Investigation of Neutron Induced Reaction Cross sections for Nuclear Data Applications

A Synopsis Submitted to The

The Maharaja Sayajirao University of Baroda

For The Degree of

Doctor of Philosophy in Science

(Physics)

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June-2022

I. Introduction

Studying fast neutron-induced reactions, especially at higher incident energies, is interesting in fundamental and applied physics. The accurate cross-section data are needed for many practical applications, including nuclear power, transmutation of radioactive waste, nuclear medicine, non-proliferation, stockpile stewardship, and fundamental physics. These applications rely on nuclear phenomena such as reactions, decay, and structure. The neutron-induced reactions can be used to describe reaction mechanisms and examine nuclear structure by restricting nuclear models in fundamental nuclear physics. Neutron activation analysis is a particularly effective approach because neutrons may penetrate deep into materials, providing information on the bulk rather than just the surface. The total production cross-sections of the residual nucleus are generally obtained for ground-state reaction products, although isomeric state production cross sections also explore our understanding of the level and decay structure of the residual nucleus [1]. Application of the activation technique is particularly fruitful if crosssections are 1 mb or more, the radioactive product has a half-life of several minutes to several days and its decay is accompanied by the emission of a gamma-ray in the range from 100 keV to several MeV and an intensity of 10% or more. In such cases, the overall irradiation time, the total counting time, and the sample transfer time between irradiation and activity determination are very favorable, allowing relatively large-scale measurement programs in a short time. Furthermore, the easy-to-use, highly selective, and well-established high-purity germanium (HPGe) spectrometry can be used to determine the activity [2].

Nuclear reactions are responsible for nucleosynthesis, or the production of elements throughout the Universe. The^{2}H, ^{3}He, and particles are the most common products of Big-Bang nucleosynthesis. The development of early stars, which create elements up to Fe, follows this initial nucleosynthesis. Different mechanisms, like neutron capture, neutrino-induced reactions, explosive events in supernovae, and the rapid-neutron process in neutron star mergers, are used to create heavier elements. The neutron energy range between 1 keV and 1 MeV is essential in astrophysics because it corresponds to the temperature regimes of the important areas for synthesizing all nuclei between iron and actinides.Apart from the astronomical purpose, there is continued interest in neutron cross sections for practical applications, such as the neutron balance in modern reactors aiming for high burn-up rates and concepts dealing with radioactive waste transmutation. Determining(n, xn) reaction cross sections areessential for developing fast reactors since the neutron balance in the reactor core is affected by the neutron multiplication caused by such reactions. At moderately high temperatures, the $(n, 2n)$ channel may dominate the (n, γ) reaction channel. This might be important in the nucleosynthesis of neutron rich isotopes and, in the r-process nucleosynthesis. Therefore, testing the assumption that multiparticle emission is negligible at astrophysics relevant energy is necessary. For this reason, measurements of the cross section for the $(n, 2n)$ reaction channel are important [3-4].

One of the most critical issues to resolve for public acceptability of nuclear fission energy generation is developing and controlling long-term radioactive danger. ADS (Accelerator Driven Subcritical Systems) for nuclear waste transmutation offers a viable solution to this challenge. The system, consisting of a sub-critical reactor and a spallation neutron source generated by a high-power accelerator with a heavy metal target, is fundamentally safe. This will be assured by online monitoring of the sub-criticality level that is both reliable and accurate. However, several obstacles must be overcome before the initial notion of ADS can be achieved and its viability established. This is significant in the context of reactor physics and neutronics, where nuclear data uncertainties play a substantial role in determining many-core and fuel cycle parameters.Nuclear research reactors are crucial to the advancement of nuclear science and technology. They're utilized in fundamental research, radioisotope manufacturing, neutron scattering, radiography, and material characterization and testing, among other applications [5-6].

Since the early 1980s, several large and excellent measurement campaigns were conducted around 14 MeV to facilitate the knowledge of cross sections relevant to the fusion community. For higher energies, the interest in the range above 14 MeV and up to several GeV is a consequence of the study of accelerator-driven systems (since early 1990) and, more recently, of the design of the IFMIF materials irradiation facility for the study of radiation damage in fusion reactors (maximum energy 55 MeV). Even for such systems, the primary energy range of relevance in a large part of the facility is below 20 MeV.The outcomes of the experiments may be used to investigate different statistical model codes and confine the parameter sets they used. Such research is also expected to shed light on the reaction mechanisms in various energy regions. The information will be used to construct the International Fusion Material Irradiation Facility (IFMIF), which will test materials for fusion power plant technologies. Despite substantial improvements in the quality of nuclear data, such as neutron interaction cross sections, little information on nuclear data uncertainties and much less on nuclear data covariance are available. In nuclear applications, covariance data is necessary to examine design parameter uncertainties [7] accurately.The available database above 14 MeV is minimal, reflecting the lack of need for such data from traditional fission and fusion reactor development. Around 14 MeV, many measurements were performed because of the widespread use of neutron generators, but often these data show discrepancies and large scatter. Studies of cross sections and isomeric crosssection ratio of neutron threshold reactions are of considerable significance for testing nuclear models and practical applications. The improved experimental data on neutron induced charged particle (n,p) and (n,α) reactions on structural materials areessential and critical for defining processes like brittlement,nuclear heating, induced radioactivity, nuclear transmutation rates, and damage caused by hydrogen and helium generation in structural materials [8].

The elements V, Cr, Ti, and Cu-based alloys have excellent properties that make them an essential structural material for reactor technology. In the fusion reactor, vanadium is considered the reactor structural material for the first wall/blanket applications due to the low activation properties of the vanadium alloys. Similarly, copper as a first wall material has been considered in reactor designs with high thermal loads on the first wall or requires a shield of high electrically conductive material surrounding the plasma to help stabilize its location. Furthermore,because of their remarkable characteristics, Titanium and chromium alloys are desirable structural materials for fusion reactors. The activation data on titanium and chromium is important for practical applications in fusion reactor technology ~e.g., estimation of activity level, hydrogen and helium gas production, nuclear heating, and radiation damage since chromium is an important constituent of structural steel. The elements ^{78}Se and ^{80}Se are also used as targets to produce ^{77}Br and ^{80m}Br, which are therapeutic radioisotopes. The(n,p) reaction produces arsenic isotopes, which are poisonous to humans and other living creatures. Cancer and other serious health problems occur due to the arsenic element.The $(n,2n)$ reaction cross sections of the ^{121}Sb, ^{123}Sb, and ^{103}Rh isotopes are essential for neutron multiplication calculations. Some of the antimony isotopes in nuclear fission have been identified as nuclides of the fission product. Therefore, fast neutron induced cross section measurements with better accuracy for antimony are essential for improving nuclear data. Threshold reactions, including (n,n') and $(n,2n)$ have been used extensively for determining the differential flux (dQ/dE) from neutron sources by foil activation techniques. The cross sections of (n,xn) reactions are necessary for activation detectors which are used to probe energy components of neutron fluence. An example of such a detector is rhodium, which is monoisotopic.Thus, it is essential to study the higher energy neutron induced reaction cross section for Rh, Sb, V, Cr, Ti, Cu, and Se elements from an application point of view.

II. Nuclear database

The nuclear data community maintains many databases for the use of its members as well as for the application communities. The suggested values for the evaluated data libraries are based on an expert review. There are also unevaluated libraries and databases where experimental data and computations are assembled and available for usage within and beyond the community. There are different data libraries which are mentioned below:

1. **Experimental Nuclear Reaction Data (EXFOR)**
2. **Evaluated Nuclear Data File (ENDF)**
3. **Reference Input Parameter Library (RIPL-3)**

The EXFOR (EXchange FORmat) database includes data on cross section reaction experiments, outgoing particles, multiplicities, fission product yields, and other topics. The EXFOR began as a standard format for exchanging nuclear measurement results between multiple data centres. It has subsequently grown into a vast measuring database containing data from over 23,000 tests. The EXFOR compilation normally follows the original references closely, although it may contain extra material or fixes for issues identified after publication. This experimental data can be used to evaluate and enhance theoretical models of neutron, photon, and charged particles induced reactions [9].

The Evaluated Nuclear Data File (ENDF), the Japanese Evaluated Nuclear Data Library (JENDL), the Fusion Evaluation Nuclear Data Library (FENDL), the Chinese Evaluation Neutron Data Library (CENDL), Talys Evaluation Nuclear Data Library (TENDL),and the Joint Evaluated Fission and Fusion File (JEFF) are the most often used evaluated libraries. The National Nuclear Data Center (NNDC) in the United States, the Japan Atomic Energy Agency (JAEA), and the Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD) manage ENDF, JENDL, and JEFF, respectively (OECD). The Atlas of Neutron Resonances is a distinct collection with assessed resonance parameters. These libraries are the most up-to-date representations of nuclear data observables [10-15].

Furthermore, the Reference Input Parameter Library (RIPL) is a database containing essential information for reaction calculations. This comprises ENSDF-evaluated structural data and reaction model inputs like the optical potential and fission barriers. Based on the reviewed structural information, experimental data, and theory work, this library proposes values or models for reaction calculations. This library is a single database that contains most of the nuclear data required to execute the reaction calculation algorithms and ensures that the calculations are accurate and consistent [16].

III. Literature Survey and Objective

The neutron induced (n, p) and $(n, 2n)$ reactions cross sections for Se, Cu, Ti, Cr, V, Rh, and Sb elements are taken from the experimental EXFOR database and evaluated data from the ENDF database. The measured cross section shows large discrepancies in the available experimental and evaluated data at the same incident neutron energies. More experimental data seems necessary for energies below 13 and above 15 MeV. The measured cross section data is scarce for neutron energy greater than 15 MeV.However, the previous measurement was restricted to energy values around 14 MeV incident neutron energies. To validate the theoretical estimations and improve the parameterization of the statistical model calculations, the experimental mapping of the $(n, 2n)$ and (n, p) excitation function is needed for an extended energy region.The reference data for the neutron flux estimation was the ^{27}Al$(n, a)^{24}$Na reaction cross sections that was known with an accuracy of 0.4–0.8 percent. IRDFF ver. 1.05 [17] probably is the best choice for this purpose because it is one of the most recent evaluations (published on October 9, 2014) and includes extensive cross sections and uncertainty information.The previous measurements used a coincidence setup of two NaI(Tl) detectors to count the annihilation γ rays of the positrons from the β^{+} decay of the reaction product nucleus.The use of the NaI (T1) detector for γ-ray or β-ray counting has a resolution problem. The recent works use high-resolution HPGe γ-ray spectroscopy to measure the activated samples.The main objective of the present thesis is to study and investigate the neutron induced reaction cross sections on Se, Cu, Ti, Cr, V, Rh, and Sb elements.

IV. Plan of the Ph.D. thesis

This thesis aims to study the behavior of the excitation function of Se, Sb, V, Cu, Ti, Cr, and Rh elements at reaction threshold to 20 MeV incident neutron energies using the neutron activation and offline γ-ray spectrometry method. In the present work, the cross section of the $(n, p)$and$(n, 2n)$ reactions was measured experimentally at different neutron energies using the activation technique. Highenergy quasi-monoenergetic neutrons in the interesting MeVregime with a tail to lower energies are produced via the ^{7}Li(p, n) reaction at the 14UD TIFR-BARC Pellatron Accelerator Facility Mumbai, India. The Pellatron accelerator irradiates metallic lithium foil with energies up to22 MeVand continues proton beamcurrent up to 200(nA). This results in neutron energies rangingbetween 8 to 20 MeV.The samples were activated along with Al- monitor foil to determine the incident neutron flux. The activities induced by the reaction products were measured using a high-resolution HPGe spectrometry system. The data analysis was carried out using the latest decay data. Detail uncertainty propagation had been performed, and the measured cross sections are reported with their uncertainties and correlation coefficients. Statistical model calculations were performed using the reaction codes TALYS (ver. 1.95) and EMPIRE (ver. 3.2.3) from the reaction threshold to 25 MeV energies [18-19].

Additionally, the effects of various combinations of the theoretical level density, optical potential, preequilibrium models and γ-ray strength function were considered forthe reproduction of the experimental data. The input parameters needed in theoretical calculations to reproduce the present and previous measurements were taken from the RIPL-3 database. The present results are compared with the earlier measurements and latest evaluations of the ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0/HE, CENDL-3.2, TENDL-2019, JENDL-5.0, and FENDL-3.2b libraries and theoretical calculations using TALYS (ver. 1.95) and EMPIRE (ver. 3.2) codes. Furthermore, the cross section of the (n, p) and $(n, 2n)$ reactions was estimated within 14-15 MeV neutron energies using different systematic formulae. These estimated cross sections by various systematic formulae were compared with the previous experimental data.

V. The present Ph.D. thesis work is divided into seven chapters as follows:

➢ Chapter 1

Chapter 1 gives a broad overview of nuclear reactions and nuclear data. Neutron-induced reactions are given special attention, and their significance is explained in thischapter. Furthermore, the status of nuclear data and its use in various energy regions were discussed briefly. This chapter also includes an overview of the neutron-induced reaction cross sections and the uncertainties in the measured cross section.

➢ Chapter 2

Chapter 2 describes the irradiation experimental setup. The neutron facility and sample details are briefly stated in this chapter. Moreover, the experimental design used for irradiation as well as the HPGe detector setup used for activated measurement is described in this chapter.

➢ Chapter 3

Chapter 3 deals with the procedures for data analysis work in which the activation cross section calculation and uncertainties in the measured cross sections are described systematically using the covariance method. In addition, the different systematic formulae for the estimates of the (n, p) and $(n, 2n)$ reactions cross sections are included in this chapter.

➢ Chapter 4

Chapter 4illustrates the statistical model calculations based on the level density, optical potential, and pre-equilibrium models. The different nuclear reaction models and two different simulation codes TALYS (ver. 1.9) and EMPIRE (ver. 3.2.3) were used for the theoretical calculations, and details about the codes are explained in this chapter.

➢ Chapter 5

Chapter 5 discusses the neutron induced $(n, 2n)$ and (n, p) reactions cross sections for ^{103}Rh, ^{121}Sb, ^{123}Sb isotopes, and ^{51}V. The present results are compared with the previous measurements and latest evaluations of the ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0/HE, CENDL-3.2, TENDL-2019 and FENDL-3.2 libraries. We also compare the experimental results with theoretical estimates using TALYS (ver. 1.95) and EMPIRE (ver. 3.2.3) codes fromthe reaction threshold to the 25 MeV energy region. Furthermore, the (n, p) and $(n, 2n)$ reactions cross section of the ^{51}V and ^{121}Sb, ^{123}Sb isotopes are estimated within 14-15 MeV neutron energies using different systematic formulae are discussed in detail. These estimated cross sections by various systematic formulae are compared with the previous experimental data.

➢ Chapter 6

Chapter 6explains the neutron induced (n, p) reaction cross sections for $^{76, 77, 78, 80}$ Se,^{52}Cr, ^{48}Ti, and ^{65}Cu isotopes. The present results are compared with the previous measurements and latest evaluations of the ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0/HE, CENDL-3.2, TENDL-2019 and FENDL-3.2 libraries. The present experimental results also compare with the theoretical estimates using TALYS (ver. 1.95) and EMPIRE (ver. 3.2.3) codes from reaction threshold to the 25 MeV neutron energy region. Furthermore, the (n, p) reaction cross section of the ^{76,77,78,80} Se isotopes are estimated within 14-15 MeV neutron energies using different systematic formulae and discussed in detail.These calculated cross sections by various systematic formulae are compared with the previous experimental data.

➢ Chapter 7

Chapter 7 summarizes the results and conclusions of the research work described in the present thesis and proposals for future work indicated in this chapter.

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➢ List of peer-reviewed publications included in the present thesis

1. **Cross sections for the (n, p) reaction of selenium isotopes within 10.5 to 19.81 MeV neutron energies**

   R. K. Singh, N. L. Singh, R. D. Chauhan, Mayur Mehta, S. V. Suryanarayana, Rajnikant Makwana, S. Mukherjee, B. K. Nayak, H. Naik, J. Varmuza, K. Katovsky

   Eur. Phys. J. Plus (2021) 136:338 **Impact Factor: 3.911**

   https://doi.org/10.1140/epjp/s13360-021-01299-x

2. **Neutron induced reaction cross section of ^{51}V with covariance analysis**

   R. K. Singh, N. L. Singh, R. D. Chauhan, Mayur Mehta, S. V. Suryanarayana, Rajnikant Makwana, S. Mukherjee, B. K. Nayak, H. Naik, Tarak Nath Nag, J. Varmuza, K. Katovsky

   Eur. Phys. J. A (2021) 57:337 **Impact Factor: 3.043**

   https://doi.org/10.1140/epja/s10050-021-00638-x

3. **Systematic study of the $(n, 2n)$ reaction cross section for ^{121}Sb and ^{123}Sb isotopes**

   R. K. Singh, N. L. Singh, R. D. Chauhan, Mayur Mehta, S. V. Suryanarayana, Rajnikant Makwana, B. K. Nayak, H. Naik, Tarak Nath Nag, J. Varmuza

   Chin. Phys. C Vol. 46, No. 5 (2022) 054002 **Impact Factor: 2.720**

   https://doi.org/10.1088/1674-1137/ac4a5a

4. **Experimental and theoretical study of the ^{65}Cu$(n, p)^{65}$Ni reaction cross section from reaction threshold up to 25 MeV**

   R. K. Singh, N. L. Singh, Mayur Mehta, R. D. Chauhan, S. V. Suryanarayana, Rajnikant Makwana, B. K. Nayak, H. Naik, Tarak Nath Nag, J. Varmuza, K. Katovsky

   (Phys Rev. C under revision)

5. **Activation cross section for the $(n, 2n)$ and (n, p) reactions on ^{103}Rh, ^{48}Ti and ^{51}Cr from reaction threshold up to 25 MeV energy region**

   R. K. Singh, N. L. Singh, Mayur Mehta, R. D. Chauhan, S. V. Suryanarayana, Rajnikant Makwana, B. K. Nayak, H. Naik, Tarak Nath Nag, J. Varmuza, K. Katovsky

   (To besubmitted in journal)

➢ Other publications in peer-reviewed journals

6. **Cross-section of $(n, 2n)$ reaction for niobium and strontium isotopes between 13.97 to 20.02 MeV neutron energies**

   Mayur Mehta, N. L. Singh, **Ratankumar Singh,** Rakesh Chauhan, Rajnikant Makwana, S. V. Suryanarayana, H. Naik, P. V. Subhash, S. Mukherjee, Jan Varmuza, Karel Katovsky

   Applied Radiation and Isotopes **Impact Factor: 1.5**

   https://doi.org/10.1016/j.apradiso.2022.110142

7. **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 (2021) 136:746 **Impact Factor: 3.228**

   https://doi.org/10.1140/epjp/s13360-021-01673-9

8. **Study of $(n, 2n)$ reaction cross sections for ^{107}Ag within the energy range of 9–22 MeV**

   Rakesh Chauhan, **R. K. Singh,** N. L. Singh, Mayur Mehta, Rajnikant Makwana, S. V. Suryanarayana, S. Mukherjee, B. K. Nayak, H. Naik, J. Varmuza, K. Katovsky

   Eur. Phys. J. Plus (2021) 136:532 **Impact Factor: 3.228**

   https://doi.org/10.1140/epjp/s13360-021-01449-1

9. **Measurement of ^{90}Zr$(n, 2n)^{89}$Zr and ^{90}Zr$(n, p)^{90m}$Y reaction cross sections in the neutron energy range of 10.95 to 20.02 MeV**

   Mayur Mehta, N. L. Singh, **R. K. Singh,** Siddharth Parashari, P. V. Subhash, H. Naik, R. D. Chauhan, R. Makwana, S. V. Suryanarayana, S. Mukherjee, A. Gandhi, J. Varmuza, K. Katovsky

   Journal of Radioanalytical and Nuclear Chemistry (2021) 328:71–81

   Impact Factor: 1.137

   https://doi.org/10.1007/s10967-021-07625-y

10. **Measurement of (n, γ) reaction cross section of ^{186}W isotope at neutron energy of 20.02±0.58 MeV**

Mayur Mehta, N. L. Singh, R. Makwana, P. V. Subhash, S. V. Suryanarayana, S. Parashari, Rakesh Chauhan, **R. K. Singh,** H. Naik, S. Mukherjee, B. Soni, S. Khirwadkar, J. Varmuza & K. Katovsky

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Impact Factor: 0.653

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11. **Measurement of neutron induced ^{86}Sr$(n, 2n)^{85}$Sr reaction cross sections at different neutron energies**

    Nidhi Shetty, Rajnikant Makwana, Mayur Mehta, S. Mukherjee, N. L. Singh, S. V. Suryanarayana, S. Parashari, **R. Singh,** H. Naik, S. C. Sharma, S. Ayyala, B. Soni, R. Chauhan

    Applied Radiation and Isotopes 154 (2019) 108866 **Impact Factor: 1.270**

    https://doi.org/10.1016/j.apradiso.2019.108866

12. **Elastic scattering for ^{6}Li + ^{51}V and systematic study of breakup threshold anomaly**

    H. Kumawat, C. Joshi, V. V. Parkar, V. Jha, B. J. Roy, Y. S. Sawant, P. C. Rout, E. T. Mirgule, **R. K. Singh**, N. L. Singh, B. K. Nayak, S. Kailas

    Nuclear Physics AVol. 1002, October 2020, 121973 **Impact Factor: 1.695**

    https://doi.org/10.1016/j.nuclphysa.2020.121973

13. **Exploring breakup coupling effect in ^{7}Li + ^{92,100}Mo elastic scattering around Coulomb barrier energies**

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    Eur. Phys. J. A (2022) 58:40 **Impact Factor: 3.043**

    https://doi.org/10.1140/epja/s10050-022-00690-1

14. **Cross-section measurement of the ^{114}Cd$(p, \gamma)^{115m}$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) **Impact Factor: 3.296** https://doi.org/10.1103/PhysRevC.105.044613

15. Inclusive α production for the ^{6}Li + ^{51}V system

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Phys. Rev. C 105, 034615 (2022) **Impact Factor: 3.296**

https://doi.org/10.1103/PhysRevC.105.034615

- ## Publications in National and International Conference Proceedings

1. **Measurement of reaction cross-section for ^{197}Au$(n, 2n)^{196}$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, Tarak Nath

   Proceedings of the DAE Symp. On Nucl. Phys. Vol. 64, B31-381 (2019).

2. **Neutron nuclear data of $(n, 2n)$ reaction for Sb Isotopes**

   R K Singh, R. D. Chauhan, S. V. Suryanarayana, Rajnikant Makwana, S. K. Mukherjee, Mayur Mehta, Tarak Nath, Bhargav Soni

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3. **Measurement of ^{100}Mo$(n, 2n)^{99}$Mo reaction cross sections using 10-20 MeV quasi-monoenergetic neutrons**

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4. **Production cross-section of the ^{99m}Tc medical isotope by using the ^{nat}Mo$(p, 2n)$ reaction**

   Siddharth Parashari, S. Mukherjee, S. V. Suryanarayana, R. Makwana, B. K. Nayak, **Ratan K. Singh,** S. C. Sharma, M. Mehta, N. L. Singh, H. Naik

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5. **Measurement of the ^{115}In$(n, 2n)^{114m}$In reaction cross-section using the quasi-monoenergetic neutrons**

   Bhargav K. Soni, Siddharth Parashari, S. Mukherjee, S. V. Suryanarayana, R. Makwana, B. K. Nayak, **Ratan K. Singh,** S. C. Sharma, M. Mehta, N. L. Singh, and H. Naik

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6. **Measurement of neutron induced reaction cross-sections for ^{86}Sr at different neutron energies**

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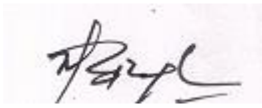

30th June 2022

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