



EVALUATION OF ^{67}Ga CROSS SECTIONS USING EXIFON CODE FOR MEDICAL APPLICATIONS

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ABSTRACT

Radioisotopes play very important roles in nuclear medicine for imaging and therapeutic applications. In the present studies, model calculation of excitation function for production of ^{67}Ga radioisotope was performed using the EXIFON code, a nuclear reaction cross sections theoretical model code, for the reaction $^{65}\text{Cu}(\alpha, 2n)^{67}\text{Ga}$. The work was performed in the incident alpha energy range of 0 - 40 MeV. Similarly, the Q-value software (interface) was used for the calculation of reaction threshold and Q-value energies of the reaction of interest and were respectively found to be 14.97 MeV and -14.10 MeV. The calculated excitation function has a peak value of 1025 mb around 25 MeV incident energy. The results from the EXIFON code were compared with the experimentally measured cross sections data retrieved from IAEA database, the EXFOR database, as well as the theoretical data from Talys code via its library, the TENDL-2019. Our results partially agree with the theoretical data from Talys code (via the TENDL-2019 library) within the investigated energy region. The results however overestimated the measured (experimental) data and only agree in shape of the excitation function. The present work does not consider the effect of shell structure during the execution of the EXIFON model code. This work could be of importance to the developers and users of nuclear reaction model codes for new developments and enhancements of the existing codes, as well as to serve as a rough guide for experimentalists during production of radioisotopes for nuclear medicine applications.

Keywords: Cross sections, Excitation function, ^{67}Ga , Nuclear reaction, Nuclear reaction model

INTRODUCTION

Despite significant improvement in the knowledge of nuclear reaction theory and in turn, improved size for the nuclear reaction database, the importance of new experimental data as well as the data from theoretical codes calculations, using reliable parameters, cannot be overemphasized. Several new radioisotope candidates for diagnosis and therapy have been emerging, leading to the quest for sufficient experimental and theoretical data for their effective validation and optimized production. The need for further nuclear data measurements (and model calculations) can also be seen in several other applications such as in alpha- and deuteron-induced reactions, high-energy accelerators as well as in low energy cyclotrons (Tarkanyi et al., 2014). Moreover, the database for the proton-induced reactions routes are well established, relative to the higher charged particles. However, following the advent of new candidate radioisotopes and additional production routes for radioisotope production optimization, there is recent surge of studies using the higher charged projectiles, especially the deuterons and alpha particles (Khandaker et al., 2014; Otuka et al., 2017; Usman et al., 2017a; Usman et al., 2016a; Usman et al., 2017b; Usman et al., 2020; Usman et al., 2016b).

The artificially produced new candidate radioisotopes are gradually finding a large number of potential applications in numerous fields of human endeavor (Art and Ayetkin, 2015). In nuclear medicine, radioisotopes mostly find very specific and important applications in diagnosis and therapy (Alharbi et al., 2012). The use of these radioisotopes for imaging or as therapeutic agents depends on their respective properties suitable for the application of interest. Two very popular modalities, the Positron Emission Tomography (PET) and Single Photon Emissions Computed Tomography (SPECT) are both used for diagnosis. The PET modality is a nuclear medicine imaging technique capable of producing a three-dimensional (3D) image of organs in the body. Emerging evidences indicate that PET has the promise of higher-quality images and greater diagnostic accuracy relative to SPECT.

The process basically determines pairs of gamma rays emitted indirectly by a positron-emitting radionuclide (tracer), which is introduced into the body on a biologically active molecule. PET producer requires the combination of a number of factors before it can become an important clinical tool. These factors include (Alharbi et al., 2012; Hod et al., 2021; Manhas et al., 2020; Ogawa et al., 2021):

- i. An imaging device must give good performance for isotope of choice.
- ii. The necessary radioisotope has to be readily available as at the time of need.
- iii. The combination of a radiopharmaceutical and a patient tissue for which the PET scan provides significant diagnostic information which is not readily available with other techniques.

SPECT imaging scan shows how blood flows to tissues and organs, capable of diagnosing tumors, infections, seizures, stroke and stress fractures in the spine. Usually, a particular radiolabeled pharmaceutical is injected into the blood stream, depending on the medical issue to be investigated. Some of the popular radioisotopes for this procedure include technium-99m, iodine-123, xeno-133, fluorine-18 and thallium-201. A radiolabeled glucose (Fluorodeoxyglucose, FDG) may be used for the examination of a tumor in brain. As gamma rays are emitted by the active isotope in the tracer, they are detected by a scanner, which are then collected by a computer, process and add them as 3D images of the investigated organ, such as the brain. Unlike the PET scan, in SPECT procedure, the tracer stays in the blood stream rather than being absorbed in the surrounding body tissues, thus limiting the images to only the areas where blood flows. SPECT scans are said to be cheaper and more readily available than PET scans (Hod et al., 2021; Manhas et al., 2020; Ogawa, et al., 2021), even though the PET scan are also. SPECT use common radiopharmaceuticals of nuclear medicine, in contrast to PET, which use positron emitters and followed by formation of two annihilation 511-keV photons.

Gallium-67 (^{67}Ga) has several important applications in nuclear medicine. It is popularly known for its usage in imaging and localization of infections (inflammatory lesions). Its usage in imaging is because of its emission of four (4) suitable gamma rays (93, 184, 296, 388 keV) (Van de Wiele, 2008) for this purpose. For localizing infection purpose, the ^{67}Ga radioisotope has been labeled in the form of ^{67}Ga -citrate for more than three (3) decades with very efficient results (Van de Wiele, 2008).

Furthermore, ^{67}Ga is Auger—electron emitting radioisotope similar to ^{125}I , with cancer therapeutic property following their significant level of cytotoxicity (Koumarianou et al., 2014; O'Donoghue and Wheldon, 1996). The emissions from this radioisotope are also found to be short-range in biological tissues, thus minimal damage to neighboring healthy cells (Usman et al., 2016b). Comparatively, the emission of 4.7 Auger and Coster-Kronig electrons per decay and the shorter half-life of ^{67}Ga ($T_{1/2} = 3.2617$ d) relative to ^{125}I ($T_{1/2} = 59.4$ d) makes the ^{67}Ga -labeled radiopharmaceuticals clinically more desirable to patients than the later (Koumarianou et al., 2014).

Experimentally, the ^{67}Ga radioisotope is usually produced in cyclotron machines and various production channels are available for this important radioisotope. Cyclotron-based radioisotope production is one of the most sophisticated technologies that allows the production of a number of isotopes of immense potential applications in the medical field (Usman et al., 2017b). Several studies in the literature have reported the production of the ^{67}Ga from induced nuclear reactions using proton, deuteron, tritium or alpha particles as activation projectiles. We have recently reported measured experimental cross section of the ^{67}Ga via the alpha particles production route in one of our previous studies (Usman et al., 2016b).

Model predictions of nuclear reactions are important in radioisotopes production as they provide reasonably reliable data where experimental data is not readily available, or, where the available data is scanty and insufficient. Moreover, the theoretical data serves as a guide to experimentalist in some cases. There are a number of nuclear reactions models capable of predicting nuclear reaction cross sections. These models include Tallys code (Koning and Rochman, 2012), Empire code (Herman et al., 2007), ALICE code (Alharbi & Azzam, 2012), Exifon code and few others. Each of these models have their strength and limitations, with some more powerful than others. The Exifon code is a simple to use code for the prediction of nuclear reactions cross sections for uncharged neutron and gamma rays and also for the charged proton and alpha particles. Though relatively old (as it has not been having significant updates in recent years) and not as sophisticated as other codes such as the Tallys code, the Exifon code can predict the cross sections of nuclear reactions in a fraction of a minute or in few minutes, depending on the maximum energy of interest. The code hold the advantage of being easy to understand and run, without requiring cumbersome activities and heavy task. There are several studies in the literature for detailed theory on the Exifon code, as well as several previous works which use the code for the calculations of cross sections (Ahmad et al., 2017; Ahmad et al., 2019; Chad-Umoren and Ebiwonjumi, 2014; Dauda, 2011; Dauda et al., 2017; Jonah, 2004; Hauser and Feshbach, 1952; Kalka et al., 1990; Kalka, 1991; Muhammed et al., 2011; Murata, 1997; Polster and Kalka, 1991).

BACKGROUND AND THEORETICAL FRAMEWORK

EXIFON, a nuclear reaction code, is an analytical and multi-step statistical model code. This code has the ability to uniquely describe angular distributions, reaction cross sections and emission spectra for pre-equilibrium, equilibrium and the collective the no-collective direct theory processes (Kalka, 1991; Chad-Umoren and Ebiwonjumi, 2014). However, a slight drawback of the code is its limitation to few projectiles which are: neutron-, proton-, and alpha-induced reactions, with the outgoing or emitted particles being neutrons, protons, alphas or photons in the outgoing channels. The development of statistical multi-step theory from the simple compound nucleus and single step direct models, were influenced by mainly three basic ideas, which include; the classification of nuclear states by their complexity or exciton numbers as proposed by Griffin, (1967), the distinctive configurations of bound and unbound state proposed by Feshbach et al. (1980) and the possible consideration of the chaotic nuclear Hamiltonian as a random matrix proposed by Agassi et al. (1975).

The concept of statistical multistep theory is becoming more relevant for a better comprehension of the mechanism of nuclear reactions, especially beyond 20 MeV energy. It involves the application of analytical model, for both processes of statistical multistep direct (SMD) and statistical multistep compound (SMC), to describe the nuclear reactions up to 30 MeV energy. The generalization of this concept can be possible to some respect: The extension to higher energies is performed including s-step direct processes for $s = 1$ up to 5. The same residual interaction is used for computing both formation and decay of the compound nucleus within SMC as well as SMD processes. Thus, there is no reference to the optical model (OM) reaction cross section. The OM cross section for charged particles was used to simulate coulomb effects only within the threshold region (Polster and Kalka, 1992). The α and γ processes are included in spin-isospin conservation when the two-body collision is considered. The calculation of Multiple Particle Emission (MPE) is generalized. Up to three decays of the compound nucleus are considered. The detailed formulation of this model makes it capable of predicting spectral emissions of photons, neutrons, protons and alphas, including pre-equilibrium, equilibrium, direct and also the MPE processes in a consistent mode. For several reaction types, nuclei and energies, calculations can be performed with one physical parameter set.

For the pure statistical multi step reaction model, a proposal was made for a unique description (a,x) emission spectra where a,b = n,p, α and γ (Neutron, Proton, Alpha and Gamma-Ray), as well as the excitation function (activation cross section). This technique is based on many body theory (Green Function Formalism) and random Matrix Physics. The (Version 2.0) EXIFON Code is a generalization of energies up to 100 MeV, and is capable of performing fast calculations of cross section from one global parameter set (Kalka, 1991). Detailed theoretical description of underlying theory to Exifon such as the Statistical Multistep Direct (SMD), the Statistical Multistep Compound (SMC) cross Section and many more can be found in the literature (Ahmad et al., 2017; Kalka, 1991; Polster and Kalka, 1991). The SMD and SMC theories are very vital in the understanding of the basis of the formation of the cross section.

METHODOLOGY

The theoretical model code EXIFON (version 2.0) was used to calculate the cross section of the reaction $^{65}\text{Cu}(\alpha, 2n)^{67}\text{Ga}$. The theoretical model is based on optical potentials, which

comprises of both statistical multistep (SMD) and statistical multistep compound (SMC). The code can make calculations for incident energies up to 100 MeV in a very fast mode and has the capacity to predict cross sections using global parameter set, with its output capable of being arranged in ENDF-6 format (Kalka, 1991).

To properly run the program, the first important step is to ensure that the existing Exifon code version on a system is working efficiently. The code is usually used or installed on a 32-bit computer operating system. In addition to specifying the target of material, the parameters of inputs as well as the outputs were properly defined. The input parameters for this study include use of alpha particles as projectile or incident particles, and the (^{65}Cu) isotope of copper as the target material. This is usually followed by prompt selection of 'excitation function' in the general option of the code. The investigation was performed in the medium energy range of 0 to 40 MeV. The incident energies of the bombarding alpha particles were specified in steps of 1 MeV in the present work. There exists a modification option, where one can make the

calculation with or without shell effect corrections. For the present study, the calculations were performed with default settings of the code. Other parameters, such as decay properties, relevant to the investigated radioisotope are summarized in in Table 2.

RESULTS AND DISCUSSION

The output results for the Exifon calculation can be found as the output data in the Exifon output directory as OUTEXI. Similarly, other DATA files such as A2N, ALF and AP (for Alpha-Neutron, Alpha-Alpha, and Alpha-Proton) in same output directory, each representing an exit channel or decay channel for production of a radioisotope, depending on the bombarded target.

The calculated cross section data were plotted as a function of alpha energy and the excitation curve obtained was compared with the Talys code data obtained from TENDL 2019 library (Koning et al., 2019), as well as the experimental literature data from EXFOR Data Library (Otuka et al., 2014). The results are discussed in the subsequent section.

Table 1: Cross-Sections for $^{65}\text{Cu}(\alpha, 2n)^{67}\text{Ga}$ Obtained using EXIFON Code.

Incident Energy (MeV)	Cross Section δ (mb)
16	118.73
17	355.5
18	617.47
19	791.09
20	876.31
21	945.72
22	984.38
23	1007.7
24	1017.8
25	1025
26	962.79
27	653.53
28	363.04
29	212.6
30	116.54
31	62.984
32	35.196
33	22.581
34	14.599
35	9.8236
36	7.4984
37	5.8638
38	4.7729
39	3.9799
40	3.5464

Similarly, the graphical plot of cross section δ (mb) against incident alpha energy (MeV) is given below.

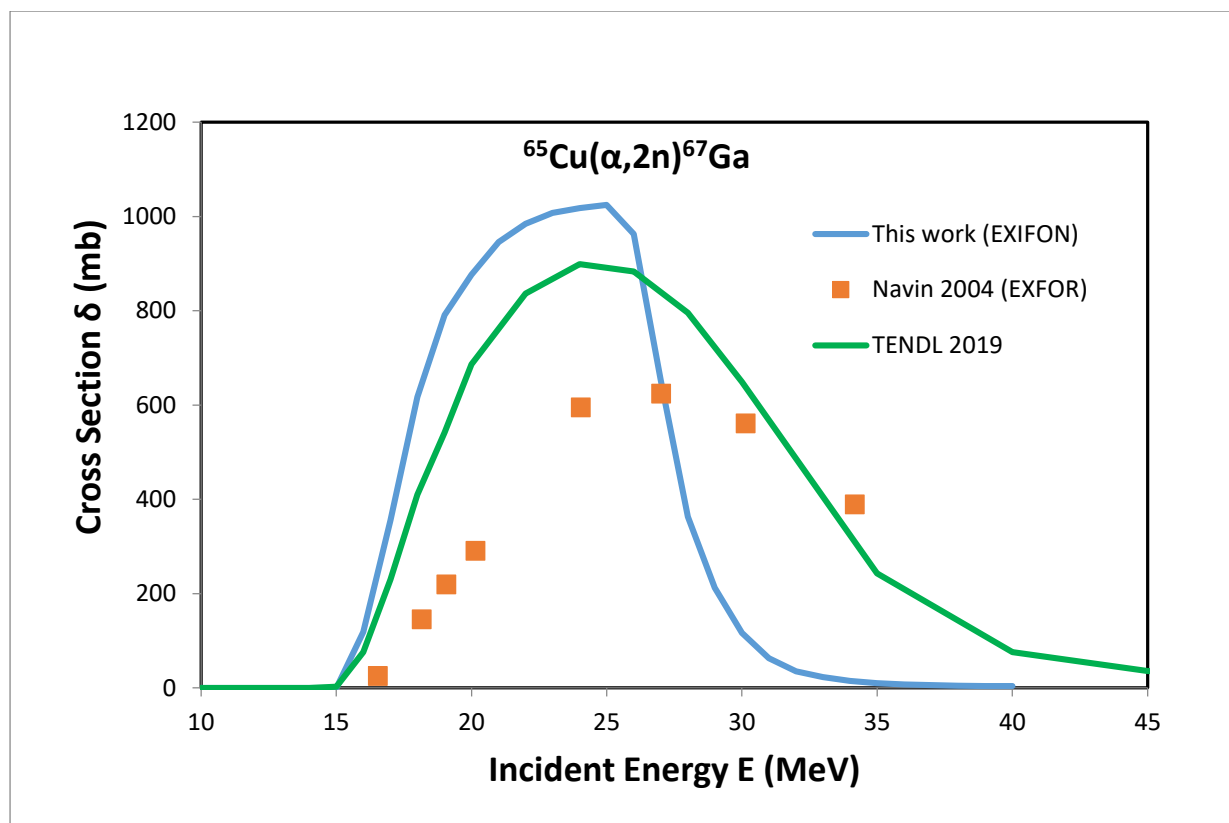


Figure 1: Excitation Function for the $^{65}\text{Cu}(\alpha, 2n)^{67}\text{Ga}$ Reaction

Evaluated cross section of Ga-67

Table 1 shows numerical values of the evaluated cross sections of ^{67}Ga from alpha bombardment of ^{65}Cu using Exifon model code. The prediction of the cross sections by the code in the present work shows partial agreement with the advanced Talys code data from TENDL 2019 library, especially up to about 20 MeV, beyond which the estimated cross sections from the Exifon code look larger than the TENDL-2019 data, up to about 26 MeV. A sharp decrease or fall in magnitude of the present cross sections relative to those reported in TENDL-2019 are observed beyond the 26 MeV

energy. Generally, this work predicts higher cross sections up to around 27 MeV incident alpha energy. As indicated in Fig. 1, the fine shape of the excitation function has its Ga-67 production peak at about 25 MeV and corresponding cross sections values of 1025 mb, which indicates that the possible region of the higher yield production prospect. Both Exifon and Talys codes (TENDL-2019) could not accurately reproduce the experimental work of Navin et al. (2004), even though all the three (3) share similar shape rather than magnitude.

Table 2: Decay Data of Ga-67 Retrieved from Nudat 2.6 (NuDat 2.6, 2011) and Q-Value Software (QCalc, 2016).

Nuclide	Half-Life (Hours)	Decay Mode (%)	E_γ (KeV)	I_γ (%)	Contributing Reaction	Q-Value (MeV)	Threshold (MeV)
^{67}Ga	78.3	EC (100)	93.31	39.2	$^{65}\text{Cu}(\alpha, 2n)^{67}\text{Ga}$	-14.102	14.971
			184.5	21.2			
			300.2	16.8			
			393.5	4.68			

CONCLUSION

The excitation function for the theoretical production of ^{67}Ga , an important radionuclide in nuclear medicine procedure was investigated via alpha induced reaction on copper (^{65}Cu) target and using Exifon nuclear reaction code. The goal of the EXIFON code calculation was achieved and the obtained results were compared with the experimental data from EXFOR library and also with a theoretical data by TALYS code via TENDL 2019 library, for validation. The comparison of the results and other data show that the prediction of ^{67}Ga via the investigated route and code is relatively good and with considerable accuracy. The present result could be useful for the developments and enhancements of another version of the Exifon code and other nuclear

reaction codes, as well as providing a useful information for production of ^{67}Ga radioisotope to experimentalist.

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