

Simulation of Excitation Functions for Sb^{123} & Sb^{124} Production from (n, na) & (n, a) Reactions Using Exifon Code

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Abstract

The widespread integration of radionuclides in diagnosing and treating diseases is now a fundamental aspect of global healthcare systems, with techniques like Positron Emission Tomography (PET) and Single Photon Emission Computed Tomography SPECT playing vital roles in cancer detection and radiotherapy treatments such as I-131 for thyroid cancer which is gaining traction. However, shortages of radionuclides can disrupt medical facilities, leading to delayed diagnoses, increased costs, and patient distress. Therefore, the exploration of non-reactor methods for radionuclide production is essential. This research specifically delves into calculating excitation functions for generating Antimony-123 and Antimony-124 via (n, an) and (n, a) reactions on stable Iodine-127 within the 1-30 MeV energy range using the EXIFON Code. These isotopes are crucial for biological and biomedical labelling, with a new dataset establishing threshold energies of 9 MeV and 20 MeV for Sb-124 and Sb-123, respectively, and corresponding cross-section value of 0.1mb, showcasing favorable agreement with data from Evaluated nuclear data file (ENDF) and experimental data (EXFOR). This is to facilitate Sb-123 and Sb-124 production using cyclotron.

Keywords: EXIFON, Radionuclide, PET, SPECT, Antimony

1. Introduction

Nuclear reaction parameters such as nuclear cross section and excitation function among others, provide fundamental information that facilitate the prediction of the possibility and production as well as purity of nuclear reaction products, or radionuclides and hence play a crucial role in the course of nuclear energy and development of fusion and fission reactors. As a consequence of this principle, nuclear reaction data describing the interactions of various projectiles such as protons, neutrons or alpha particles with target nuclei are becoming increasingly important for optimizing radionuclide production, analysis of impurities and radionuclide purification as well as ascription of the most suitable route for radionuclide production with high efficiency (Iwamoto, 2007). In nuclear theory, quantum mechanics is used to predict the probability that a specific nuclear reaction will occur under certain conditions (Hibstie et al., 2018). This quantitative prediction is the nuclear cross section which can be measured in the laboratory with the use of experimental techniques.

Radioisotope production for use in nuclear medicine has already become one of the most important contributions of nuclear physics because radionuclides play a central role in life saving applications especially nuclear diagnostics and therapeutics. An example of radionuclide therapy is treatment of thyroid disease with radioiodine while nuclear diagnostics involves application of short half-life nuclide

attached to suitable radiopharmaceutical followed by measurement of nuclear emission as a result of accumulation and movement of activity from outside the body (Qaim, 2012). With the use of external detectors, it's used to capture and form images from the emission of radiation by the radiopharmaceutical; to detect disorders of the human body's vital organs such as the heart, liver, brain and others. This process is called emission tomography which includes PET and SPECT (Zubaida & Ahmad, 2019). However, as the medical cases requiring nuclear diagnosis and therapy are on steady rise, the nuclear medical services are vulnerable to failure in the event of radioisotope shortage. Thus if anything hitches the availability of radioisotope, most of nuclear medical services may halt leading to use of inferior procedures bereft of the needed precision and may cause fatalities. This has happened in the United States during the shortage of ^{99}Mo which is used in producing Technetium. The shortage was due to stoppages of nuclear reactors that are needed in the production of the radionuclides (Ruth, 2014). Hence with the teeming morbidity of the Nigerian population a crisis may unfold if steps are not taken to ensure radioisotope availability, this is the basis behind exploring the various routes of radionuclide production including use of cyclotrons, particle accelerators as well as low enrichment. This research seeks to obtain reaction cross sections for (n, na), and (n,a) reactions on stable ^{127}I within the energy range of 0 - 30MeV and to evaluate the excitation functions for the reactions. The findings will facilitate the production of Sb-123 and Sb-124 using other routes apart from the use of nuclear reactors.

Radioisotopes produced from (n, na) and (n, a) reactions on ^{127}I have been found to play important medical roles. ^{127}I (n, na) and ^{127}I (n, a) produces Antimony 123 and 124 respectively. Sb-123 is a stable isotope useful for production of Sb-124 which has a half-life of 60.20 days and decays via β emission producing X-rays and gamma rays with energy of 27 – 2700KeV (Almeida et al., 2010). This radionuclide is used as radio labelling agents to trace anatomical and physiological processes for detection of cancers. It is also used in animal organ distribution studies for pharmacological evaluations. Production routes of medical radionuclides need to be optimized in order to produce higher yields and to achieve high purity (Ahmad & Koki, 2017) and for this to be accomplished, nuclear data which comprises reaction cross section, excitation function, reaction energetic and angular distribution etc. need to be evaluated (Chadwick, 1995). Threshold and maximum cross sections with corresponding incident energies facilitate selection of best reaction route and how to get maximum yield. Such evaluated data are usually validated through comparison with the IAEA evaluated (ENDF) and experimental (EXFOR) nuclear data libraries.

EXIFON Code is a FORTRAN based nuclear reaction code that uses analytical model for the description of excitation function of particle induced reactions within the statistical multistep model (SMD/SMC). The code is adept in calculating cross sections for the pre-equilibrium, equilibrium and emission spectra for incident energies up to 100 MeV and was utilized in this work for its ability to provide smooth, continuous and precise nuclear reaction data over a wide incident energy range. Further, the code is fast and easy to handle for cross section predictions from one global parameter set.

2. Theoretical Background

Description of nuclear reactions are usually achieved with the use of several nuclear models which are linked together to calculate nuclear cross sections. One of the most important models however, is the Statistical (or Compound nuclear) model which belongs to the group of reaction models in the slow order along with the pre-equilibrium model (Hilaire, 2000). This model has been receiving attention over the years for description of reactions in the low and intermediate region up to 100 MeV. The models provides that a compound nucleus is formed with high enough excitation energy so that the incident particle –

target nucleus interaction may excite many states (Ahmad et al., 2017). In addition, it assumes the incident energy is shared among individual components of the nucleus which equilibrate fully before nuclear decay takes place. This implies no correlation of compound nucleus formation with the decay process and hence the existence of independent expressions to describe the ingredients of the process as summed up in equation (I). These processes include Statistical Multistep Direct (SMD) and Multistep Compound (SMC). Same residual interaction was utilized in computing both the formation and decay within SMD and SMC. Thus reference is not tenable to the optical model (OM) reaction cross section. OM cross section for charged particles was used for simulation of Coulomb effects in the threshold region (Ahmad & Koki, 2017).

Multiple particle emission (MPE) calculation is generalized and up to three decays of the compound nucleus are considered. The statistical model is formulated in detail for predicting emission spectra for neutrons, protons, alpha particles and photons with inclusion of equilibrium, pre-equilibrium, direct and MPE processes with high degree of consistency. One physical parameter set for several energies and reaction types was employed in performing calculations. Statistical multistep models are very successful in describing nuclear reactions at energies up to about 100 MeV (Kalka, 1992). These models enable the description of direct, pre-equilibrium and equilibrium processes in a constant way for a wide number range and various reaction channels e.g. neutrons, protons, alpha particles, and gamma rays. The application of Statistical Multistep Model to heavy nuclei requires the consideration of fission as a competing process to particle and gamma-ray emissions. Therefore, Statistical Multistep Models should be extended to the fission channel.

2.1 Statistical Multistep Model

In the Statistical Multistep Model, the total emission spectrum of the process (a, xb) is divided into three main parts (Ahmad & Koki, 2017).

$$\frac{d \sigma_{a,xb}(E_a)}{dE_b} = \frac{d \sigma_{a,b}^{SMD}(E_a)}{dE_b} + \frac{d \sigma_{a,b}^{SMC}(E_a)}{dE_b} + \frac{d \sigma_{a,xb}^{MPE}(E_a)}{dE_b} \quad (1)$$

The first term on the right-hand side of the Equation (1) represents the statistical multistep direct (SMD) part which contains from single step to five-step contributions. Besides particle-hole excitations also collective photon excitations are considered. The second term represents the statistical multistep compound (SMC) emission which is based on a master equation. Both terms together (SMD+SMC) represent the first chance emission process (Pandey et al., 2011). The last term of the Equation (1) represents multiple particle emission (MPE) reaction which includes the second chance, third chance emissions, etc. These terms are summarized below:

$$\frac{d \sigma_{a,xb}^{MPE}(E_a)}{dE_b} = \sum \frac{d \sigma_{a,cb}(E_a)}{dE_b} + \sum \frac{d \sigma_{a,cdb}(E_a)}{dE_b} + \dots \quad (2)$$

3. Method

Theoretical calculations of nuclear cross-sections were performed using nuclear model code EXIFON which is a FORTRAN based computer program that outputs nuclear reaction data for a wide range of incident energies.

3.1 Computer Code EXIFON 2.0

EXIFON code is based on an analytical model for the description of excitation function of particle induced reactions within a statistical multistep direct and multistep compound reactions (SMD/SMC) model. The computer code is very important for several technical applications if the experimental data are not available or unable to measure the reaction cross-sections due to the experimental difficulties. This code is capable of calculating equilibrium and pre-equilibrium emission cross-section, and valid for excitation energy of the compound nucleus up to 100 MeV. EXIFON code provides continuous and smooth description of nuclear reactions over a wide energy and mass range which is based on an analytical model for statistical multistep direct and multistep compound reactions (SMD/SMC model). It predicts emission spectra, angular distribution and activation cross-section for neutrons, protons, alpha particles and photons. Multiple particle emissions are considered for up to three decays of the compound system. EXIFON is a fast, easy to handle code which predicts cross-sections from one global parameter set. The only adjustable quantity is the pairing shift. The INPEXI code creates input files for EXIFON 2.0 from mass and shell – correction tables. The MAKE6 code transforms EXIFON output into an ENDF-6 format file (Ebiwonjumi, 2014).

3.1.1 General Features of EXIFON Code

Computer: PC/AT, Programming language: FORTRAN 77, Characters: ASC II (IBM International Graphic), Memory size: 300kbyte, Number of subroutines: 13, Records; about 1600, running time: about 20 seconds per incident energy.

3.1.2 Standard Parameters Set

The following global parameters are used:

Strength of surface – delta interaction:	$F0 = 27.5\text{MeV}$
Radius Parameter:	$r0 = 1.21 + 4.0A^{-2/3} - 15A^{-4/3}$
Fermi energy:	$Ef = 33\text{MeV}$
Potential energy:	$V0 = 52 - 0.3E\alpha \text{ MeV}$
Pairing shift:	$\Delta = 12.8A^{-1/2} \text{ MeV}$
Phonon (Breit-Weigner) width:	$\Delta w = 14\text{MeV}$
Optical Model Potential OM:	for protons, neutrons and alpha (Kalka, 1992).

In the calculations, the following parameters can be changed; Strength of surface-delta interaction F0, Radius parameter r0, Fermi energy Ef, Phonon width Δw and global OM parameter for protons.

3.2 Procedure

Inputting data in Exifon code involves target nucleus specification in which user add mass number A of the target nucleus. This is followed by specifying the incident particle (neutron, proton or alpha), the

first incident energy, and then the incident energy step. Subsequently, the cross-section corresponding to each particular energy step will be generated. The output data (OUTEXI) for the calculation are then stored in the output directory, also DAT file name is stored in the set output directory. The computer program Python was used to generate a code that can convert the results which comes in complex notepad format into a simple format for easy processing.

3.2.1 Input Data and Output Data files

For each target nucleus, input data files are necessary. They contain all binding energies BC and shell correction energies dW, as well as phonon parameters. They can be created with code INPEXI.EXE. One input data file contains all the information for neutron, proton and alpha induced reactions. Output data files are equally necessary for each target nucleus. The emission spectra of neutrons, protons and alpha particles are stored in the file AXN.DAT, AXP.DAT, AXA.DAT and AXG.DAT respectively. A short documentation is written to OUTEXI after each run.

3.3 Data Analysis Procedure

The data of this work has been arranged in proper order and organized by using tabulation method. These organized data was analyzed and described graphically with the help of spreadsheet (Microsoft Excel). Results generated from this analysis were then compared with the experimental (EXFOR data library) and evaluated nuclear data libraries (ENDF), from the International Atomic Energy Agency (IAEA), to validate the calculated result.

4. Results and Discussion

The computed reaction cross-sections based on neutron energy using (n, na) and (n, a) reactions are given in Figure (1–2) below. Figure 1 shows I-127 (n, na) many bodies reaction for the production of Sb-123. The threshold energy is 20 MeV with corresponding cross section of 0.1mb, up to 2.4 mb of the research maximum energy 30 MeV. Sb-123 is an important radionuclide useful in biomedical and biological labeling. The (n, na) reaction was compared with experimental (EXFOR) and evaluated (ENDF) in the IAEA data library and it shows good agreement. Figure 2 shows (n, a) reaction which is the break-up reaction for the production of Sb-124, with threshold energy 9 MeV and corresponding reaction cross section 0.1 mb. For the maximum energy within this research limitation, the reaction cross section is 26.8 mb. The (n, a) reaction was compared only with evaluated nuclear data (ENDF), and there was agreement up to 19.0 MeV neutron energy where the latter exhibited maximum cross section which commence regression downwards while the Exifon values appear to mount higher possibly to reach maximum cross section in higher energy beyond the scope of this work as shown in Figure 2. Sb-124 is an important radionuclide useful in biomedical and biological labeling. It decays by β -emission, gamma and X-rays, with energy altering from 0.027 to 2.700 MeV.

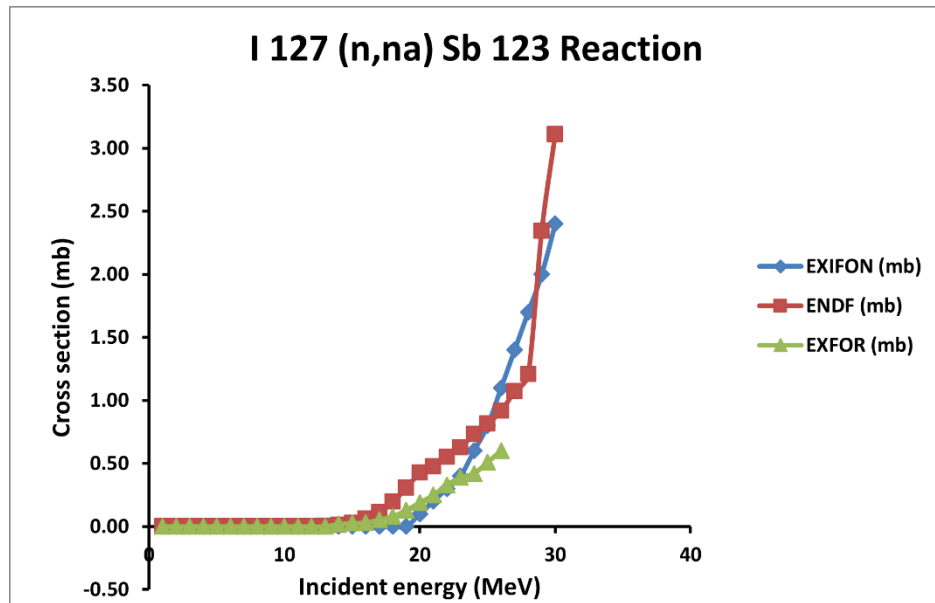


Figure 1: Excitation function of I-127 (n, na) Sb-123 reaction

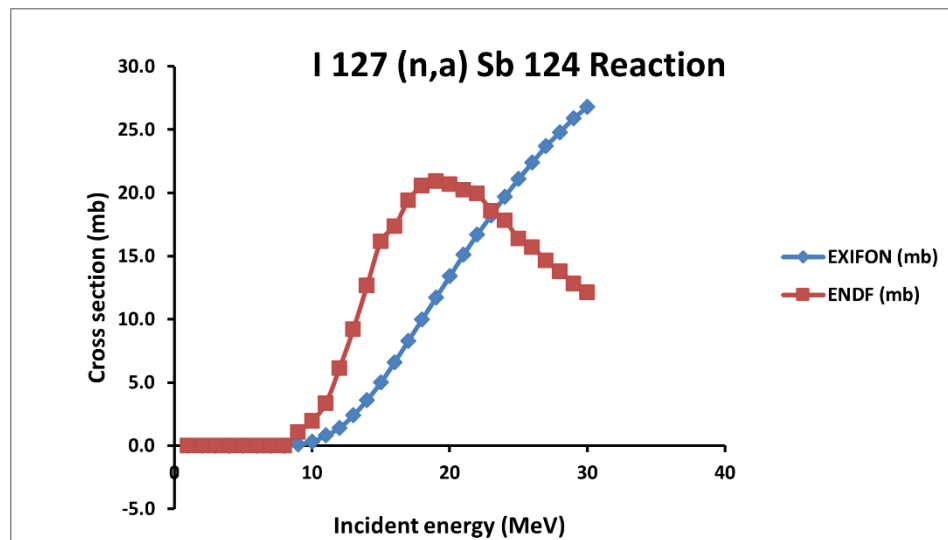


Figure 2: Excitation function of I-127 (n, a) Sb-124 reaction

5. Conclusion

In compliance to the original goal, a new data set for the production cross sections of the Sb-123 and Sb-124 radionuclides through the neutron bombardment on natural Iodine have been reported in the energy range of 1 - 30 MeV using the EXIFON Code. The evaluated cross sections have much importance in several fields such as nuclear medicine, trace elemental analysis and improvement of model calculations. The calculated data having rendered favorable agreement with both evaluated nuclear data

file (ENDF) and experimental nuclear data (EXFOR) has demonstrated good validity. Hence, it can be assertively indicated that radioisotope production to generate important radionuclides such as Sb-123 and Sb-124 that are a crucial part of modern medical diagnosis and therapy has the potential of contributing to national development through the provision of nuclear medical services to the citizens. Certainly, the evaluations will go a long way to augmenting the supply and availability of these radionuclides and as well avoid the hazardous route of reactor production as well as the untoward consequences of their shortage in nuclear medical facilities.

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