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Production cross sections of $^{nat}Zn(\alpha,x)^{68,69}Ge,^{66,67}Ga,^{65}Zn$ reactions up to 46.3 MeV

Sk Wasim Raja[1,3,a], **R. Acharya**[2,3,b], **S. Dey Chaudhuri**[1]

[1] Radiochemistry Division (BARC), Variable Energy Cyclotron Centre, 1/AF, Bidhan Nagar, Kolkata 700064, India
[2] Radiochemistry Division, Bhabha Atomic Research Centre, Mumbai 400085, India
[3] Homi Bhabha National Institute, Anushaktinagar, Mumbai 400094, India

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Abstract Three medicinally important radioisotopes of Ga, $^{66,67,68}Ga$, having half-lives ranging from minutes to days, could be produced from $^{nat}Zn(\alpha,x)$ reactions. They have different applications in diagnosis and therapy in nuclear medicine depending on their decay properties and half-lives. ^{68}Ge ($T_{1/2}$ = 270.95 days) produced from $^{nat}Zn(\alpha,x)$ reactions decays to ^{68}Ga ($T_{1/2}$ = 67.71 min) via electron capture (100%). So ^{68}Ga can be produced for its practical application in medicine using $^{68}Ge/^{68}Ga$ generator where in-house medical cyclotron facilities are unavailable. Production and radiochemical separation of a particular radioisotope from natural zinc target with minimum contamination from other isotopes require accurate activation cross section data of all the produced radionuclides. Activation cross sections of long-lived radioisotopes of $^{68,69}Ge$, $^{66,67}Ga$ and ^{65}Zn produced from alpha irradiation of natural Zn targets were measured from their respective threshold up to 46.3 MeV. Using the experimentally measured cross section data and stopping power of natural Zn, thick target yields were calculated for these medicinally important radionuclides. The experimentally measured cross section data were compared with the earlier published literature data and theoretical data from TENDL-2021 nuclear data library based on the TALYS-1.96 code. Cross section data of the present work are found to be in good agreement with the literature data within experimental uncertainties for ^{68}Ge, ^{66}Ga and ^{65}Zn while there are significant differences for ^{67}Ga and ^{69}Ge.

1 Introduction

Medicinally important radionuclides for positron emission tomography (PET) applications are generally routinely produced using light charged particle beams (proton, deuteron and alpha) from medical cyclotrons located close to the hospitals across the globe. Positron emitting radionuclides such as ^{18}F ($T_{1/2}$ = 1.83 h), ^{11}C ($T_{1/2}$ = 20.4 min), ^{13}N ($T_{1/2}$ = 9.96 min) etc. are produced via proton irradiation and ^{15}O ($T_{1/2}$ = 2.05 min) is produced in deuteron induced reaction in cyclotron facilities near to hospitals [1]. As an alternate to this, radionuclide generators could be used for production of medicinally important radioisotopes in house without cyclotrons [2]. Here the parent nuclide of long half-life decays to short half-life daughter nuclide and the daughter nuclide is chemically separated from the parent for their subsequent uses. Though reactor produced $^{99}Mo/^{99m}Tc$ generator is very common, cyclotron produced $^{68}Ge/^{68}Ga$ generator is gaining interest in recent days [3].

Three isotopes of Ga, $^{66,67,68}Ga$, could be produced from alpha induced reactions on natural Zn targets. ^{66}Ga ($T_{1/2}$ = 9.49 h), because of its long half-life and suitable decay data, is used as a tracer to study time taking biological processes and can be a suitable alternate in PET in hospitals for therapeutic uses [4]. ^{67}Ga ($T_{1/2}$ = 3.2617 days) is used in single-photon emission computed tomography (SPECT) imaging in diagnosis of cancers, inflammatory and/or infectious diseases in clinical practice. As an alternate of ^{125}I, ^{67}Ga labeled compounds are used for cancer therapy [5]. ^{68}Ga ($T_{1/2}$ = 67.71 min) is useful for PET imaging of prostate cancer and it is a rapidly growing PET nuclide for cancer diagnostics [6]. The different radioisotopes of Ga have different diagnostic and therapeutic applications owing to their different decay properties, particularly half-lives ranging from minutes to days. So production and radiochemical separation of

[a] e-mail: sw.raja@vecc.gov.in(corresponding author)
[b] e-mail: racharya@barc.gov.in(corresponding author)

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a particular radioisotope with minimum contamination from other isotopes is often necessary [7]. Long-lived ^{69}Ge ($T_{1/2}$ = 270.95 days) and ^{65}Zn ($T_{1/2}$ = 243.93 days) radio nuclides are also produced in alpha induced reactions on natural Zn. ^{65}Zn is used for labeling anticancer agent and brain tumor imaging [8]. Hence, accurate activation cross section data of all the produced radionuclides are also often necessary for optimization of the production of medicinally important nuclide of choice from natural zinc target.

^{68}Ga can be used in practical purpose only using in-house cyclotrons. Due to comparatively short half-life of 67.71 min of ^{68}Ga, its use in medicine is limited where medical cyclotron facility is unavailable on site. However, it can be produced from its long-lived parent ^{68}Ge ($T_{1/2}$ = 270.95 d). High activity ^{68}Ge could be produced using proton and alpha beams from cyclotron via ^{69}Ga(p,2n)^{68}Ge and ^{66}Zn(α,2n)^{68}Ge reactions [9]. So ^{68}Ge/^{68}Ga generator could be an alternate for its production and application in medicine where in-house medical cyclotron facilities are unavailable. On demand separation of ^{68}Ga can be done economically in no-carrier added form with high specific activity from this generator system for a long period of ~ 1 year or more [10]. ^{68}Ga could be directly produced in a cyclotron from proton bombardment of ^{68}Zn via ^{68}Zn(p,n)^{68}Ga reaction. 15 MeV proton beam was found suitable for production of high activity ^{68}Ga with minimum radionuclide impurities [11]. ^{68}Ge/^{68}Ga generators are preferred over cyclotron production because of its comparatively lower cost, availability, simple and fast method to prepare radiopharmaceuticals by extracting ^{68}Ga and labeling with suitable biomolecules.

Accurate cross section data are required to optimize the production of these medicinally important radionuclides with respect to suitable target-projectile combination, beam energy range and production of other nuclides for their easy radiochemical separation. There are only few discrete measurements of activation cross sections in alpha induced reaction on natural zinc available in literature [12–15]. The produced radionuclides from ^{nat}Zn(α,x) reactions have important applications in medicine and industry. Nagame et al. [12] irradiated natural Zn with 40 MeV alpha beams to determine the optimum conditions for the production of medicinally important ^{67}Ga. They had also measured formation cross sections of radionuclides ^{63,65}Zn, $^{65,66.67,68}$Ga and ^{66,67,68,69}Ge having half-lives from few minutes to days in the energy range of 10 to 40 MeV using stacked foil activation technique and measured the gamma activity without chemical separation. Aikawa et al. [13] recently measured production cross sections of ^{68,69}Ge and ^{66,67}Ga from ^{nat}Zn(α,x) reactions up to 50.7 MeV.

Systematic study on alpha induced reaction on natural Zn is scarce. In the present work, we have studied in detail the production cross sections of long-lived radionuclides produced in the alpha induced reactions on natural Zn from their respective threshold up to 46.3 MeV. Three stacks of natural Zn foils were irradiated with initial alpha beam energies of 40 and 35 MeV at K = 130 AVF cyclotron at Variable Energy Cyclotron Centre, Kolkata, India. Long-lived nuclides of ^{68,69}Ge, ^{66,67}Ga and ^{65}Zn were identified in the measured gamma ray spectra using HPGe detector. Activation cross sections of these radionuclides were measured and thick target yields were calculated from the measured cross section data. The data of the present work were compared with the earlier published literature data [12–16] and theoretical data based on TALYS-1.96 code [17] obtained from TENDL-2021 nuclear [18] data library. The experimentally measured cross section data of ^{68,69}Ge, ^{66,67}Ga and ^{65}Zn from ^{nat}Zn(α,x) reactions reported in the present work will be useful to optimize their production and calculate the contamination from other produced radioisotopes.

2 Experimental details

The experiment was performed using alpha beam from K = 130 AVF cyclotron at Variable Energy Cyclotron Centre (VECC), Kolkata, India. Standard stacked foil activation method was used for irradiation and induced activity was measured offline using HPGe detector based high resolution gamma ray spectrometry to measure the cross sections of long-lived radionuclides produced in alpha induced reactions on natural zinc (^{nat}Zn(α,x) ^{68,69}Ge, ^{66,67}Ga, ^{65}Zn) from their respective threshold up to 46.3 MeV. Experimental details are described in the following subsections.

2.1 Targets and irradiations

The targets were high purity natural Zn foils (thickness: 50 μm, purity: 99.9%). Zn has five naturally occurring isotopes ^{64}Zn (48.63%), ^{66}Zn (27.90%), ^{67}Zn (4.10%), ^{68}Zn (18.75%) and ^{70}Zn (0.62%) [19]. High purity Al (18 μm thickness) and Cu foils (25.4 μm thickness) were used as catcher foils and beam current monitors, respectively. A large sheet of Zn, Al and Cu foils were cut into 12 × 12 mm^2 size individual pieces. In the stacks, there was one Al catcher foil after each Zn foil and in each stack; one Cu foil was inserted to monitor beam current. There were four zinc targets in each stack. A typical stack containing target-catcher foil arrangements is shown in Fig. 1. The foils were screwed on the target holder. The target holder containing the stack was placed in the cyclotron beam line and irradiated with incident alpha beam energy of 48 and 35 MeV. Three different stacks were irradiated each for 1.0 h using 48 and 35 MeV initial alpha beam from cyclotron of current ~ 71, 61 and ~ 30 nA, respectively. 6 mm diameter collimated alpha beam was used. During irradiation, target holder containing the stack was cooled with water circulation in a closed loop. Using

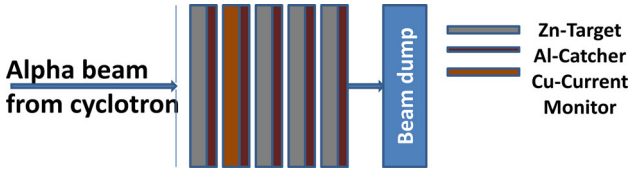

Fig. 1 A typical representation of a stack used for irradiation. After each Zn target, one Al catcher foil was kept and Cu foil was used to monitor beam intensity

SRIM 2013 code, beam energy degradation in the foils of the stack was calculated [20]. The average energy loss in the target foils varied from 3.4 to 4.6 MeV for the stacks irradiated with 48 MeV alpha beams and 4.2–8.9 MeV for the stack irradiated with 35 MeV alpha beams. Average energy in the middle of the target foil was taken as the Zn targets were 50 μm thick. The uncertainties in energy varied from 0.5 to 1.3 MeV depending on the position of the targets in the different stacks considering uncertainties of 0.5% in initial alpha beam energy, ~ 1% in target thickness and beam energy straggling.

2.2 Data acquisition

Sufficient cooling time was given after the irradiation to allow decaying of short-lived reaction products. When radiation doses were in the permissible limit, the target holder was removed from the cyclotron beam line, transferred safely in the radiochemistry laboratory and kept inside fume hood. It was then opened, the target with associated catcher foil were individually separated and packed in small polythene pouch and mounted on Perspex plate for counting the gamma activity. The counting was started after providing sufficient cooling time for the different radionuclides from the end of irradiation. Initially, targets were placed at a distance of 20–25 cm for counting to maintain point like geometry of the sample to the detector and minimize pulse pile up effect. Later, after a cooling period of ~ 120 days, targets were placed close to the detector at a distance of 3 cm for counting the long-lived radioisotopes. The dead time of the detector was always kept below 5%. Each target was counted several times to follow the decay curve and to identify interfering radionuclides and sufficient counts were collected for each gamma ray peaks of interest to reduce statistical uncertainty. The counting was done for more than 6 months. Typical gamma ray spectrum of Zn target irradiated with 46.3 MeV alpha beams and acquired after 30 h and 120 days of cooling is shown in Fig. 2. The activity was measured using 50% relative efficiency HPGe detector coupled to PC based 4 k MCA. It had energy resolution of 1.8 keV at 1332 keV of ^{60}Co. Biasing the detector, signal processing and data collection were done by DSA-1000 (Canberra Digital System). GENIE 2000 software was used for data analysis. ^{152}Eu and ^{133}Ba

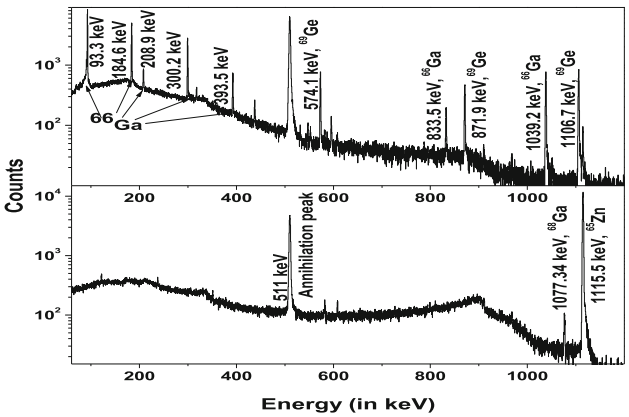

Fig. 2 A typical gamma ray spectra of Zn target irradiated with 46.3 MeV alpha beams and measured after 30 h (above) and 120 days (below) of cooling period. Gamma ray peaks of the studied radionuclides are marked in the spectra

point sources were used for energy and efficiency calibrations of the HPGe detector. Efficiencies (ε) of the interested gamma rays (E_γ) were determined using fifth order polynomial fitting of $\ln(\varepsilon) = \sum_{j=0}^{j=5} a_j (ln(E_\gamma))^j$ where a_j are fitting coefficients.

2.3 Calculations of cross sections

Cross sections (σ(E)) were calculated using the following well known activation equation [21]:

$$\sigma(E) = \frac{\lambda C(E)}{\varepsilon_\gamma I_\gamma t \rho \varphi \left(1 - e^{-\lambda t_{irr}}\right) e^{-\lambda t_{cool}} \left(1 - e^{-\lambda t_{count}}\right)}$$

where, C (E) is the net counts under the photo peak of the radionuclide, λ is the decay constant of the radioisotope (s^{-1}), I_γ and ε_γ are the absolute intensity and absolute efficiency of the detector for the detected gamma ray, t is the target foil thickness (cm), ρ is the atomic density of the target (cm^{-3}), φ is the beam intensity (s^{-1}), and $t_{irr}, t_{cool}, t_{count}$ are irradiation, cooling and measurement time (s), respectively.

The nuclear spectroscopic data like decay mode, half-life, gamma ray energy and intensity of the studied radio nuclides along with their production routes, Q values and threshold energies (E_{th}) are given in Table 1. They were taken from the ENSDF evaluation obtained from the NuDat 3.0 database [22]. Q-tool [23] based on atomic mass evaluation by Meng et al. [24] was used to calculate Q-values and threshold energies (E_{th}). Activity of ^{67}Ga formed in the alpha induced reaction on natural Cu (^{nat}Cu(α,x) ^{67}Ga, beam monitor reaction), which was present at different location of the stacks, was measured to determine alpha beam intensity. IAEA recommended cross-sections of σ = 297.4 mb at E_α = 28.6 MeV, σ = 27.1 mb at E_α = 46.9 MeV and σ = 297.4 mb at E_α = 28.4 MeV for the three stacks irradiated with 48 and 35 MeV

Table 1 Decay data and reaction parameters of the studied long-lived radio nuclides produced from $^{nat}Zn(\alpha,x)$ reactions

Nuclide	Half-life ($T_{1/2}$)	Decay Mode (%)	E_γ (keV)	I_γ (%)	Contributing Reactions	Q-Value (MeV)	Threshold, E_{th} (MeV)
^{69}Ge	39.05 (10) h	EC + β^+ (100)	574.11(10)	13.3(18)	$^{66}Zn(\alpha,n)$	− 7.4	7.9
			871.98 (10)	11.9(16)	$^{67}Zn(\alpha,2n)$	− 14.5	15.4
			1106.77(10)	**36**	$^{68}Zn(\alpha,3n)$	− 24.7	26.2
					$^{70}Zn(\alpha,5n)$	− 40.4	42.7
					IT decay (100%) of ^{69m2}Ge ($T_{1/2}$ = 2.81 μs) and ^{69m1}Ge ($T_{1/2}$ = 5.1 μs)		
^{68}Ge	270.95 (16) d	EC (100)	–	–	$^{64}Zn(\alpha,\gamma)$	3.4	
					$^{66}Zn(\alpha,2n)$	− 15.6	16.6
					$^{67}Zn(\alpha,3n)$	− 22.7	24.0
					$^{68}Zn(\alpha,4n)$	− 32.9	34.8
					$^{70}Zn(\alpha,6n)$	− 48.6	51.4
^{68}Ga	67.71 (8) min	EC + β^+ (100)	**1077.34 (5)**	**3.22**	^{68}Ge (EC)		
^{67}Ga	3.2617 (5) d	EC (100)	93.310 (5)	38.81(3)	$^{64}Zn(\alpha,p)$	− 4.0	4.2
			184.576(10)	21.410(10)	$^{66}Zn(\alpha,t)$	− 14.5	15.4
			300.217 (10)	**16.64(12)**	$^{67}Zn(\alpha,tn)$	− 21.6	22.9
			393.527 (10)	4.56 (24)	$^{68}Zn(\alpha,t2n)$	− 31.8	33.7
					$^{70}Zn(\alpha,t4n)$	− 47.5	50.2
					EC and β^+ decay (100%) of ^{67}Ge ($T_{1/2}$ = 18.9 min)		
^{66}Ga	9.49 (3) h	EC + β^+ (100)	833.5324 (21)	5.9 (3)	$^{64}Zn(\alpha,d)$	− 13.0	13.8
			1039.220 (3)	**37.0(20)**	$^{66}Zn(\alpha,tn)$	− 25.8	27.3
					$^{67}Zn(\alpha,t2n)$	− 32.8	34.8
					$^{68}Zn(\alpha,t3n)$	− 43.0	45.6
					EC + β^+ decay (100%) of ^{66}Ge ($T_{1/2}$ = 2.26 h)		
^{65}Zn	243.93 (9) d	EC + β^+ (100)	**1115.539 (2)**	**50.04(10)**	$^{64}Zn(\alpha,2pn)$	− 20.3	21.6
					$^{66}Zn(\alpha,2p3n)$	− 39.3	41.7
					$^{67}Zn(\alpha,2p4n)$	− 46.4	49.2
					$^{68}Zn(\alpha,2p5n)$	− 56.6	59.9
					EC + β^+ decay (100%) of ^{65}Ga ($T_{1/2}$ = 15.2 min)		

Gamma rays marked in bold were used in cross section calculations

Table 2 Measured cross section data for ^{68,69}Ge, ^{66,67}Ga and ^{65}Zn radio nuclides produced from ^{nat}Zn(α,x) reactions

Energy (MeV)	Cross sections (mb)				
	^{68}Ge	^{69}Ge	^{66}Ga	^{67}Ga	^{65}Zn
46.3 ± 0.7	46.3 ± 3.8	74.0 ± 5.5	100.1 ± 9.2	209.2 ± 15.9	415.9 ± 30.3
43.5 ± 1.0	57.3 ± 4.7	70.6 ± 5.2	135.2 ± 12.3	180.6 ± 13.3	344.8 ± 25.1
42.2 ± 0.9	64.2 ± 5.2	73.6 ± 5.5	172.2 ± 15.8	165.6 ± 12.2	341.9 ± 24.9
39.2 ± 1.1	80.8 ± 6.5	64.9 ± 4.8	232.4 ± 21.3	112.6 ± 8.4	234.2 ± 17.1
37.8 ± 1.1	111.4 ± 8.5	59.8 ± 4.4	285.8 ± 26.1	90.9 ± 6.7	193.8 ± 14.1
34.5 ± 1.2	127.9 ± 9.6	43.4 ± 3.3	318.3 ± 29.1	52.6 ± 3.9	89.9 ± 6.6
33.1 ± 1.2	141.8 ± 10.6	39.4 ± 2.9	337.8 ± 30.8	54.9 ± 4.1	66.6 ± 4.9
32.9 ± 0.5	163.3 ± 12.8	51.7 ± 4.0	424.1 ± 38.9	62.6 ± 4.7	101.2 ± 7.4
29.4 ± 1.3	111.5 ± 8.4	50.7 ± 3.8	267.0 ± 24.5	114.4 ± 8.5	30.6 ± 2.2
23.4 ± 0.7	58.5 ± 5.3	144.2 ± 10.7	102.0 ± 9.6	392.9 ± 29.0	9.4 ± 0.6
16.5 ± 0.8		93.6 ± 6.9		239.6 ± 17.6	
11.8 ± 0.8		11.6 ± 1.1		34.3 ± 2.5	

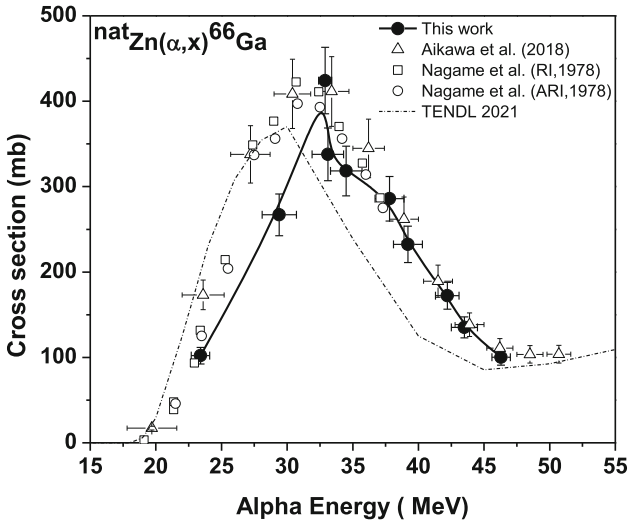

Fig. 3 Excitation function of ^{nat}Zn(α,x)^{66}Ga reaction and comparison with literature data and TENDL-2021 data

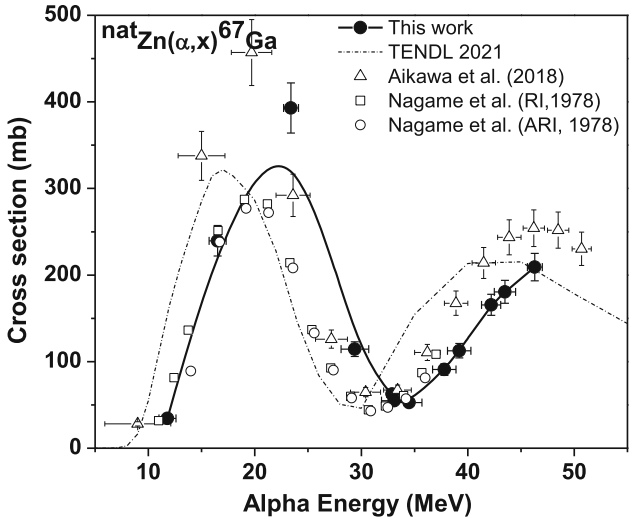

Fig. 4 Excitation function of ^{nat}Zn(α,x)^{67}Ga reaction and comparison with literature data and TENDL-2021 data

initial alpha beam energies for the beam monitor reaction ^{nat}Cu(α,x)^{67}Ga were used for beam intensity determinations [25].

3 Results and discussion

Experimentally measured production cross sections along with total uncertainties for the long-lived radionuclides ^{68,69}Ge, ^{66,67}Ga and ^{65}Zn produced from ^{nat}Zn(α,x) reactions from their respective threshold up to 46.3 MeV are given in Table 2. They are shown in Figs. 3, 4, 5, 6, 7 along with earlier published data from literature and theoretical data from TENDL-2021 nuclear data library [18] based on the TALYS-1.96 code [17] without adjusting the model parameters for comparison purpose. Our data are spline fitted to guide the eye in the spectra. The thick target integral yields of all these radionuclides were also calculated from the experimentally measured cross section data, compared with the literature data whenever available and presented in Figs. 8, 9. The cross section and yield data of the earlier published work used for comparison purpose with the present data were taken from EXFOR [26]. Cross sections were calculated using the standard activation equation. High intensity gamma rays free from any interference (marked bold in Table 1) were used to calculate cross sections. The total uncertainty in the measured cross sections was in the range of 7.5–9.5%. It was

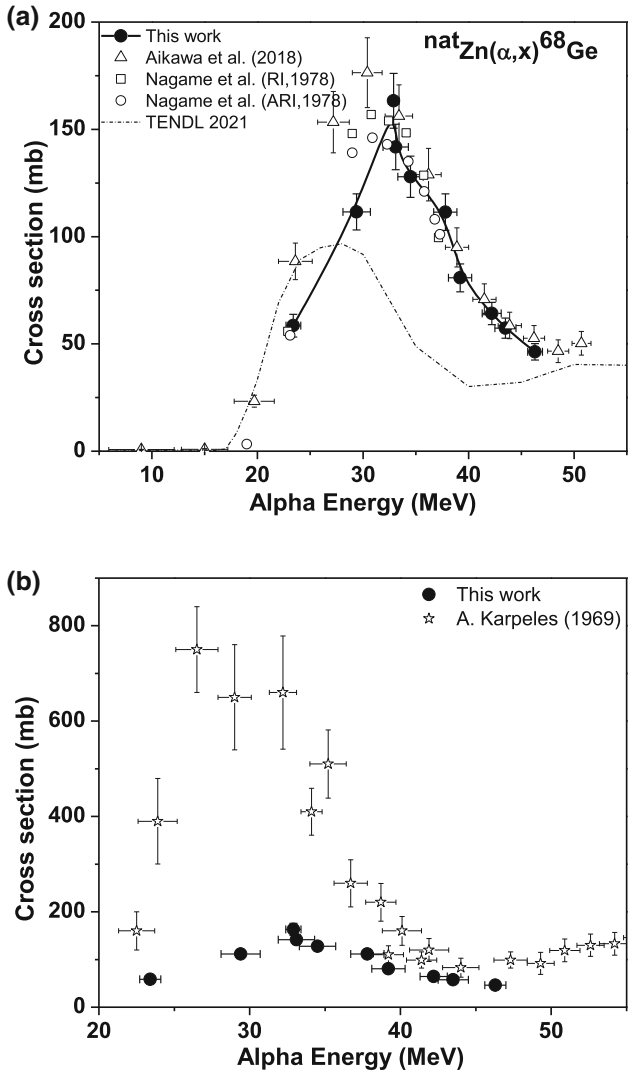

Fig. 5 **a** Excitation function of $^{nat}Zn(\alpha,x)^{68}Ge$ reaction and comparison with literature data and TENDL-2021 data. **b** Excitation function of $^{nat}Zn(\alpha,x)^{68}Ge$ reaction. Present data are compared with data reported by A. Karpeles [15]

calculated according to the error propagation rule by taking square root of quadratic summation of independent uncertainty contributions from thickness of the foil (1%), detector efficiency (4%), alpha beam intensity (6%), gamma ray intensity and counting statistics of the gamma rays. Uncertainties in gamma ray intensities were almost negligible (< 0.7%) for all the studied radionuclides except ^{66}Ga for which it is 5.4% and statistical uncertainties (%) were in the range of 0.8–2.0, 0.9–1.5, 1.3–2.7, 1.6–5.5 and 0.3–0.9 for 1039.2, 300.2, 1077.3, 1106.77 and 1115.5 keV of ^{66}Ga, ^{67}Ga, ^{68}Ge, ^{69}Ge and ^{65}Zn, respectively. Individual reactions are separately discussed in the following subsections.

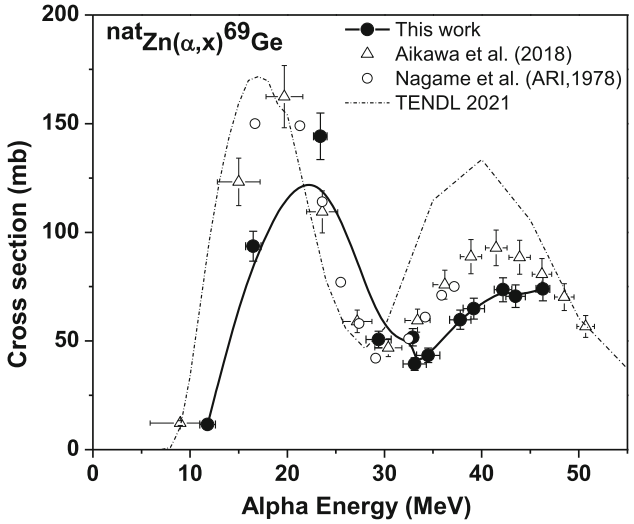

Fig. 6 Excitation function of $^{nat}Zn(\alpha,x)^{69}Ge$ reaction and comparison with literature data and TENDL-2021 data

3.1 Cumulative production cross sections of $^{nat}Zn(\alpha,x)^{66}Ga$

^{66}Ga ($T_{1/2} = 9.49$ h) is produced from ^{nat}Zn directly by the reactions listed in Table 1 and electron capture and β^+ decay (100%) of ^{66}Ge ($T_{1/2} = 2.26$ h). ^{66}Ga decays to stable ^{66}Zn by EC and β^+ processes (100%) with the emission of several gamma rays. As counting of 1039.220 keV of ^{66}Ga was done after a cooling period of 30 h, the measured cross sections are cumulative cross sections of ^{66}Ga. The experimentally measured cross section data along with earlier published data [12–14] and TENDL-2021 [18] data are shown in Fig. 3 and the comparison shows that present data are in good agreement in shape with these data. However, our data are lower in magnitude below 32 MeV and above this energy the amplitudes of our cross section data match well with the published data.

3.2 Cumulative production cross sections of $^{nat}Zn(\alpha,x)^{67}Ga$

^{67}Ga ($T_{1/2} = 3.2617$ d) is produced from ^{nat}Zn by direct reactions as listed in Table 1 and from the electron capture and β^+ decay (100%) of ^{67}Ge ($T_{1/2} = 18.9$ min) which is produced by $^{64}Zn(\alpha,n)$, $^{66}Zn(\alpha,3n)$, $^{67}Zn(\alpha,4n)$ and $^{68}Zn(\alpha,5n)$ reactions having thresholds of 9.6, 29.7, 37.2 and 47.9 MeV, respectively. As ^{67}Ge completely decays to ^{67}Ga when counting of ^{67}Ga activity was done after a cooling period of 3 days, the measured cross section is cumulative cross section. ^{67}Ga decays to stable ^{67}Zn by EC (100%) process with the emission of several gamma lines. 300.2 keV (16.64%) of ^{67}Ga was used for cross section calculations. The measured cross section data were compared with published literature data [12–14] along with TENDL-2021 data [18] as shown in Fig. 4. Present data are found to be in good agreement in shape

(double humped distribution) but the amplitude of cross sections deviate significantly in the entire energy range. The maximum in the cross section for the first hump appears at about 20 MeV but the magnitude of maximum cross section value varies widely among the different measurements; it varies from 280 to 460 mb. There is only one literature available for cross section measurements above 37 MeV for this radioisotope and our data is lower (~ 20%) than the data of Aikawa et al. [13]. More measurements are required to minimize the discrepancies in the cross section data for this medicinally important radionuclide.

3.3 Independent production cross sections of ^{nat}Zn(α,x) ^{68}Ge for ^{68}Ge/^{68}Ga generator

^{68}Ge ($T_{1/2}$ = 270.953 days) is produced from ^{nat}Zn directly by the reactions listed in Table 1. ^{68}Ge ($T_{1/2}$ = 270.95 ± 0.16 days) decays to ^{68}Ga ($T_{1/2}$ = 67.71 ± 0.08 min) via electron capture (EC, 100%) process. ^{68}Ga disintegrates partially by positron emission (88.88%) with a maximum energy of 1899.1 keV and partially by electron capture (11.11%) to stable ^{68}Zn. ^{68}Ga can be obtained using a ^{68}Ge/^{68}Ga generator without the in house cyclotron facility for their uses in medicine. There are no gamma rays for decay of ^{68}Ge to ^{68}Ga. ^{68}Ga was in equilibrium with ^{68}Ge and the production of ^{68}Ge was measured indirectly using 1077.34 keV gamma rays of ^{68}Ga after a cooling period of four months. In this time, ^{68}Ga produced directly from ^{nat}Zn is considered to be decayed completely. The experimentally measured cross section data were compared with literature data [12–15] along with TENDL-2021 data [18] as shown in Fig. 5a and b. As it can be seen from Fig. 5a, present data are in good agreement in shape and magnitude with the earlier published data above 32 MeV but below this energy our data are much lower in magnitude. Though TENDL-2021 data are much lower in magnitude compared to the experimental data, the shape of excitation function is well reproduced with maximum in cross section at lower energy compared to experimentally determined values. Cross section data reported by Karpeles [15] are much higher in magnitude compared to our data and all the previously reported data. Hence, the data are presented separately in Fig. 5b with a comparison of our data.

3.4 Independent production cross sections of ^{nat}Zn(α,x) ^{69}Ge

^{69}Ge ($T_{1/2}$ = 39.05 h) is produced directly from ^{nat}Zn by the reactions listed in Table 1 and IT decay (100%) of the two metastable states of ^{69m2}Ge ($T_{1/2}$ = 2.81 μs) and ^{69m1}Ge ($T_{1/2}$ = 5.1 μs). ^{69}Ge decays by EC and β^+ (100%) processes to stable ^{69}Ga followed by the emission of several gamma rays. 1106.77 keV gamma rays were measured for

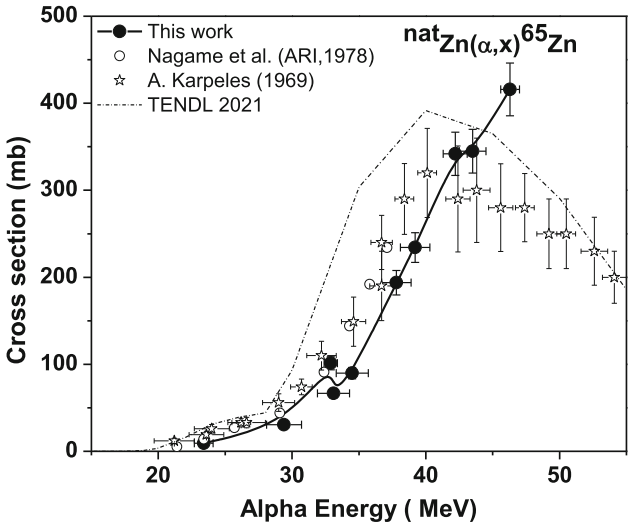

Fig. 7 Excitation function of ^{nat}Zn(α,x)^{65}Zn reaction and comparison with literature data and TENDL-2021 data

^{69}Ge after 50 h cooling period. The experimentally measured cross section data, earlier published data from literature [12, 13] along with TENDL-2021 data [18] are shown in Fig. 6. Our data match in shape but not in amplitude with the data from TENDL-2021 and earlier works. Magnitudes of our data are lower than the earlier reported data both at below 20 MeV and above 35 MeV. Though the ^{68,69}Ge, ^{66,67}Ga and ^{65}Zn radionuclides were produced from the same target under similar experimental conditions, significant disagreements in cross section data for ^{69}Ge and ^{67}Ga radionuclides were observed when we compared our data with the available published literature data. Experimental measurement of cross section is simple with respect to target irradiation and counting but there are often some unknown and unavoidable discrepancies in the measurements, which may result such disagreements [21].

3.5 Cumulative production cross sections of ^{nat}Zn(α,x)^{65}Zn

^{65}Zn ($T_{1/2}$ = 243.93 d) is produced from ^{nat}Zn directly by the reactions as listed in Table 1 as well as EC and β^+ decay (100%) of ^{65}Ga ($T_{1/2}$ = 15.2 min). ^{65}Zn decays to stable ^{65}Cu by EC and β^+ processes (100%) followed by the emission of intense gamma ray at 1115.5 keV. The measured cross sections are cumulative cross sections as counting of 1115.5 keV gamma rays were done after a cooling period of 122 days when ^{65}Ga decayed completely to ^{65}Zn. Our data are shown in Fig. 7 along with earlier published data [12, 15] and TENDL-2021 data [18]. Present data are in good agreement both in shape and in amplitude with the published literature data. Compared to the experimentally measured data, TENDL-2021 data are larger in magnitude. However,

the shape of excitation function is well reproduced in the studied energy range.

3.6 Thick target integral yields of ^{66,67}Ga, ^{68,69}Ge and ^{65}Zn

Thick target yields were deduced for these radionuclides from the measured cross sections assuming that the total incident energy is absorbed in the target using the following equation [27]:

$$Y = \frac{N_A H}{M} I\left(1 - e^{-\lambda t}\right) \int_{E_2}^{E_1} \left(\frac{dE}{d(\rho x)}\right)^{-1} \sigma(E) dE$$

where, N_A: Avogadro's number (mol^{-1}), H: isotopic abundance of the target, M: atomic mass of the target (g mol^{-1}), I: beam current (s^{-1}), dE/d(ρx): stopping power (MeV g^{-1} cm^2), σ(E): energy dependent cross section of the studied nuclear reaction (cm^2), λ: decay constant (s^{-1}), t: irradiation time (s), E_1: initial beam energy (MeV) and E_2: beam energy (MeV) below which the reaction cross section is insignificant. Thick target yield (Y) is calculated on the assumption that the target is irradiated for 1 h with 1 μA current and its unit is MBq/ μAh.

Experimentally measured cross sections were used for yield calculations and the stopping power of natural zinc was taken from SRIM 2013 over the energy range from their respective threshold up to 46.3 MeV. Experimentally thick target yields are measured generally at few discrete incident energies. A comparison of the theoretically calculated integral yields from the cross section data with experimentally measured yield data at few bombarding energies provides a benchmark experiment for the experimentally measured excitation function. The calculated integral yields for all the produced long-lived radionuclides from alpha induced reactions on ^{nat}Zn are shown in Figs. 8, 9 and 10 along with the earlier published literature data [12, 14, 16] taken from EXFOR [26] for comparison purposes. Among the yield data available in literature, Krasnov et al. [16] only experimentally measured thick target yields by irradiating thick samples of pure zinc metal produced by zone melting using stacked foil irradiation method whereas other literature yield data are theoretically calculated from their respective experimentally measured cross section data.

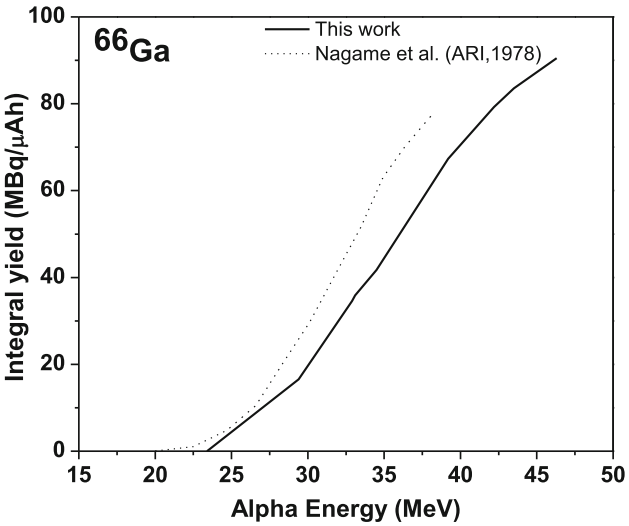

Fig. 8 Integral yield of ^{66}Ga with the earlier published literature data

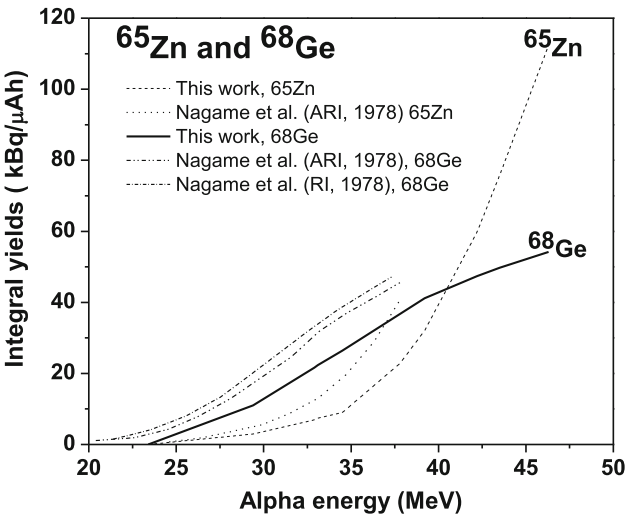

Fig. 9 Integral yield of ^{65}Zn and ^{68}Ge with the earlier published literature data

4 Conclusions

Excitation function of medicinally important radionuclides, ^{66,67}Ga, ^{68,69}Ge and ^{65}Zn, produced from the alpha particle induced reactions on natural zinc were measured from their respective threshold up to 46.3 MeV. Standard stacked foil activation method was used for irradiation and the induced gamma activity was measured offline using high resolution HPGe detector based gamma ray spectrometry system. The experimentally measured cross sections data were compared with the literature data as well as the TENDL-2021 predicted theoretical data based on the TALYS-1.96 code. The experimentally measured data of the present work are found to be in good agreement for ^{66}Ga, ^{68}Ge and ^{65}Zn and there are significant differences for ^{67}Ga and ^{69}Ge with the earlier published literature data. Thick target yields were calculated from the measured cross sections and compared with the yield data from the literature.

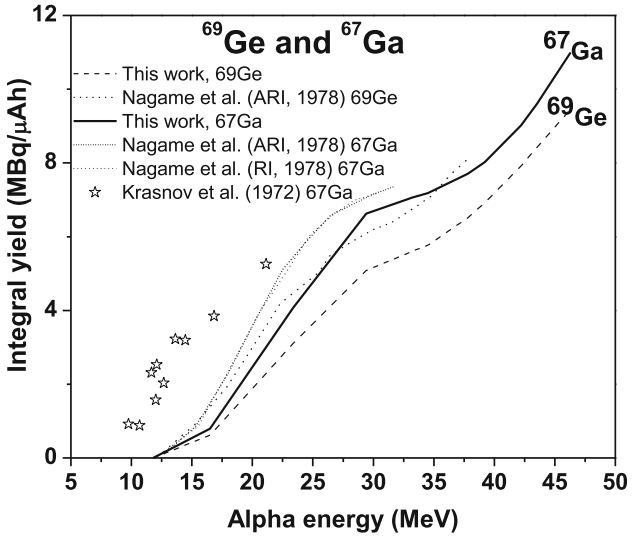

Fig. 10 Integral yield of ^{69}Ge and ^{67}Ga with the earlier published literature data

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Data Availability This manuscript has no associated data or the data will not be deposited. [Authors' comment: All the relevant data are given in the paper.]

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