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CALCULATED RADIOACTIVITY YIELDS OF GALLIUM-67 USING MATLAB CODES

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ABSTRACT

CALCULATED RADIOACTIVITY YIELDS OF GALLIUM-67 USING MATLAB CODES. In nuclear medicine, gallium-67 (^{67}Ga) is potentially applied for imaging a certain type of tissue. In this investigation, ^{67}Ga is theoretically studied in terms of its potential radioactivity yields at the end of various energetic proton bombardments. Nuclear cross-sections derived from the Talys Evaluated Nuclear Data Library (TENDL) 2017 were used as the input files, while a Matlab code was developed to perform the yield calculations of ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga nuclear reactions to produce ^{67}Ga. Two different targets – enriched ^{67}Zn and ^{nat}Zn targets – were simulated in the calculations. The calculated yields suggested that a maximum of 27.37 MBq/µAh could be achieved when enriched ^{67}Zn target was irradiated with 15-MeV protons, whereas 46.99 MBq/µAh could be generated following a 30 MeV proton bombardment of enriched ^{68}Zn target. Various radioactive gallium impurities, i.e. ^{63,64,65,66,68,70}Ga and stable ^{69}Ga isotope were also expected to be generated mostly via (p,n) and (p,2n) reactions when ^{nat}Zn target was used in the ^{67}Ga production. In contrast, radioactive ^{66}Ga and ^{68}Ga impurities were mainly produced following bombardment of enriched ^{67}Zn and ^{68}Zn targets. This study can be used as a reference for future ^{67}Ga radionuclide production.

Keywords: Ga-67, Nuclear cross-section, Proton bombardment, Radioactivity yield, TENDL 2017

ABSTRAK

PERHITUNGAN YIELD RADIOAKTIVITAS GALLIUM-67 MENGGUNAKAN MATLAB. Dalam kedokteran nuklir, gallium-67 (67Ga) berpotensi untuk diaplikasikan dalam pencitraan jaringan tertentu yang ada di dalam tubuh. Dalam penelitian ini, 67Ga dipelajari secara teoritis terutama mengenai potensi yield radioaktivitasnya pada saat akhir iradiasi dengan berkas proton dengan berbaga energi. Tampang lintang reaksi nuklir yag diperoleh dari Talys Evaluated Nuclear Data Library (TENDL) 2017 digunakan sebagai data input, sedangkan software Matlab digunakan untuk menghitung yield reaksi nuklir 67Zn(p,n)67Ga dan 68Zn(p,2n)67Ga untuk memproduksi 67Ga. Dua target yang berbeda – 67Zn yang diperkaya and Zn alam (natZn) – disimulasikan dalam perhitungan tersebut. Hasil perhitungan menunjukkan bahwa proton dengan energi 15 MeV yang ditembakkan ke target diperkaya 67Zn dapat menghasilkan yield radioaktivitas sebesar 27,37 MBq/µAjam, sedangkan untuk energi proton sebesar 30 MeV dapat dihasilkan yield sebesar 46,99 MBq/µAjam. Berbagai impuritas radioaktif Ga, antara lain ^{63,64,65,66,68,70}Ga dan impuritas isotop stabil 69Ga dapat dhasilkan melalui reaksi nuklir (p,n) dan (p,2n) jika Zn alam digunakan di dalam produksi 67Ga. Sebaliknya, impuritas radioaktif 66Ga dan 68Ga diprediksikan dapat dihasilkan dari iradiasi target diperkaya 67Zn dan 68Zn. Hasil studi ini dapat dijadikan sebagai referensi untuk produksi radionuklida 67Ga di masa yang akan datang.

Kata kunci: Ga-67, Nuclear cross-section, Proton bombardment, Radioactivity yield, TENDL 2017

INTRODUCTION

Gallium-67 (^{67}Ga) is a gamma-emitting radioisotope with a half life of 3.3 days suitable as a diagnostic radioisotope for Single Photon Emission Computed Tomography (SPECT) modality or scintigraphy in nuclear medicine. Recent development suggests that ^{67}Ga is potentially employed to study a wide range of diseases, including for detection of Kaposi sarcoma lesions [1], Pulmonary Mycobacterium mucogenicum and Mycobacterium phocaicum Infection [2], osteomyelitis in the diabetic foot [3], Spondylodiscitis imaging [4] and some other diseases [5-10]. When ^{67}Ga is labeled to chemical compound such as citrate, it can be applied for early assessment of psoas muscle abscess [11]. Yet, ^{67}Ga could also be used to monitor therapeutic effectiveness in a patient with relapsing Polychondritis [12]. Furthermore, ^{67}Ga could even be employed as a therapeutic radioisotope as recently reported by Othman and co-workers [13], in which they discovered that ^{67}Ga could be as effective as ^{111}In radioisotope for therapy. Previous research suggested various types of chemical complexes which could be labelled with ^{67}Ga [14-16]; thus makes it more flexible to be used in nuclear medicine applications.

While potential use of ^{67}Ga has been well demonstrated elsewhere, its production requirements still need further attention to obtain much better radioactivity yield. Latest experimental cross-sections for ^{68}Zn(p,2n)^{67}Ga nuclear reaction has been published by Pupillo et al [17], which indicates the maximum excitation function around 20 MeV incident protons. The incoming ion beam can be generated using cyclotrons or accelerators which have also been applied for thin layer activation analysis [18] and adsorption studies [19-20] as well as ^{18}F radionuclide production [21-22].

Recent theoretical study using a Monte Carlo code (MCNPX code) [23] predicted that for incident proton energy range between 15 and 26 MeV, the expected radioactivity yield was 196.86 MBq/µAh for ^{68}Zn(p,2n)^{67}Ga nuclear reaction, whereas lower yield of 88.44 MBq/µAh was expectedly obtained for ^{67}Zn(p,n)^{67}Ga nuclear reaction. Theoretical and experimental data on the end-of-bombardment (EOB) yield of ^{67}Ga have been limited; thus further studies are required.

In this theoretical work, radioactivity yield of two different nuclear reactions relevant for ^{67}Ga production – ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga – are calculated using Matlab code, while the corresponding nuclear cross-sections are calculated using the TENDL 2017. In addition, various impurities are also predicted in ^{67}Ga production, which have not been reported elsewhere.

MATERIALS AND METHODS

In this present study, two different targets – natural zinc (^{nat}Zn) target and enriched zinc targets (^{68}Zn and ^{67}Zn targets) – were simulated in the calculations. The range of various energetic protons in the investigated targets were calculated using the Stopping and Range of Ions in Matters (SRIM) 2013 code, while the nuclear cross-sections for ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga and the other relevant nuclear reactions were calculated using the Talys Evaluated Nuclear Data Library (TENDL) 2017 [24 – 28]. In addition, a Matlab code was developed for calculations of the end-of-bombardment (EOB) yields of the investigated nuclear reactions using the stopping power data derived from the SRIM calculations and the excitation functions obtained from the TENDL 2017. The calculation procedures have been described elsewhere [29-32].

The computer hardware specifications used in these calculations are as follows:

Device name	LAPTOP-5A5RPLDL
Processor	Intel®Core™ i7-8750H CPU@ 2.20 GHz 2.21 GHz
Installed RAM	8.00 GB
Device ID	231CFEB0-2A05-4595-894C-6D0E8A44E99A
System type	64-bit operating system, x64-based processor

In order to study the dependence of proton beam dose on the radioactivity yield, the calculations were simulated for a fixed proton beam current of 50 µA while the irradiation time was varied from 12 to 120 minutes, creating proton doses ranging from 10 to 100 µAh. For these purposes, several proton energies (9 MeV, 11 MeV, 18 MeV, 26 MeV and 30 MeV) were simulated based on the currently available cyclotrons in Indonesia. In addition, in these calculations, Matlab code was used since it is relatively easy for a simple yield equation, it is a high-performance language for technical computing that integrates computation in an easy-to-use environment where problems and solutions are expressed in familiar mathematical notation.

RESULTS AND DISCUSSION

TALYS-Evaluated Nuclear Cross-sections

As mentioned in Section 2, two nuclear reactions are considered in this work, namely ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga; thus two enriched targets (^{67}Zn and ^{68}Zn targets) are studied. According to the SRIM 2013 calculations, 2.07 mm-thick ^{67}Zn target should be prepared for ^{67}Ga production when the target is bombarded with 30-MeV proton beam. However, a slightly thicker enriched ^{68}Zn target (2.10 mm thick) should be used in the ^{67}Ga production for irradiating it with 30-MeV protons.

Based on the TENDL 2017 evaluated excitation function of ^{67}Zn(p,n)^{67}Ga reaction the threshold energy is 1.98 MeV with a maximum nuclear cross-section of 563.64 mbarn at proton incident energy of 9 MeV as can be seen in Figure 1. In contrast, higher threshold energy (12.16 MeV) is required for ^{68}Zn(p,2n)^{67}Ga reaction, though its maximum nuclear cross-section is higher than that of ^{67}Zn(p,n)^{67}Ga reaction, which is 675.88 mbarn at proton energy of 18 MeV.

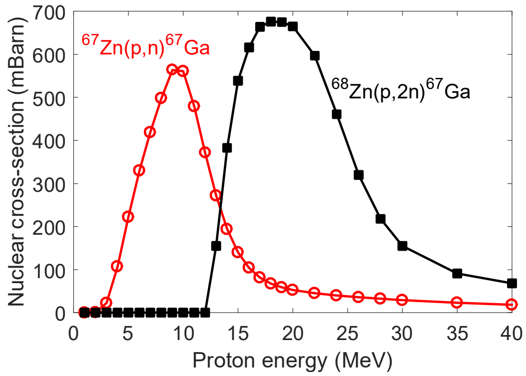

Figure 1. TENDL 2017 nuclear cross-section of ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga nuclear reactions.

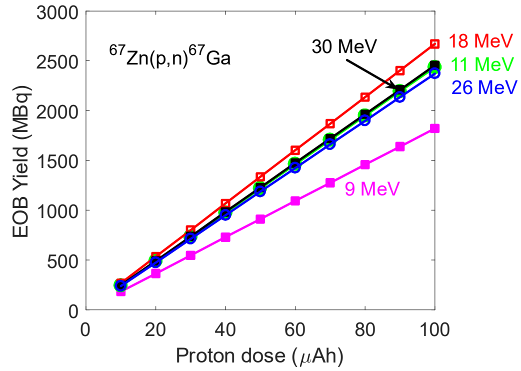

Figure 3. Calculated ^{67}Ga yields at selected proton doses and energies for ^{67}Zn(p,n)^{67}Ga nuclear reaction.

EOB Yields of ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga Nuclear Reactions

The proton energy dependence of ^{67}Ga radioactivity yield at the EOB for ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga nuclear reactions is shown in **Figure 2**. In the case of ^{67}Zn(p,n)^{67}Ga reaction, as low as 4.09 MBq/µAh could be produced when enriched ^{67}Zn target is irradiated with 5-MeV protons. The EOB yield increases with increasing proton energy and reaches its maximum yield of 27.37 MBq/µAh at 15-MeV protons. The yield saturates for proton incident energy of greater than 15 MeV.

Again from Figure 2, the EOB yield for ^{68}Zn(p,2n)^{67}Ga nuclear reaction remains low (as low as 4.18 MBq/µAh) when enriched ^{68}Zn target is bombarded with 15-MeV protons. However, the ^{67}Ga radioactivity yield increases dramatically to a maximum value of 46.99 MBq/µAh as the proton energy is increased to 30 MeV. The EOB yield is expected to level off for proton energy of greater than 30 MeV (not shown in Figure 2).

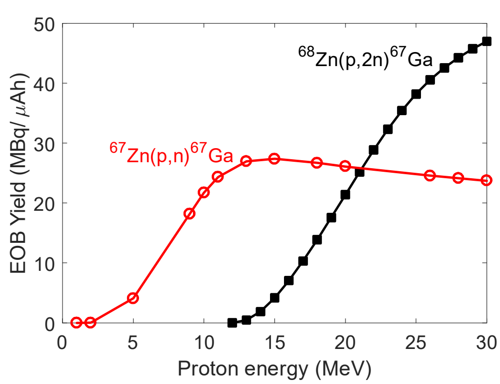

Figure 2. Calculated EOB yields of ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga nuclear reactions.

To achieve a high level of radioactivity yield applicable for scintigraphy in nuclear medicine, one should play around with the proton dose. For ^{67}Zn(p,n)^{67}Ga reaction, the expected EOB yields of various proton doses ranging from 10 to 100 µAh for 9 MeV, 11 MeV, 18 MeV, 26 MeV and 30 MeV protons can be found in Figure 3. When ^{67}Zn target is irradiated with 9 MeV-protons at a proton dose of 100 µAh, the EOB yield could reach up to 1820.9 MBq which would be sufficient for diagnosing 10 patients. At the same irradiation parameters, for incident protons of 18 MeV, the expected EOB yield is 2669 MBq, which could be used for diagnosing up to 14 patients. Moreover, there is no significant different in the EOB yield when ^{67}Zn target is bombarded with 11 MeV, 26 MeV and 30 MeV protons, in which around 2400 MBq is resulted from ^{67}Ga production at 100 µAh proton dose.

For ^{68}Zn(p,2n)^{67}Ga reaction, the calculated EOB yields of various proton doses ranging from 10 to 100 µAh for 18 MeV, 26 MeV and 30 MeV protons are shown in Figure 4. Note that there would be no radioactivity yield resulted from proton bombardment of ^{68}Zn target when the incident proton energy is lower than 12 MeV. When 18 MeV, 26 MeV and 30 MeV protons are bombarded into ^{68}Zn target at 100 µAh proton dose, the expected radioactivity yields are 1387.5 MBq, 4055.2 MBq and 4699 MBq respectively, which would be enough to diagnose 7, 21 and 25 patients respectively.

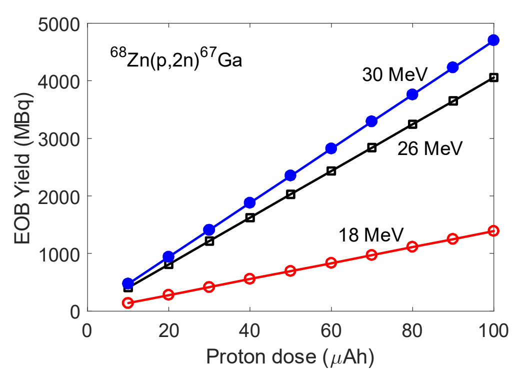

Figure 4. Calculated ^{68}Ge yields at selected proton doses and energies for ^{68}Zn(p,2n)^{67}Ga nuclear reaction.

Predicted Impurities

Possible impurities in cyclotron-based radionuclide production can be predicted from their corresponding nuclear reactions and cross-sections. For enriched ^{67}Zn target, the expected impurity is ^{66}Ga radionuclide which is due to $^{67}Zn(p,2n)^{66}Ga$ nuclear reaction. The ^{66}Ga impurity emits positron with a half life of 9.49 hours. As can be seen in Figure 5, the proton threshold energy for $^{67}Zn(p,2n)^{66}Ga$ nuclear reaction is 13.21 MeV; thus no ^{66}Ga impurity would be generated at proton energy lower than 13 MeV.

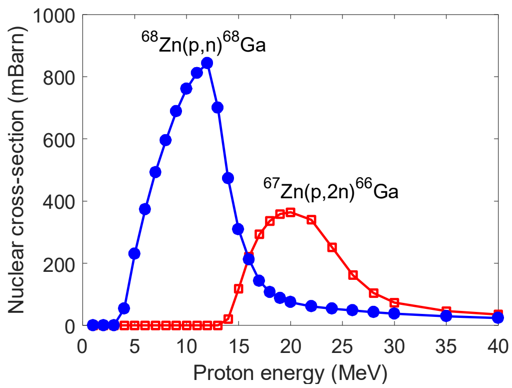

Figure 5. TENDL 2017 excitation functions of $^{67}Zn(p,2n)^{66}Ga$ and $^{68}Zn(p,n)^{68}Ga$ nuclear reactions.

For enriched ^{68}Zn target, ^{68}Ga radionuclide is expected to be an impurity as a result of $^{68}Zn(p,n)^{68}Ga$ nuclear reaction. Similar to ^{66}Ga impurity, ^{68}Ga impurity also emits positron, though the half life is much shorter (67.71 minutes). Again, as shown in Figure 5, the proton threshold energy for $^{68}Zn(p,n)^{68}Ga$ nuclear reaction is 3.94 MeV; therefore ^{68}Ga impurity would always be generated when producing ^{67}Ga through $^{68}Zn(p,2n)^{67}Ga$ nuclear reaction.

When the target of interest is natural zinc (^{nat}Zn) containing several Zn atoms such as ^{64}Zn, ^{66}Zn, ^{67}Zn, ^{68}Zn and ^{70}Zn, there would be several nuclear reactions involved in the ^{67}Ga production, which eventually create various impurities. The TENDL 2017 nuclear cross-sections for ^{64}Zn, ^{66}Zn, ^{68}Zn and ^{70}Zn following (p,n) and (p,2n) nuclear reactions are depicted in Figure 6. The lowest threshold energy (1.97 MeV) is predicted for $^{70}Zn(p,n)^{70}Ga$ reaction while the highest threshold energy (18.60 MeV) is expected for $^{64}Zn(p,2n)^{63}Ga$ reaction.

According to the EOB yield calculations, a maximum of 8.7 MBq/µAh radioactivity yield is produced

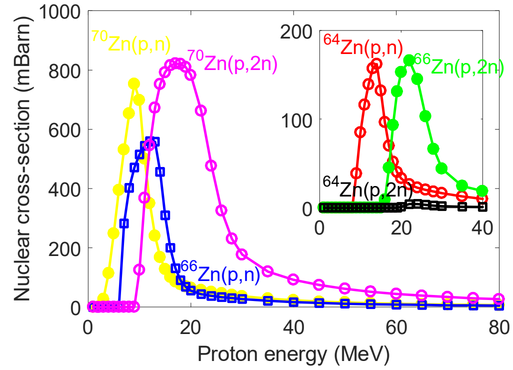

Figure 6. TENDL 2017 excitation functions of proton-irradiated natZn target.

when ^{nat}Zn is irradiated with 15-MeV protons, 18-MeV protons and 30-MeV protons respectively. While the EOB yield is relatively high and sufficient for diagnosing several patients, deep attention should be taken care of particularly dealing with impurities such as $^{63,64,65,66,68,70}Ga$ and stable isotope ^{69}Ga as listed in Table 1. Most impurities would be positron emitting radionuclides with half lives ranging from as short as 32.4 seconds to 3.26 days as a result of (p,n) and (p,2n) nuclear reactions.

Comparison with Published Work

There have been a limited number of publications related to ^{67}Ga production yield, though available experimental work and theoretical calculations can be collected for comparisons. As seen in Table 2, this calculated work closely agrees with semi-experimental data [30-31] as well as the MCNPX calculation [20] with differences ranging from 0.13% to 13.66% for proton

Table 2. Comparison of calculated and semi-experimental ^{67}Ga yields from $^{68}Zn(p,2n)^{67}Ga$ nuclear reaction.

E_p range (MeV)	Radioactivity yield (MBq/µAh)			
	This calculated work	Semi-experiment [33]	MCNPX calculation [23]	Semi-experiment [34]
25-14	207.93	208.2	193.95	193.14
26-15	209.67	222	196.86	204.61
26-18	153.68	178	146.86	162.8

Table 1. Various impurities predicted during production of 67Ga radionuclide

Isotopes of Zn	Natural abundance (%)	Nuclear reaction	Threshold energy (MeV)	Decay mode	Half life
^{64}Zn	49.2	$^{64}Zn(p,n)^{64}Ga$	8.12	Positron	2.627 min
		$^{64}Zn(p,2n)^{63}Ga$	18.60	positron	32.4 s
^{66}Zn	27.7	$^{66}Zn(p,n)^{66}Ga$	6.09	Positron	9.49 h
		$^{66}Zn(p,2n)^{65}Ga$	15.33	positron	15.2 min
^{67}Zn	4.0	$^{67}Zn(p,n)^{67}Ga$	1.98	EC	3.26 d
		$^{67}Zn(p,2n)^{66}Ga$	13.21	positron	9.49 h
^{68}Zn	18.5	$^{68}Zn(p,n)^{68}Ga$	3.94	Positron	67.71 min
		$^{68}Zn(p,2n)^{67}Ga$	12.16	EC	3.26 d
^{70}Zn	0.6	$^{70}Zn(p,n)^{70}Ga$	1.97	Beta	21.14 min
		$^{70}Zn(p,2n)^{69}Ga$	9.22	stable	-

energy range between 26 and 14 MeV. In the semi-experimental work by Takács *et al* [30] and Szelecsényi *et al* [31], thick target yields were calculated from their experimental nuclear cross-sections. Therefore, further experimental investigations are required for better comparisons.

In Indonesia, currently there are three cyclotrons which accelerate protons up to 9 MeV in Gading Pluit Hospital, Jakarta, 11 MeV in Dharmais Cancer Hospital, Jakarta and 18 MeV in Siloam Hospital, Jakarta. Based on the calculated nuclear cross-sections and threshold energies for ^{67}Ga production, the three available cyclotrons can be used to generate ^{67}Ga radionuclide via Zn(p,n)^{67}Ga nuclear reaction. In contrast, only Siloam Hospital's cyclotron is capable of producing ^{67}Ga radionuclide via ^{68}Zn(p,2n)^{67}Ga nuclear reaction since the threshold energy is 11.98 MeV. In addition, predicted impurities also greatly depend on the cyclotron employed in ^{67}Ga production. Using Gading Pluit cyclotron, three radionuclides, such as ^{70}Ga, ^{68}Ga and ^{64}Ga may contribute to the presence of impurities, whereas two more isotopes, such as ^{64}Ga and ^{69}Ga could be present in ^{67}Ga production using Dharmais Hospital's cyclotron. Furthermore, all listed isotopes in **Table 1** could become impurities when ^{67}Ga is produced using Siloam Hospital's cyclotron.

CONCLUSION

Production yields of ^{67}Ga radionuclide have been calculated using a developed Matlab code for ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga nuclear reactions. In the EOB yield calculations, the stopping powers were derived from the SRIM-2013 calculations while the nuclear cross-sections were derived using the TENDL 2017. The TENDL 2017 excitation functions indicate that the threshold energy is 1.98 MeV with a maximum nuclear cross-section of 563.64 mbarn at proton incident energy of 9 MeV for ^{67}Zn(p,n)^{67}Ga reaction, whereas for ^{68}Zn(p,2n)^{67}Ga reaction the threshold energy is 12.16 MeV with maximum nuclear cross-section of 675.88 mbarn at proton energy of 18 MeV. The expected maximum EOB yields of ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga reactions are 27.37 MBq/µAh at 15-MeV protons and 46.99 MBq/µAh at 30 MeV protons, respectively. Based on the calculated nuclear cross-sections, various impurities could be generated following bombardment of enriched ^{67}Zn, enriched ^{68}Zn and ^{nat}Zn targets. Two impurities (^{66}Ga and ^{68}Ga) are predicted to be produced when protons hit enriched ^{67}Zn and ^{68}Zn targets. On the other hand, ^{63,64,65,66,68,70}Ga and stable ^{69}Ga isotope are expected to be generated mostly via (p,n) and (p,2n) reactions when ^{nat}Zn target is used in the ^{67}Ga production. Comparisons with other theoretical and semi-experimental work indicate that this work is in a good agreement with other published works, though further experimental work is needed for better comparisons. In addition, all available cyclotrons in Indonesia can be used to produce ^{67}Ga, though the impurities present during the production depend on the type of cyclotron.

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