

EVALUATION OF EXCITATION FUNCTION OF NUCLEAR DATA ON PROTON INDUCED REACTIONS ON IRIDIUM

Temitope Funmilayo Ojo and Idris Ahmad

Department of Physics, Bayero University Kano, Nigeria

Abstract

In recent times, the production of radioisotopes for diagnostic and therapeutic purposes has become very important. This study aims to provide current cross-section production and theoretical results of proton induced reactions of iridium isotope on incident energy range of 1 – 30 MeV in order to improve the reliability of nuclear stimulations. The excitation functions for the production of $^{191,192,193}\text{Ir}$; $^{191,192,193,194}\text{Pt}$; and $^{189,190,192}\text{Os}$ from (p,p) , (p,α) , (p,g) , $(p,2n)$, (p,np) , $(p,n\alpha)$, and $(p,3n)$ reactions on Iridium-193 nucleus respectively were investigated using EXIFON code. The threshold energies for the reactions were 10 MeV, 20 MeV, 7 MeV, 9 MeV, 12 MeV, 16 MeV and 1 MeV with maximum cross-sections of 215.6 mb, 0.7 mb, 3.9 mb, 1300 mb, 1.2 mb, 0.6 mb and 1293.8 mb respectively. The results obtained were compared to the evaluated nuclear data files (ENDF) gotten from the IAEA nuclear data bank and our results showed close agreement with the evaluated data. The calculated data were aimed to enhance the production of these radioisotopes, in order to prevent medical mishaps and even shortages.

Keywords: Cross section, ENDF, Excitation function, EXIFON, Nuclear reaction.

1. Introduction

Nuclear reaction database is a form of theoretical tool that provides particle induced reaction cross-sections for isotopes. The isotope technology has contributed to developing advanced forms of radioisotopes, applicable in both medicine and industry. The evaluation of the cross-section from nuclear structure of radionuclides could be probed experimentally or can be evaluated by using systematics or theoretical model calculations and this could include proton induced reaction channels [1][2]. Nuclear reaction cross-sections, especially with incident particle energy above 0.1 MeV, have direct relation to nuclear data files when they are compared to experimental data and evaluated cross-sections from nuclear data files. This is mostly possible when nuclear models and computer codes are used to calculate these cross-sections [3]. Nuclear cross-sections are important in nuclear reaction because of their applications in many areas of physics, such as nuclear astrophysics, nuclear energy, and national security [4]. Several nuclear models are used to calculate both proton and neutron induced reaction cross-sections; this is due to the fact that some radioisotopes, like Iridium, are difficult to evaluate experimentally. Hence, evaluated nuclear data are required [5].

It is of vital importance that new reactions be made available for monitoring of charged particle beams (mostly proton particle beams, as attempted in this work). It is also important that the target nucleus have well-characterized decay data, wide thickness range, and high incident particle energy monitor reaction applications. By expanding the measurements of excitation functions of charged particle beams as well as calculating statistical models and optimizing medical isotope production modalities, more options will be open for malignant cancer therapy and medical imaging [6]. As remarkably important radiation therapy can be, industrial radiography may expose workers to occupational accidents such as radiodermatitis, which is due to the high penetration of the radioactive isotope in the human body. Although radionuclide

Correspondence Author: Temitope F.O., Email: topejanejo92f@gmail.com, Tel: +2348138401019, +2348157303688 (IA)

Transactions of the Nigerian Association of Mathematical Physics Volume 15, (April - June, 2021), 113 –120

imaging such as angiography and bone scintigraphy has been used by clinicians to examine the vascular status of damaged tissues or bones of patients, so as to know what stage of damage has been caused on the human tissue, it is quite advisable to prevent frequent exposure to radioisotopes like Ir-192 that can cause these damages[7].

Nuclear fusion has been one of the all-time attractive sources of energy in terms of safety and environmental impact. Nuclear fusion reactors have been studied to help in understanding excitation functions and nuclear cross-sections. These nuclear data are being used to investigate structural materials of nuclear fusion reactors, develop nuclear model mechanisms from nuclear reactions and also contribute to fusion reactor technology [8].

The separation of a neutron from Iridium-193 by photonuclear reaction can lead to the production of a stable radioisotope, Iridium-192. Iridium is one of the rare platinum group elements on earth and it can have medical and industrial purposes (such as industrial monitoring, ignition plugs, and satellite systems) [9]. The reaction $^{193}\text{Ir}(n,\gamma)$ leads to the formation of ^{194}Ir by determining the effective resonance neutron cadmium transmission factor of the reaction. This reaction, $^{193}\text{Ir}(n,\gamma)^{194}\text{Ir}$, is used in calculating experimental resonance energy and q-value using a reliable cadmium ratio method[10]. $^{193}\text{Ir}(p,pn)$ produces the radioisotope ^{192}Ir , a good product of iridium spallation, by bombarding iridium with proton of up to 87 MeV of energy[11].

Medical and industrial radionuclides need to have optimum production route as well as higher yield and purity. This is solely because nuclear data is highly necessary for calculating excitation functions, cross-section values, incident energies and threshold frequencies [12]. The International Atomic Energy Agency (IAEA) has been leading this research for about five decades and which is made available online via the IAEA nuclear data services website. During the course of this research, innovation, propagation, advancement, and further, through the nuclear data library, the evaluation of reaction cross-section and other calculations are validated through comparison[13]. However, a growing deficiency crisis of nuclear reactors as well as reactor shutdown is a growing concern as a lot of procedures had to be cancelled or postponed; this challenge has led to an increase in tumours and malignant diseases[14]. This situation may bring fatalities, not to mention ethical and socioeconomic effects, on hospitals. Hence there is need to find other alternative routes of production of radionuclides to take care of any eventual shortages. The main objectives of this research work are to obtain maximum and minimum cross-section (with and without shell structure) for the reactions, their corresponding incident energies, and also to investigate nuclear model calculations on the excitation functions with EXIFON code and to relate the radioisotopes produced to their uses in nuclear medicine.

2. THEORETICAL BACKGROUND

It is very important to have knowledge of nuclear reaction cross-sections from practical point of view and for theoretical understanding of nuclear interactions. The interaction of a nucleus with light particles such as alpha, proton, neutron etc., is of foremost importance as it changes the configuration of the nucleus and is useful in transformations of the nucleus into various other nuclides [15].

The Fermi gas equilibrium model was developed by Harp, Miller and Berne in 1968 is widely known as HMB model [16]. In 1971, Gadioli et al developed and perfected the original Griffin model into what came to be known as Exciton model[3]. Exciton and hybrid models are only special cases of a more general master equation approach, originally proposed by Pauli in 1928[17].

The concept of statistical multistep processes has over the years, become more and more important for the understanding of nuclear reaction mechanism, especially above 20 MeV[18].

These models enable the description of scattering process that takes place in two ways; namely direct and compound (pre-equilibrium and equilibrium) nucleus formation processes and these are called shape elastic and compound elastic scattering respectively [19].

The calculation of Multiple Particle Emission (MPE) is generalized. Up to three decays of the compound nucleus are considered. Calculations are performed with one physical parameter set for several nuclei, several energies, and several reaction types. The application of a 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.

STATISTICAL MULTISTEP REACTION

In the Statistical Multistep Model, the total emission spectrum of the process (a,xb) is divided into three main parts[16].

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

Where $\sigma_{a,xb}$ = the cross-section for (a,xb) process, $\sigma_{a,b}^{SMD}$ = the cross-section for direct reaction for the production of 'b' from 'a' and E_b = the excitation energy for the formation of 'b'.

In equation (1), the first term on the right-hand side represents the statistical multistep direct (SMD) part which consists of

up to five-step contributions from single-step. Collective phonon excitations are also considered besides particle-hole excitations. The second term on equation (2) stands for statistical multistep compound (SMC) emission, and this is based on a master equation. On bringing both terms together, we have (SMD+SMC) which represents the first-chance emission process [20]. The last term in equation (1) stands for the multiple particle emission (MPE) which consists of second chance emissions, third chance emissions, etc[20].

We summarize these terms below as:

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

In order to keep the model manageable, a simple two-body interaction is assumed [18]:

$$I(r_1, r_2) = -4\pi \frac{F_0}{A} [X_{nl}(R)]^{-4} \delta(r_1 - r_2) \delta(r_1 - R) \quad (3)$$

With the residual interaction, $F_0 = 27.5\text{MeV}$ taken from nuclear structure considerations. The factor $[X_{nl}(R)]^{-4}$ consists of the wave function at the nuclear radius $R = r_0 A^{1/3}$

The single-particle state density of proton particles p with mass μ_p is given by:

$$\rho(E_p) = \frac{4\pi V \mu_p (2\mu_p E_p)^{1/2}}{(2\pi \hbar)^3} \quad (4)$$

$$= (4.48 \times 10^{-3} \text{ fm}^{-3} \text{ MeV}^{-3/2}) r_0^3 A E_p^{1/2}$$

where $V = \frac{4\pi R^3}{3}$ is the nuclear volume.

The single-particle state density of bound particles (at Fermi energy) [18] is then defined by

$$g = 4\rho(E_F) \quad (5)$$

3. METHODOLOGY

A nuclear model computer code called EXIFON was used in this work to obtain nuclear reaction cross-sections on a target nucleus within the incident energy range of 1 – 30 MeV. An output data (OUTEXI) was used for this calculation which was then stored in the output directory with the file name DAT. The computer program Python was used to generate a code that can convert the results into simple formats.

The results gotten from this analysis were compared to evaluated nuclear data files (ENDF); The evaluated data (ENDF) was extracted from the IAEA nuclear data library.

3.1 Computer Code EXIFON

Computer codes have been developed to fill in for technical difficulties or to solve experimental difficulties in measuring nuclear reaction cross-sections. EXIFON code is a computer code which 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 code can calculate equilibrium and pre-equilibrium emission cross-section and is valid for excitation energy of the compound nucleus up to 100 MeV.

EXIFON code is a fast, easy to handle code which predicts cross-sections from one global parameter set. The only adjustable quantity is the pairing shift. It 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, and alpha particles and photons.

Results obtained in this work were properly arranged and organized using tables. The organized data were studied and arranged using a spreadsheet.

3.2 Data Analysis Procedure

The nuclear reaction data produced in this work has been properly arranged and organized using tabulation method. These organized data described graphically with the help of Microsoft Excel spreadsheet were analysed and interpreted accordingly by comparing with nuclear data files.

4. RESULTS AND DISCUSSION

The results of the excitation function calculations based on proton particles are given in figure (1-7) along with their available evaluated data. Experimental data (EXFOR) was not found for the following proton-induced reactions.

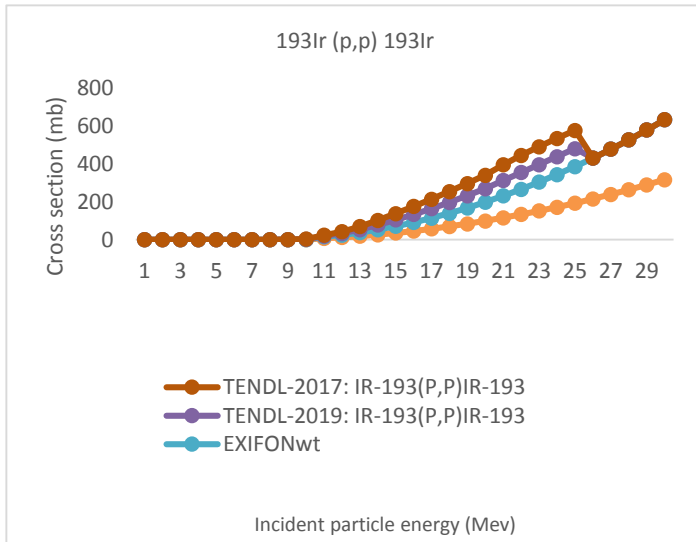


Fig 1 Excitation function for Ir-193 (p,p) Ir-193 reaction

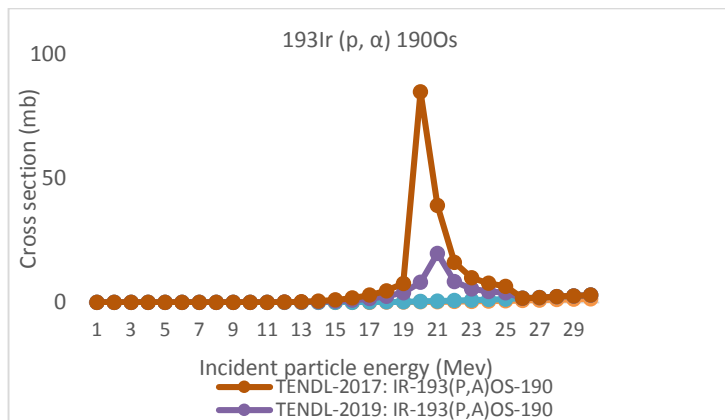


Fig 2 Excitation function for Ir-193 (p, alpha) Os-190 reaction

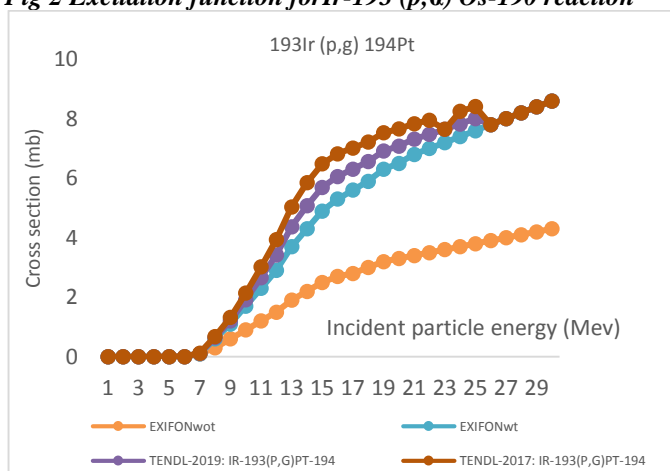


Fig 3 Excitation function for Ir-193 (p, gamma) Pt-194 reaction

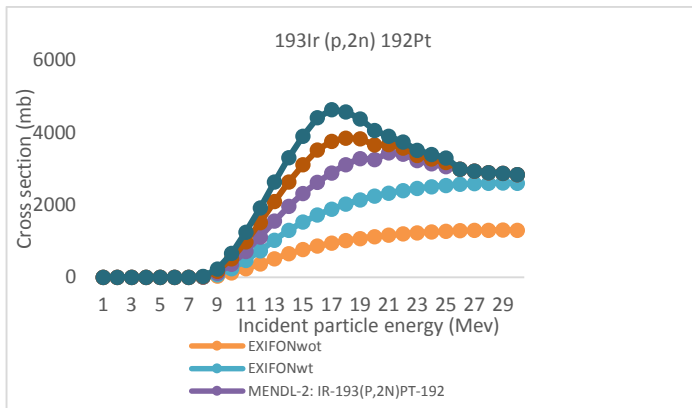


Fig 4 Excitation function for Ir-193 (p,2n) Pt-192 reaction

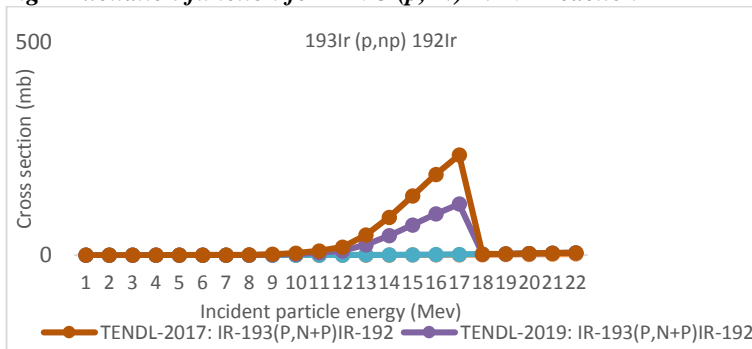


Fig 5 Excitation function for Ir-193 (p,np) Ir-192 reaction

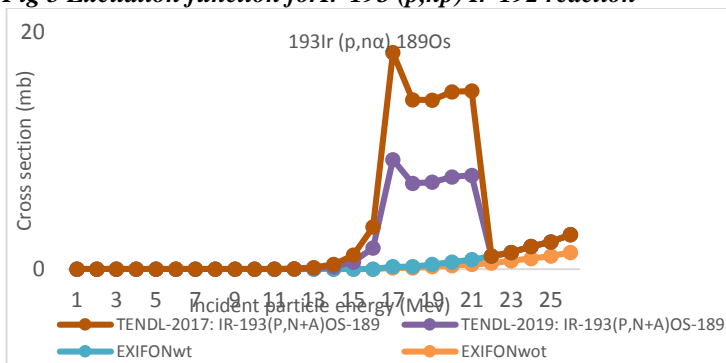


Fig 6 Excitation function for Ir-193 (p,nα) Os-189 reaction

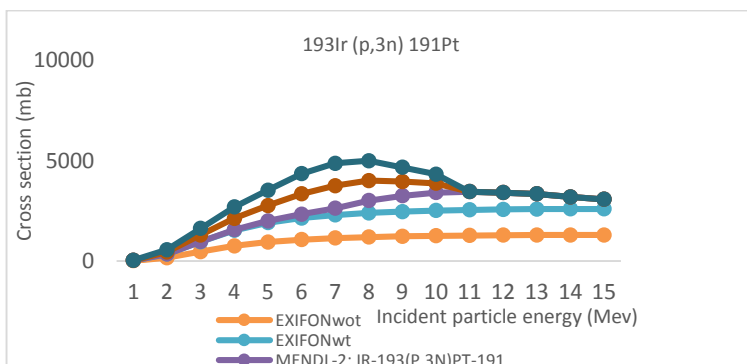


Fig 7 Excitation function for Ir-193 (p,3n) Pt-191 reaction

From the results above, there is little difference between the excitation functions with shell corrections and those without shell correction.

Fig. 1 shows Ir-193 (p,p) Ir-193 reaction, which is an elastic scattering because the proton particle passes through Ir-193 and ejects with the same nuclei and is unexcited.

Ir-193 is a non-radioactive isotope and is used in special alloys, and forms an alloy with osmium, which is used for pen tips and compass bearings.

According to my data gotten from EXIFON code, the threshold energy is given as 10 MeV, and both evaluated data (ENDF) show a close agreement with the calculated data.

Osmium-190 is a non-radioactive isotope of Osmium with natural abundance of 26.26%. It is used for cardiologic studies, laser frequency stabilization, and as a tracer in geological studies.

Fig 2 shows a transfer reaction Ir-193 (p,α) Os-190, for the production of Osmium-190, by transferring a proton nucleon between the beam and thereby accompanied by the emission of an alpha particle. From the above graph, the threshold incident energy was 14 MeV and the evaluated data appeared to have a peak cross-sectional value of 0.767 millibarn at an incident energy of 20 MeV.

Platinum-194 is a non-radioactive isotope of Platinum, useful for the production of backgroundless tritium targets. It is also useful for research and applications in mass cytometry, which is a mass spectrometry technique.

The reaction Ir-193 (p,γ) Pt-194 in Fig. 3 is a proton capture which shows a threshold incident energy of 7 MeV, which seems to be in close value with the evaluated data (ENDF).

The reaction Ir-193 (p,2n) Pt-192 in Fig. 4 is a break-up reaction, where an energetic proton breaks up target nucleus Ir-193 leading to the production of Pt-192. From the graph in Fig.4 above, the threshold incident energy is about 8 MeV and it closely agrees with the evaluated data (ENDF) given.

Figure 5 shows the excitation function for the production of Ir-192, produced from the reaction Ir-193 (p,np) Ir-192 and applied in brachytherapy. The graph shows a many-body reaction with a threshold incident energy of about 9 MeV while the calculated data rises with increasing cross-section before coming to rest.

Figure 6 shows the reaction Ir-193 (p,nα) Os-189 for the production of Osmium-189. The threshold energy for the production of Osmium-189, which is useful for research purposes in physics, is 13 MeV.

Platinum-191 is used as a target material for biological and biomedical labelling. Figure 7 shows a threshold incident energy for the Ir-193 (p,3n) Pt-191 spallation reaction as 1 MeV for the production of Platinum-191. The evaluated data (ENDF) appeared to have a peak cross-section of 1119.76 millibarns at an incident particle energy of 7 MeV.

Table 1: Nuclear Data Summary for Exifon with and without shell corrections compared with the ENDF data.

REACTION	PRODUCT	THRESHOLD ENERGY (MeV)				MAXIMUM CROSS SECTION (mb)			
		EXIFONwt	EXIFONwot	ENDF (MENDL)	ENDF (TENDL)	EXIFONwt	EXIFONwot	ENDF (MENDL)	ENDF (TENDL)
193Ir (p,p) 193Ir	Iridium-193	10	10	-	10	215.6	316.5	-	96.42
193Ir (p,α) 190Os	Osmium-190	20	20	-	13	0.7	1.4	-	0.767
193Ir (p,g) 194Pt	Platinum-194	7	7	-	6	3.9	4.3	-	0.793
193Ir (p,2n) 192Pt	Platinum-192	9	9	7	7	1300	1301	1140	893.2
193Ir (p,np) 192Ir	Iridium-192	12	12	-	8	1.2	1.9	-	118
193Ir(p,nα) 189Os	Osmium-189	16	16	-	13	0.6	1.4	-	9.04
193Ir (p,3n) 191Pt	Platinum-191	1	1	1	1	1293.8	1294.2	909	1119.76

Table 1 shows the threshold energies and maximum cross-sections for different proton interactions with Iridium-193. The evaluated data ENDF were gotten for both MENDL (Medium Energy Nuclear Data Library) and TENDL (TALYS-based Evaluated Nuclear Data Library). The complete threshold energies and maximum cross-sections for both EXIFON, MENDL and TENDL nuclear data were gotten for the reactions 193Ir(p,2n)192Pt and 193Ir(p,3n)191Pt only, while the other reactions Ir-193(p,p)Ir-193, Ir-193(p, α)Os-190, Ir-193 (p, g) Pt-194, Ir-193(p, np)Ir-192, and Ir-193(p, na)Os-189 had only EXIFON and TENDL evaluated data.

5. CONCLUSION

The excitation function of the proton-induced nuclear reaction channels of ^{193}Ir (p,p) ^{193}Ir , ^{193}Ir (p, α) ^{190}Os , ^{193}Ir (p,g) ^{194}Pt , ^{193}Ir (p,2n) ^{192}Pt , ^{193}Ir (p,np) ^{192}Ir , ^{193}Ir (p, $n\alpha$) ^{189}Os , ^{193}Ir (p,3n) ^{191}Pt on iridium isotope were discussed. The reaction cross-section and excitation functions for the given reactions have been studied and evaluated in the incident energy range of 1 – 30 MeV. The evaluated nuclear data file (ENDF) results was compared with the calculated EXIFON code values. The obtained data will be very useful in producing the radioisotopes Osmium189, Osmium 190, Platinum191, Platinum 192, Platinum 194, Iridium 192, Iridium 193 with more efficiency and purity. These radioisotopes can be applied in nuclear medicine for diagnosis and therapy purposes. Providing alternative routes for the production of radionuclides using nuclear reactions will definitely reduce or stop the crisis accompanied with radioisotope shortages.

REFERENCES

- [1] O. Iwamoto, "Development of a Comprehensive Code for Nuclear Data Evaluation, CCONE, and Validation Using Neutron-Induced Cross Sections for Uranium Isotopes," *J. Nucl. Sci. Technol.*, vol. 44, no. 5, pp. 687–697, 2007, doi: 10.1080/18811248.2007.9711857.
- [2] I. T. Al-Alawy and R. S. Mohammed, "Nuclear Data Relevant to the Production of Radioiodine I-123, I-125 by Indirect Route and Medical Applications," *Int. Lett. Chem. Phys. Astron.*, vol. 63, pp. 90–97, 2016, doi: 10.18052/www.scipress.com/ilcpa.63.90.
- [3] Y. H. Asres, M. Mathuthu, and M. D. Birhane, "Analysis of reaction cross-section production in neutron induced fission reactions on uranium isotope using computer code COMPLET," *Appl. Radiat. Isot.*, vol. 139, no. November 2017, pp. 81–85, 2018, doi: 10.1016/j.apradiso.2018.04.025.
- [4] J. E. Escher, J. T. Burke, F. S. Dietrich, N. D. Scielzo, I. J. Thompson, and W. Younes, "Compound-nuclear reaction cross sections from surrogate measurements," *Rev. Mod. Phys.*, vol. 84, no. 1, pp. 353–397, 2012, doi: 10.1103/RevModPhys.84.353.
- [5] S. Cowell, P. Talou, T. Kawano, and M. B. Chadwick, "Evaluation of iridium (n,xn) reactions," 2007, doi: 10.1051/ndata:07571.
- [6] A. S. Voyles *et al.*, "Excitation functions for (p,x) reactions of niobium in the energy range of $E_p = 40\text{--}90$ MeV," *Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms*, vol. 429, no. June, pp. 53–74, 2018, doi: 10.1016/j.nimb.2018.05.028.
- [7] G. I. Kurmuş and M. Gönül, "Chronic radiodermatitis developing after iridium-192 exposure: A case report," *Cutan. Ocul. Toxicol.*, vol. 34, no. 3, pp. 242–244, 2015, doi: 10.3109/15569527.2014.944648.
- [8] T. Siddik, "Theoretical cross-sectional calculation of some structural fusion material on (n, α)-induced reactions," *Indian J. Phys.*, vol. 93, no. 7, pp. 921–925, 2019, doi: 10.1007/s12648-018-1349-3.
- [9] M. E. Korkmaz, O. Agar, and M. Yigit, "Investigation of spherical and cylindrical natural Iridium targets by photonuclear reaction," *EPJ Web Conf.*, vol. 154, pp. 1–3, 2017, doi: 10.1051/epjconf/201715401027.
- [10] M. G. Budak, M. Karadag, and H. Yücel, "Determination of effective resonance energy for the $^{193}\text{Ir}(n,\gamma)^{194}\text{Ir}$ reaction by the cadmium ratio method," *Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms*, vol. 373, pp. 85–90, 2016, doi: 10.1016/j.nimb.2016.03.026.
- [11] L. Yafpe, "The Spallation Of Iridium With Protons Of Energies 9-87 MeV G . R . Grant ' For personal use only . The purpose of the present investigation was twofold : (a) to investigate half-lives and other decay characteristics of the spallation products of iridi," vol. 41, 1963.
- [12] E. Akgun, E. Ozgenc, and E. Gundogdu, "Therapeutic Applications of Radiopharmaceuticals: An Overview," *Fabad J. Pharm. Sci.*, vol. 46, no. 1, pp. 93–104, 2021.
- [13] L. S. M. Ooi, S. S. M. Sun, H. Wang, and V. E. C. Ooi, "New mannose-binding lectin from the rhizome of sarsaparilla *Smilax glabra* Roxb. (Liliaceae)," *J. Agric. Food Chem.*, vol. 52, no. 20, pp. 6091–6095, 2004, doi: 10.1021/jf030837o.
- [14] M. S. Rosenthal, "Ethical issues in radioisotope shortages: Rationing and priority setting," *J. Nucl. Med. Technol.*, vol. 38, no. 3, pp. 117–120, 2010, doi: 10.2967/jnmt.110.081117.
- [15] K. Iida, A. Kohama, and K. Oyamatsu, "Formula for proton-nucleus reaction cross section at intermediate energies and its application," *J. Phys. Soc. Japan*, vol. 76, no. 4, pp. 1–6, 2007, doi: 10.1143/JPSJ.76.044201.
- [16] I. Ahmad and F. S. Koki, "Calculation of Reactions Cross Section for Neutron-Induced Reactions on ^{127}I Isotope," *Int. J. Med. Physics, Clin. Eng. Radiat. Oncol.*, vol. 06, no. 03, pp. 344–359, 2017, doi: 10.4236/ijmpcero.2017.63031.
- [17] J. R. Dunning, G. B. Pegram, G. A. Fink, and D. P. Mitchell, "Interaction of Neutrons with Matter," *Phys. Rev.*, vol. 48, no. 3, pp. 265–280, 1935, doi: 10.1103/PhysRev.48.265.

- [18] S. Yamoah and M. Asamoah, "Calculations of Excitation Functions of (n, p), (n, α) and (n, 2n) Reaction Cross-Sections for Stable Isotopes of from Reaction Threshold to 20 MeV," *J. Nucl. Part. Phys.*, vol. 2013, no. 4, pp. 100–107, 2013, doi: 10.5923/j.jnpp.20130304.07.
- [19] S. Hilaire, "Statistical Nuclear Reactions," no. April, p. 105, 2000.
- [20] H. Kalka, *A model for statistical multistep reactions (code EXIFON)*, vol. 21, no. 23. International Atomic Energy Agency, 1990.