

**STUDY OF REACTION CROSS-SECTIONS FOR
ADVANCED REACTORS AND ASTROPHYSICAL
APPLICATIONS**

A SYNOPSIS SUBMITTED FOR THE PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY
IN PHYSICS

BY

VIBHUTIBEN R. VASHI
(REGISTRATION NO: FOS/2185)

UNDER THE GUIDANCE OF

Dr. RAJNIKANT MAKWANA



DEPARTMENT OF PHYSICS

FACULTY OF SCIENCE

THE MAHARAJA SAYAJIRAO UNIVERSITY OF BARODA

VADODARA - 390002, GUJARAT, INDIA.

DECEMBER 2022

● Introduction :

The studies related to nuclear technology including the design of nuclear reactors, fusion devices, and accelerators as well as nuclear astrophysics research about the origin of the chemical elements and other nuclear research such as nuclear medicines, national securities, and basic and industrial applications depend on nuclear data [1,2]. Nuclear data can be defined in different ways. The definition of nuclear data for the reactor community is typically the cross-sections and related information; the information about the nuclear levels and properties for the nuclear structure study group; the information of alpha, gamma, and beta radiation for radio chemists, etc [3]. The present study is focused on the cross-section measurement of nuclear reactions for the advancement of nuclear reactors and technology as well as for astrophysical applications.

Energy is an important part of powering human technologies and for sustainable economic growth. The electricity generated from conventional fossil fuels (coal) contaminates the climate by emitting CO₂ in the air and it is the biggest environmental challenge affecting the planet and humanity. Moreover, it should be noted that the reserves of coal are also limited. Also, the secure energy supply at the most affordable prices in the context of increasing energy requirements is a challenge for developing countries like India. Therefore, a more accessible, sustainable, and affordable low-carbon energy power option is required. If we explore all possible facilities to generate electricity, the most inevitable energy resource is nuclear power [4, 5]. The advanced nuclear reactors provide clean and reliable energy with advanced safety options and optimized cost-effective designs. These reactors are the backbone of our carbon-free future. Till today, ten percent of the world's electricity is generated by nuclear power but to control climate change, we need a greater amount of clean and reliable energy.

The collaborative efforts to develop next-generation nuclear energy systems are taken to provide the future energy needs of the world. The Uranium based fast

reactors [6–8], Thorium based advanced heavy water reactor (AHWR) [9, 10], Uranium based compact and high-temperature reactors (CHTR) and Th-U cycle based accelerator driven sub-critical systems (ADSs) [11–13] are the most important candidates of the present time to enhance the nuclear power generation to fulfil the increasing energy demand. Nowadays, the hybrid ADS reactors are promising due to their capability of incinerating long-lived minor actinides (Np, Am, Cm etc.) and fission product (^{129}I , ^{135}Cs , ^{99}Tc , ^{93}Zr etc) originated in conventional reactors and also produce the nuclear energy [14, 15]. The concept of an ADSs is to couple a fission reactor to a particle accelerator and the basic process is nuclear transmutation. In this system, the high-energy proton beam was delivered from the accelerator and bombarded on a target to produce a very intense neutron source called the spallation process. These neutrons contribute to transmuting fission products. In addition to neutrons, the other particles such as protons, alpha particles, and fission fragments will be produced with energy covering the full range up to the GeV range [16]. In recent years, the reactions using these particles became more interesting for finding out suitable materials for the mentioned applications and for withstanding radiation [17]. Therefore, the cross-section data of not only neutrons but also charged-induced reactions are important for the fabrication of different components of advanced reactors such as shielding design, estimation of waste and radiation damage, nuclear heating, transmutation effects, and radiation dose. Also, charged particle-induced reactions are important for the development of medical accelerators.

Besides, designing nuclear reactors, the nuclear data are required to address the quest of an interdisciplinary branch of Physics: nuclear astrophysics such as the origin of the chemical elements, the inner working of our sun, the evolution and explosions of stars, and the origin, composition, age, and ultimate fate of the Universe [18]. These puzzles could be solved by getting deeper knowledge about nuclear processes in an astrophysical environment. E. M. Burbidge, G. R. Burbidge, W. A. Fowler, and F. Hoyle (B^2HF) [19] as well as A. G. W. Cameron [20] independently came up

with the ideas of nucleosynthesis of the chemical elements in 1957. There are eight astrophysical processes such as the Hydrogen burning phase, Helium burning phase (Triple α process), α process, e process, s process, r process, **p** process and x process that can explain the origin of the all known stable and unstable isotopes [19]. Among them, the s process, p process, and r process are responsible for the nucleosynthesis of elements heavier than iron [21].

The present work is a contribution to the astrophysical p process. There are about 35 nuclei near the proton-rich side (neutron deficient nuclei) valley of β stability lie between ^{74}Se being the lightest and ^{196}Hg the heaviest, called p nuclei, are bypassed by the reaction paths of both r and s processes [22]. The origin of these nuclei can be explained by several processes summarized as p process [23–25]. The p process includes various nuclear reactions like photodisintegration of already created s and r nuclides via (γ, p) , (γ, n) , and (γ, α) also their inverse capture reactions are involved [27]. To estimate theoretically the abundance distribution, and the synthesis of p nuclei requires an extended reaction network calculation involving 20,000 reactions on 2000 stable and unstable nuclei [26]. The origin of the p nuclides is challenging due to there being no direct observations found in the stars and supernova remnants, as their contribution to elemental abundances is small and no element is dominated by a p isotope. There is no clear evidence found whether the p process is a single or multiple independent astrophysical scenarios. There are number of assumptions are made for the production of p nuclei like type II supernovae [28], the νp process in neutrino driven winds of SNII [29], type Ia supernovae [30], the rp -process in matter accreted on the surface of neutron stars [24], and others [26]. By considering these quests, it's a small contribution from our side to gain knowledge about the astrophysical p process reaction network.

• Motivation and Objectives :

Due to the significant importance of neutron-induced reactions for fundamental research in nuclear physics and astrophysics as well as in nuclear reactors demand improved and high-precision cross-section data. During the literature survey, it is found that the cross-section database for neutron-induced reactions for some reactions is rather scarce or discrepancy is observed in the literature [34]. Therefore, our aim is to measure accurate neutron cross-section data to fulfil the requirement of data for nuclear reactors advancement.

Further, the literature survey of astrophysical p process suggests that it is necessary to improve the understanding of the driving reaction mechanism behind the origin of the chemical elements which is the fundamental quest but it is complex in nature and involves the interplay of different research fields. Also, numerous experimental efforts are carried out in recent years for charged particle (proton, α) induced reactions [31–33] but still data are insufficient. Therefore, the aim of our present work is to contribute to the existing database of measured cross sections relevant to the astrophysical p process and to check the validation of the statistical model code calculation for an extensive set of nuclides. This is how the uncertainties in the abundance of the p process arising from the input can also be constrained. Also, the proton-induced reactions important for the advanced reactors are also insufficient [34].

Basically, the multidisciplinary work has been carried out by measuring proton-induced reaction cross sections, required for nuclear reactors and astrophysics. Also, proton-induced reactions are important for medical sectors and other applications are mentioned.

The following objectives have been fulfilled in the present work :

- The neutron-induced reactions were studied for $(n, 2n)$, (n, n') , (n, f) reaction channels upto 20 MeV of neutron energy. The quasi monoenergetic neutrons

were used to carry out the experiment. The neutrons were generated by $^7\text{Li}(p, n)$ reaction at BARC-TIFR pelletron facility, Mumbai, India. The neutron activation analysis technique (NAA) was used for data analysis. The neutron flux was calculated using flux monitor reactions. The reaction channels are studied for Au, In, U, and Th samples which are widely used for neutron flux calculations and for other purposes in nuclear reactors.

- The proton-induced reactions were studied on natural Cadmium target. The studied reactions are important for advanced nuclear reactors and astrophysical applications. Also, these reactions have significant importance in the medical sector. The stacked foil activation technique was used for the analysis. The 16 MeV of proton beam was delivered from BARC-TIFR pelletron facility, Mumbai, India.

Table 1: The spectroscopic data of selected neutron and proton induced nuclear reactions.

reaction	E(level) (MeV)	Isotopic abundance [38] (%)	Threshold energy [39] (MeV)	Product nucleus	J^π	Decay mode	Half-life [40]	Prominent γ -ray energy [40] keV	branching intensity (%)
$^{197}\text{Au}(n, 2n)$	0.0	100	8.114	^{196}Au	2^-	ϵ : 93.00 % β^- : 7.00 %	6.183 d	355.73 (5)	87
$^{115}\text{In}(n, n')$	0.3362	95.71	0.0	^{115m}In	$\frac{1}{2}^-$	IT : 95.00 % β^- : 5.00 %	4.486 h	336.24 (25)	45.9 (1)
$^{232}\text{Th}(n, f)$	0.0	100	0.0	^{97}Zr	$\frac{1}{2}^+$	β^- : 100.00 %	16.91 h	743.36 (3)	93
$^{238}\text{U}(n, f)$	0.0	99.2745	0.0	^{97}Zr	$\frac{1}{2}^+$	β^- : 100.00 %	16.91 h	743.36 (3)	93
$^{114}\text{Cd}(p, \gamma)$	0.3362	28.73	0.0	^{115m}In	$\frac{1}{2}^-$	IT : 95.00 % β^- : 5.00 %	4.486 (4) h	336.24 (25)	45.9 (1)
$^{114}\text{Cd}(p, n)$	0.1903	28.73	2.247	^{114m}In	5^+	IT : 96.75 % ϵ : 3.25 %	49.51 (1) d	190.27 (3)	15.56 (15)
$^{112}\text{Cd}(p, \gamma)$	0.3917	24.13	0.0	^{113m}In	$\frac{1}{2}^-$	IT : 100.00 %	99.476 (23)m	391.698 (3)	64.94 (17)
$^{110}\text{Cd}(p, n)$	0.0	12.49	4.703	^{110g}In	7^+	ϵ = 100.00 %	4.92 (8) h	937.478 (13)	68.4 (19)
$^{110}\text{Cd}(p, n)$	0.0621	12.49	4.703	^{110m}In	2^+	ϵ = 100.00 %	4.92 (8) h	657.75 (5)	97.74
$^{110}\text{Cd}(p, 2n)$	0.0	12.49	12.829	^{109g}In	$\frac{9}{2}^+$	ϵ = 100.00 %	4.159 (10) h	203.3 (1)	74.2
$^{110}\text{Cd}(p, 2n)$	0.6501	12.49	12.829	$^{109m1}\text{In}$	$\frac{1}{2}^-$	IT = 100.00 %	1.34 (7) m	649.8 (2)	93.51 (9)
$^{110}\text{Cd}(p, 2n)$	2.1018	12.49	12.829	$^{109m2}\text{In}$	$\frac{19}{2}^+$	IT = 100.00 %	0.209 (6) s	673.52 (8)	97.6 (3)

m \rightarrow minute, h \rightarrow hour, d \rightarrow day, y \rightarrow year

- The experimental work consists of the irradiation of the targets mentioned in

the table followed by their gamma ray spectrometric analysis. The off-line gamma ray counting of the irradiated samples was performed using a High Purity Germanium (HPGe) Radiation detector connected to a 4096 channel analyzer and precalibrated using ^{152}Eu source.

- The simulation of the experimental data was carried out with the Hauser-Feshbach statistical model prediction, using TALYS [35], EMPIRE [36], and ALICE [37] codes.
- Further, the theoretical study has been also carried out using TALYS, EMPIRE, and ALICE codes, for the deuteron induced reactions on some nuclear materials of the periodic table which are important in various fields of nuclear physics and technology as well as in nuclear medicines.

• Outline of the Thesis :

The present work is bifurcated in the following chapters in the thesis.

Chapter 1 - An overall view of the nuclear reaction mechanism is discussed in this chapter. The importance of the nuclear data of charged particle and neutron-induced reactions in advanced nuclear reactors is discussed. A brief history of nuclear astrophysics and then the astrophysical processes contributing to the origin of the chemical elements are presented. The importance of nuclear data for astrophysical applications is discussed.

Chapter 2- A detailed description of the experimental arrangement and methodology of the BARC-TIFR facility utilized to carry out the neutron and proton-induced nuclear reactions is presented. The activation analysis technique used for the cross-section measurement is discussed in this chapter.

Chapter 3 - A brief overview of the statistical nuclear model codes used for the present simulation is included in this chapter. The statistical nuclear model codes such as TALYS, EMPIRE, and ALICE used for theoretical cross-section predictions

and a particle transport code MCNP used to calculate proton energy degradation have been discussed.

Chapter 4 - A complete description of the methodology was followed to study the neutron-induced reactions for Au, In, Th, and U samples for the respective reaction channels $^{197}\text{Au}(n, 2n)$, $^{115}\text{In}(n, n')$, $^{232}\text{Th}(n, f)$, and $^{238}\text{U}(n, f)$ is discussed. The importance of the studied reactions in the nuclear reactor has been portrayed. The chapter contains the detail of the experimental set-up, cross-section measurement technique, and theoretical calculations followed by obtained results and discussion.

Chapter 5 - The proton-induced reactions were studied for the application of advanced nuclear reactors and astrophysics. A detail of the standard approximation of the Gamow window for astrophysical interest followed by experimental methodology, and data analysis. The detail of the different nuclear models from the statistical nuclear model codes used for the present study has been discussed. The S-factor determination is also presented. Finally, the discussion of the obtained results.

Chapter 6 - A systematic study of charged particle induced nuclear reactions has been carried out for some of the elements used for the particle accelerator and nuclear reactors using the statistical nuclear model codes TALYS, EMPIRE, and ALICE.

Chapter 7 - This chapter contains the summary and conclusion obtained through the results of the neutron and charged particle-induced nuclear reactions. Also, the future scope of such kind of work is discussed.

Bibliography

- [1] R. A. Forrest, Energy Procedia **7**, 540-552 (2011).
- [2] Michael S. Smith, Journal of Nuclear Science and Technology, Supplement **2**, 19-24 (August 2002).
- [3] R. B. Firestone, Overview of nuclear data, Lawrence Berkeley National Lab.(LBNL), Berkeley, CA (United States) (2003).
- [4] S. S. Kapoor, Pramana-J. Phys **59**, 941 (2002).
- [5] P. K. Vijayan *et al.*, Progress in Nuclear Energy, Volume **101**, 43-52 (2017).
- [6] L. Mathieu *et al.*, “Proportion for a very simple Thorium Molten Salt reactor, in Proceedings of the Global International Conference”, Tsukuba, Japan, Paper No. **428** (2005).
- [7] Fast Reactors and Accelerator Driven Systems Knowledge Base, IAEA-TECDOC-1319: Thorium fuel utilization: Options and Trends (Nov. 2002).
- [8] A. Nuttin *et al.*, Proc. Nucl. Energy, **46**(1):77-99 (2005).
- [9] R. K. Sinha, and A. Kakodkar, Nucl. Eng. Des. **236**, 683 (2006)
- [10] S. Ganesan, Creation of Indian Experimental Benchmarks for Thorium Fuel Cycle, IAEA Coordinated Research Project on Evaluated Data for Thorium-Uranium fuel Cycle, Third Research Co-ordination Meeting, 30 January to 2 February 2006, Vienna, Austria, INDC (NDS) - 0494 (2006).

- [11] F. Carminati, R. Klapisch, J. P. Revol, Ch. Roche, J. A. Rubio and C. Rubbia, An Energy Amplifier for Cleaner and Inexhaustible Nuclear Energy Production Driven by Particle Beam Accelerator, CERN Report No. CERN/AT/93-47 (ET) (1993).
- [12] C. Rubbia, J. A. Rubio, S. Buono, F. Carminati, N. Fietier, J. Galvez, C. Geles, Y. Kadi, R. Klapisch, P. Mandrillon, J. P. Revol and Ch. Roche, Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier, CERN Report No. CERN/AT/95-44 (ET) (1995).
- [13] E. D. Arthur, S. A. Schriber and A. Rodriguez (Editors), The International Conference on Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, Nevada, USA, (1994), AIP Conf. Proc., Vol. **346** (1995).
- [14] E. Andrade-II *et al.*, <http://arxiv.org/abs/1608.07501v1>.
- [15] C. D. Bowman *et al.*, Nucl. Instr. Meth. A **320**, 336, (1992).
- [16] F. -R. Lecolley *et al.*, in International Conference on Nuclear Data for Science and Technology, 26 September - 1 October 2004, Santa Fe, edited by R. C. Haight, M B. Chadwick, T. Kawano, and P. Talou, AIP Conf. Proc. No. **769**, p. 61 (AIP, New York, 2005).
- [17] T. Allen *et al.*, Mater. Today **13**, 14 (2010).
- [18] Michael S. Smith *et al.*, International Conference on Nuclear Data for Science and Technology (2007).
- [19] E. M. Burbidge *et al.* Rev. Mod. Phys. **29**, 547 (1957).
- [20] A. G. W. Cameron, Publ. Astron. Soc. Pac. **69**, 201 (1957).
- [21] S. Goriely, Journal of Nuclear Science and Technology, Supplement **2**, p. 536-541 (August 2002).

- [22] D. L. Lambert, *Astron. Astroph. Rev.* **3**, 201 (1992).
- [23] S. E. Woosley and W. M. Howard, *Astrophys. J. Suppl.* **36**, 285 (1978).
- [24] H. Schatz *et al.*, *Phys. Pep.* **294**, 167 (1998).
- [25] S. Goriely, J. José, M. Hernanz, M. Rayet, M. Arnould, *Astron. Astroph.* **383**, L27 (2002).
- [26] M. Arnould, and S. Goriely, *Phys. Rep.* **384**, 1 (2003).
- [27] A. C. Larsen *et al.*, *Phys. Rev. C* **93**, 045810 (2016).
- [28] M. Rayet, N. Prantzos, and M. Arnould, *Astron. Astrophys.* **227**, 271 (1990).
- [29] S. Wanajo, H. -T. Janka, and S. Kubono, *Astrophys. J.* **729**, 46 (2011).
- [30] C. Travaglio *et al.*, *Astrophys. J.* **739**, 93 (2011).
- [31] E. Somorjai *et al.*, *Astron. Astrophys.* **333**, 1112 (1998).
- [32] J. Bork *et al.*, *Phys. Rev. C* **58**, 524 (1998).
- [33] Gy. Gyürky *et al.*, *Phys. Rev. C* **68**, 055803 (2003).
- [34] IAEA-EXFOR Database, <http://www.nds.iaea.org/exfor>.
- [35] A. J. Koning, S. Hilaire, S. Goriely, TALYS user manual, A nuclear reaction program, NRG-1755 ZG PETTEN, The Netherlands, (2017).
- [36] M. Herman *et al.*, “EMPIRE (ver. 3.2.3): Nuclear Reaction Model Code System for Data Evaluation”, User’s Manual, INDC(NDS)-0603 (2015).
- [37] M. Blann, *Phys. Rev. Lett.* **27**, 337 (1971).
- [38] K. J. R. Rosman, and P. D. P. Taylor, *Pure Appl. Chem.* **70**, 217 (1998).

- [39] Qtool: calculation of reaction Q-values and threshold, Los Alamos National Library, Official website: http://cdfc.sinp.msu.ru/services/calc_thr/calc_thr.html.
- [40] NuDat 2.7 β 2011, National Nuclear Data Center, Brookhaven National Laboratory, Official website: <https://www.nndc.bnl.gov/nudat2/>.

List of Publications

Publications in Peer-Reviewed Journals

1. **Systematic study of (p, n) and (p, 2n) reactions on ^{110}Cd**
Vibhuti Vashi, Rajnikant Makwana, B. Quintana , M. H. Mehta, R. K. Singh, B. K. Soni, R. Chauhan, S. Mukherjee, M. Abhangi, S. Vala, N. L. Singh, G. B. Patel, S. V. Suryanarayana, B. K. Nayak, S. C. Sharma, T. N. Nag, and Y. Kavun, under review in Radiation Physics and Chemistry.
2. **Investigation of (d, 3n) reaction cross section using theoretical nuclear codes calculations on some nuclear materials**
Y. Kavun, **V. Vashi**, and R. Makwana, App. Rad. and Iso. **189**, 110426 (2022).
3. **Cross-section measurement of the $^{114}\text{Cd}(\text{p}, \gamma) ^{115\text{m}}\text{In}$ reaction for nuclear reactor and astrophysical applications**
Vibhuti Vashi, Rajnikant Makwana, B. Quintana , M. H. Mehta, B. K. Soni, S. Mukherjee, R. K. Singh, R. Chauhan, P. M. Prajapati, M. Abhangi, S. Vala, N. L. Singh, G. B. Patel, S. V. Suryanarayana, B. K. Nayak, S. C. Sharma, T. N. Nag, and Y. Kavun, Phys. Rev. C **105**, 044613 (2022).
4. **Measurement of cross sections for flux monitor reactions using quasi-monoenergetic neutrons**
Vibhuti Vashi, Rajnikant Makwana, S. Mukherjee, B. K. Soni, M. H. Mehta, S. Parashari, R. K. Singh, R. Chauhan, S. V. Suryanarayana, B. K. Nayak, S. C. Sharma, H. Naik, N. L. Singh, T. N. Nag, Eur. Phys. J. Plus **136**:746 (2021).

Publications in Proceedings of the National Conferences

1. **Theoretical study of $^{109}\text{Ag}(\alpha, n)$ reaction from the threshold up to 20 MeV**

V. R. Vashi, R. J. Makwana, and R. K. Singh, manuscript has been presented as poster in DAE Symposium on Nuclear Physics (2022).

2. **Measurement of isomeric cross-section at energy of astrophysical interest**

Vibhuti Vashi, R. Makwana, B. Quintana, M. Mehta, S. Mukherjee, B. Soni, M. Abhangi, S. Vala, N. Singh, R. Singh, G. Patel, A. Hingu, S. Suryanarayana, B. Nayak, S. Sharma, T. N. Nag, Proceedings of the DAE Symp. on Nucl. Phys. **65**, 498 (2021).

3. **Measurement of reaction cross-section for $^{197}\text{Au}(n, 2n)^{196}\text{Au}$ reaction**

Vibhuti Vashi, R. Makwana, S. Mukherjee, B. Soni, M. H. Mehta, S. Parashari, R.K. Singh, S.V. Suryanarayana, B.K. Nayak, S.C. Sharma, H. Naik, and Taraknath, Proceedings of the DAE Symp. on Nucl. Phys. **64**, 381 (2019).

Other publications

1. **Cross-sections for production of ^{115m}In by quasi-monoenergetic neutrons within 7-20 MeV**

Akash Hingu, Bhargav Soni, Siddharth Parashari, Rajnikant Makwana, P.M. Prajapati, **Vibhuti Vashi**, Mayur Mehta, R. Palit, S.V. Suryanarayana, B.K. Nayak, K. Katovsky, S. Mukherjee, Radiation Physics and Chemistry **199**, 110270 (2022).

2. **Experimental and theoretical cross sections of $^{115}\text{In}(n, n')$ reaction at 19 and 16 MeV using quasi-monoenergetic neutrons**

Akash Hingu, Bhargav Soni, S. Mukherjee, Rajnikant Makwana, Siddharth

Parashari, **Vibhuti Vashi**, Mayur Mehta, R. Palit, S. V. Suryanarayana, Proceedings of the DAE Symp. on Nucl. Phys. **65**, 357 (2021).

3. **Gamma-ray shielding study of different concrete compositions as a reactor shielding**

Bhargav Soni, Rajnikant Makwana, S. Mukherjee, S. S. Barala, **V. Vashi**, S. Parashari, Rakesh Chauhan, K. Katovsky, Proceedings of the DAE Symp. on Nucl. Phys. **64**, 944 (2019).