cross-section.

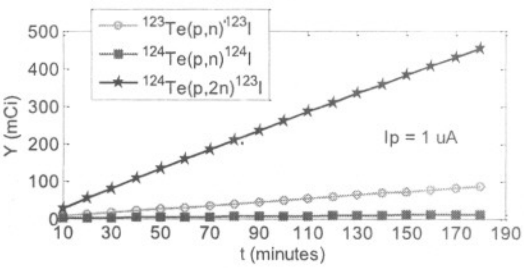

Fig. 11 Predicted EOB yields for ^{123}Te(p,n)^{123}I, ^{124}Te(p,n)^{124}I, and ^{124}Te(p,2n)^{123}I nuclear reactions at proton beam current of 1 μA.

At a beam current of 1μA, the maximum EOB yields for ^{123}Te(p,n)^{123}I, ^{124}Te(p,n)^{124}I and ^{124}Te(p,2n)^{123}I nuclear reactions after 3 hours bombardment are 12.4 mCi, 87.2 mCi and 454 mCi respectively. In other words, at proton beam energy of 12 MeV for production of I-123 from Te-123 target and I-124 from Te-124 target, and 22 MeV for production of I-123 from Te-124 target, the associated EOB yields are predicted to be 30.6 mCi/μA.hr, 4.2 mCi/μA.hr and 159.3 mCi/μA.hr respectively at their corresponding optimum thickness.

A case study was done in order to verify if the EOB prediction was close enough to the experimental results. The data were taken from the experiments conducted by R. C. Barrall, et al [12] in which they bombarded a 300-μm-electroplated Tellurium target with 11.5 MeV and 15 MeV proton beams at a current of 133 μA for 2 hours. As shown in Table 1, the calculated results are very close to the experimental data with accuracy of 10% or less. Nevertheless the calculated EOB yields in Table 1 are not the optimum yields they should have gotten. Based on the SRIM simulation, with a 11.5-MeV proton beam, the optimum target thickness should have been 0.56 mm to get a maximum EOB yield of 2.1 mCi/μA.hr, whereas at a proton beam of 15 MeV, the optimum target thickness should have been 0.87 mm to get nearly 9.9 mCi/μA.hr. Therefore a huge fraction of the yields must have been lost due to improper target thickness (too thin Te targets) in the experiments.

Table 1 Comparison of experimental and calculated EOB yields

Ep (MeV)	Ep (μA)	t (hr)	Thickness (μm)	EOB Yield (mCi/μA.hr)	
				Experiment [14]	Calculation
11.5	33	2	300	1.0	1.1
15	33	2	300	3.2	3.4

CONCLUSION

Enriched Ni target thickness, proton beam current and irradiation time are among the very important parameters to consider for the purpose of successful ^{64}Cu, ^{123}I and ^{124}I production using the BATAN's cs-30 cyclotron. For a 26.5-MeV proton beam, the optimum target thickness is nearly 1.5 mm which yields up to 560 mCi/μA.hr at the end of the bombardment. The calculated results indicate that for ^{123}Te(p,n)^{123}I, ^{124}Te(p,n)^{124}I and ^{124}Te(p,2n)^{123}I nuclear reactions, the targets should be made thinner with larger incidence angles. The calculated EOB yield could reach up to 13.62 Ci of ^{123}I at proton energy of 22 MeV, beam current of 30 μA if the ^{124}Te is irradiated over a period of 3 hours. Comparisons with some selected experimental data indicate that the much-lower-than-expected EOB yields are mainly due to incorrect target thickness prepared for the irradiation. Nevertheless these calculations are in good agreement with the previous predicted data with a maximum difference of less than 10%.

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