

Chapter 1

Introduction

1.1 Fundamentals of Nuclear Reactor Physics

Towards 21st century, the world-wide increasing demand of energy and electricity called for a re-evaluation of presently available energy sources over a longer period of time. With this, the important goal is for the development of advanced systems which are cost-effective, environmental friendly and efficient [1]. Renewable energy like hydropower, wind, solar and geothermal are most favorable sources due to advantages like less emission of greenhouse gases, lower maintenance and easy to implementation. Despite the advantages of the renewable sources; these sources have some disadvantages like poor energy conversion ratio, high cost and they are not available year around. In order to improve the standard of living for everyone, researchers have to develop new energy sources.

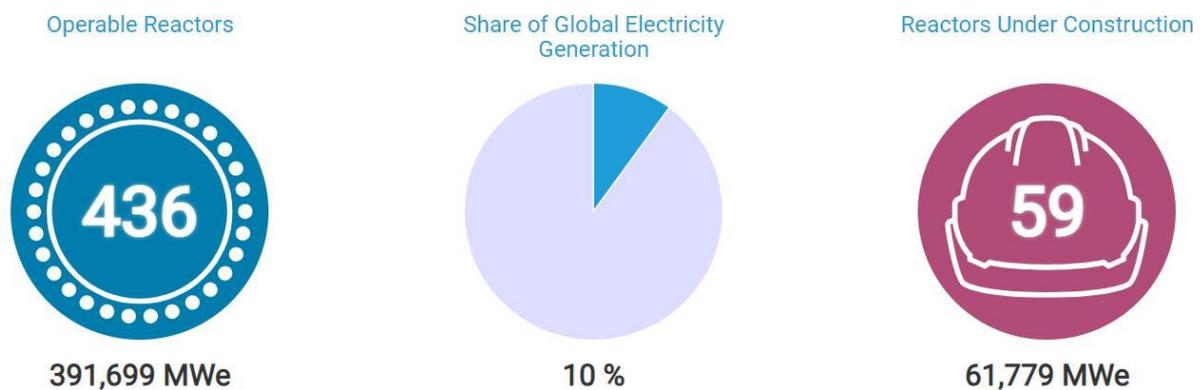


Figure 1.1 Worldwide reactor database

Fission reactors: Currently, about more than 430 fission reactors are in operational conditions which supplies about 10-15% of the world's electricity in around 30 countries as presented in *Figure 1.1* [2]. In fission reactors, the energy is produced by a controlled nuclear chain reaction. In this process, the uranium-235 ($^{235}_{92}\text{U}$) atom absorbs a neutron which splits into fast-moving fission products releasing energy and free neutrons as shown in *Figure 1.2*. These free neutrons

will be absorbed by other ^{235}U atoms and trigger other fission events. The collision of these fission products releases the energy which is converted into the thermal energy and utilized for the energy production in the fission reactor [3-5]. However fission reactor has two major limiting factors in the operation, (i) availability of nuclear fuel is limited and (ii) production of radioactive waste. This perspective called for a requirement of an effective reactor and fuel cycle strategies including fusion reactor and accelerators [6-10].

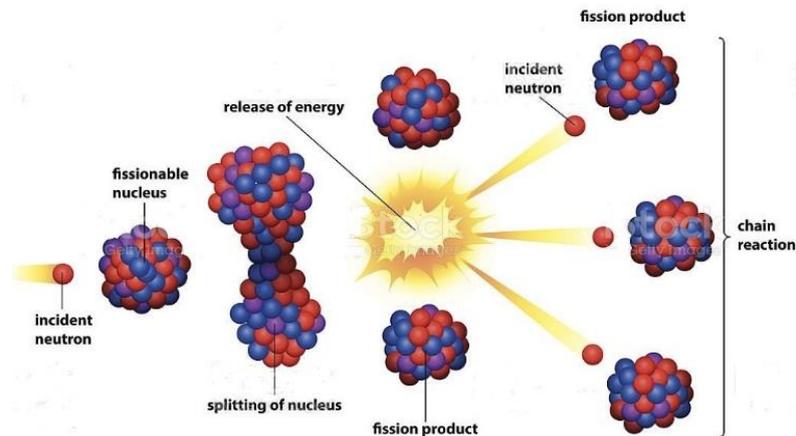
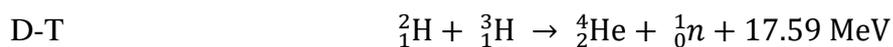
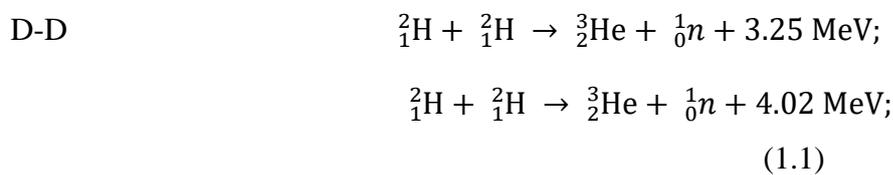


Figure 1.2 Chain reaction in fission reactor

Fusion reactors: The most favorable example of fusion around us is the stars and the Sun which are burning for billions of years using this process. Fusion reactions take place in a plasma in which two or more light atomic nuclei, usually deuterium (D) and tritium (T), combine and fuse together to form heavier nuclei with releasing massive amount of energy. The widely considered fusion reactions based on hydrogen isotopes are deuterium-deuterium (D-D) and deuterium-tritium (D-T) in which the least difficult fusion reaction is D-T reaction because it is feasible at lower temperature than the D-D reaction.



Fusion reaction process only occurs at a very high temperature which provide enough energy to overcome the electrostatic repulsion and fuse together. The temperature will reach about 10-15 times higher than the temperature in the centre of the sun. In DT- reaction, 3.5 MeV α -particle (helium nucleus); 14.1 MeV neutrons are produced and in total 17.59 MeV energy is

released per fusion reaction. This energy is released in the form of kinetic energies of product nuclei which could be converted into electricity using conventional technology. An advantage of fusion reaction is that it is virtually limitless, safe, clean and affordable. With these advantages, fusion reactions have two major challenges: (i) heating of fusion fuel up to tens of million degree temperature, and (ii) confinement of fuel in the central area of the reactor. The fusion reactions, however, has a disadvantage that these are charged particle reactions. Thus to overcome the coulomb repulsion of the positively charged particles, very high kinetic energy is required to interact nuclei with the particles which is achieved by using particle accelerators.

Accelerators: A variety of accelerators are available which operates on a different technical principles. The basic principle of the accelerators is that the interaction of the charged particles to very high speeds and energies with electromagnetic fields dividing them in two types: (i) Electrostatic Accelerators uses electrostatic fields to accelerate particles e.g. Van de Graff and Cockcroft Walton Generators; (ii) Electrodynamic or Electromagnetic accelerators uses changing electromagnetic fields to accelerate particles e.g. Cyclotron, Betatron. Accelerators are useful in nuclear physics experiments, particle interaction studies as well as medical and industrial applications. In present thesis work, the experimental work has been carried out by using charged particle accelerators which is discussed in detailed in next chapter 2.3.

1.2 Reactor Materials

A major challenge in construction and development of a nuclear reactor is the selection and design of its structural materials. High performance structural materials are essential to the development and design of the planned fusion reactors as well as to the advancement of the fission reactors that are now in operation. The materials are subjected to extreme conditions of unprecedented fluxes of high-energy neutrons, and intense thermo mechanical stresses. This environment will degrade the reactor materials which leads to reduce the performance and cause the sudden failure of the reactors [11]. The energetic neutrons in fission-fusion reactor will damage the materials and produce radio-activities by nuclear transmutation. In fission reactor, a diverse range of structural materials are used as presented in *Figure 1.3*. The degradation in mechanical and chemical properties of the materials is observed due to radiation induced defects in the reactor components. Similarly in the operation condition at 300 to 350 °C, the piping and heat exchanger experience thermal aging and complex water chemistry issues may induce corrosion and stress corrosion cracking.

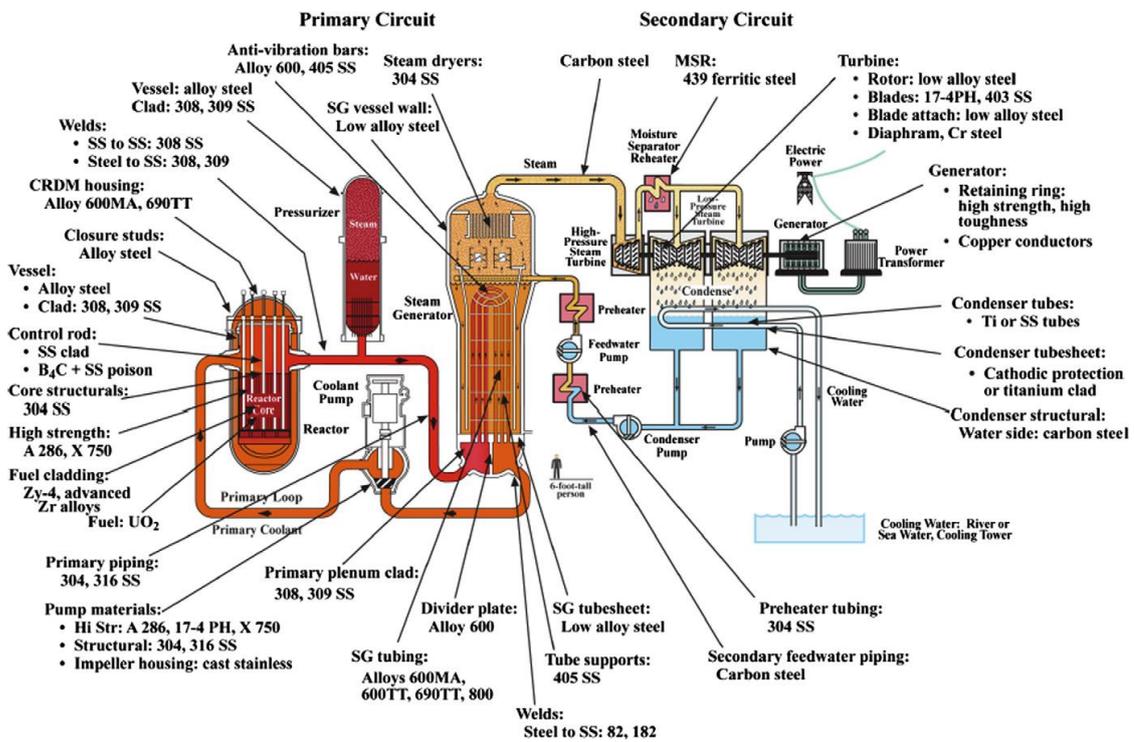


Figure 1.3 Schematic diagram of fission reactor with materials [3]

Figure 1.4 [12] depicts a schematic representation of a fusion reactor in which the initial wall, blanket, and divertor components absorb powerful neutrons produced by the DT-reaction. The materials that optimize the radiation-induced breakdown process will yield substantially larger quantities of transmutant due to the high energy neutrons. Furthermore, it is suggested that the initial wall and divertor components will encounter a heat flux of 1 to 10 MW/m² under steady state conditions. This is greater than the maximum heat flux that the structural materials in a fission reactor would be subjected, which is approximately 1 MW/m² for fuel cladding. Therefore there is a challenge for researcher to develop the variety of structural, tritium generation and reactor coolant materials are about five times higher than the core structural materials for existing fission reactors [13, 14]. In case of Accelerator Driven System (ADS), materials are divided into fuel, structural and coolant materials categories. The major issue in this case is the development of fuel due to lack of experimental work to characterize the basic properties of the materials, their fabrication process and their behavior under high temperature and high flux (proton and neutron) environment [15-17].

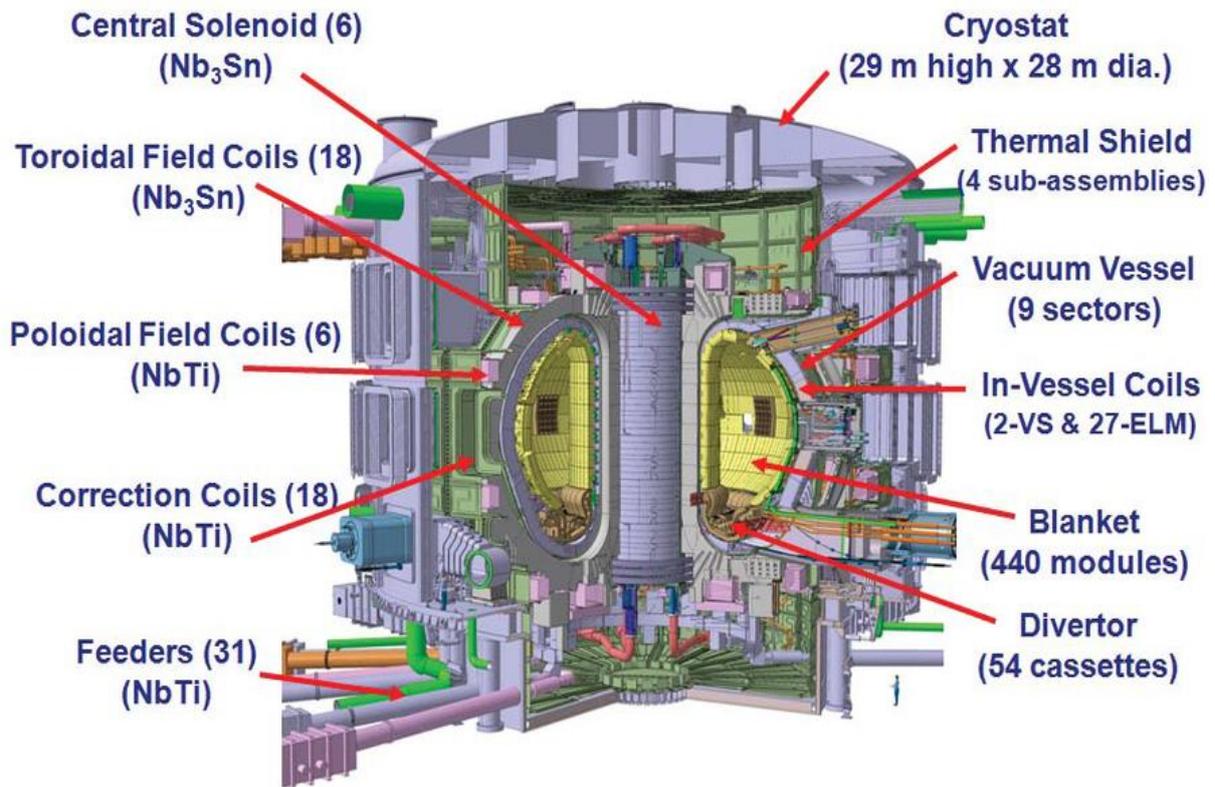


Figure 1.4 Schematic diagram of fusion reactor with structural materials [5]

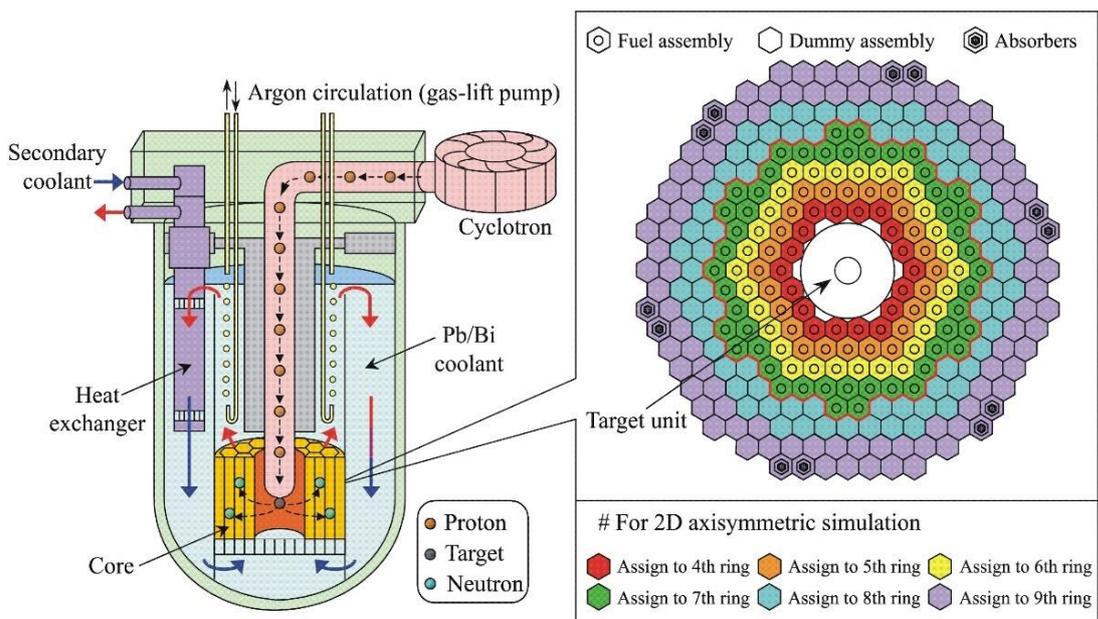


Figure 1.5 Schematic diagram of Accelerator Driven System (ADS) [6]

1.3 Neutron interactions

As discussed above, neutrons are produced during the operation of nuclear reactors. These neutrons interact with the surrounding materials which produced more neutrons and other particles through different reaction channels. *Figure 1.6* shows the type of reactions when a neutron interacts with a nucleus [18].

- **Elastic Scattering- (n,n):** A neutron interacts with a nucleus and bounces off in a different direction by transfer of some energy to it. Sometimes the nucleus absorbs a neutron and re-emits it. The fraction of energy transfer of neutrons depends on the angle of the collision with the nucleus. The target nucleus gains the energy and moves with an increased speed.
- **Inelastic Scattering (n,n γ):** A high energy neutron collides with heavy nuclei, neutron is absorbed to form a compound nucleus which will be in excited state. It de-excites by emitting neutron of lower energy with a γ -photon of remaining energy.
- **Radiative Capture (n, γ):** The most common nuclear reaction in which the compound nucleus formed by emission of γ -photon. In other words, the product nucleus has mass number increased by one.
- **Charged Particle (n,p) (n, α):** A nucleus absorbs a neutron to form a compound nucleus which de-excites by emitting a charged particle like proton or alpha particle. This type of reaction is also called as transmutation. In this type of reaction one element transforms into another by nuclear reaction.
- **Fission (n,f):** The most important reaction presented in *Table 2.1* of the next chapter.

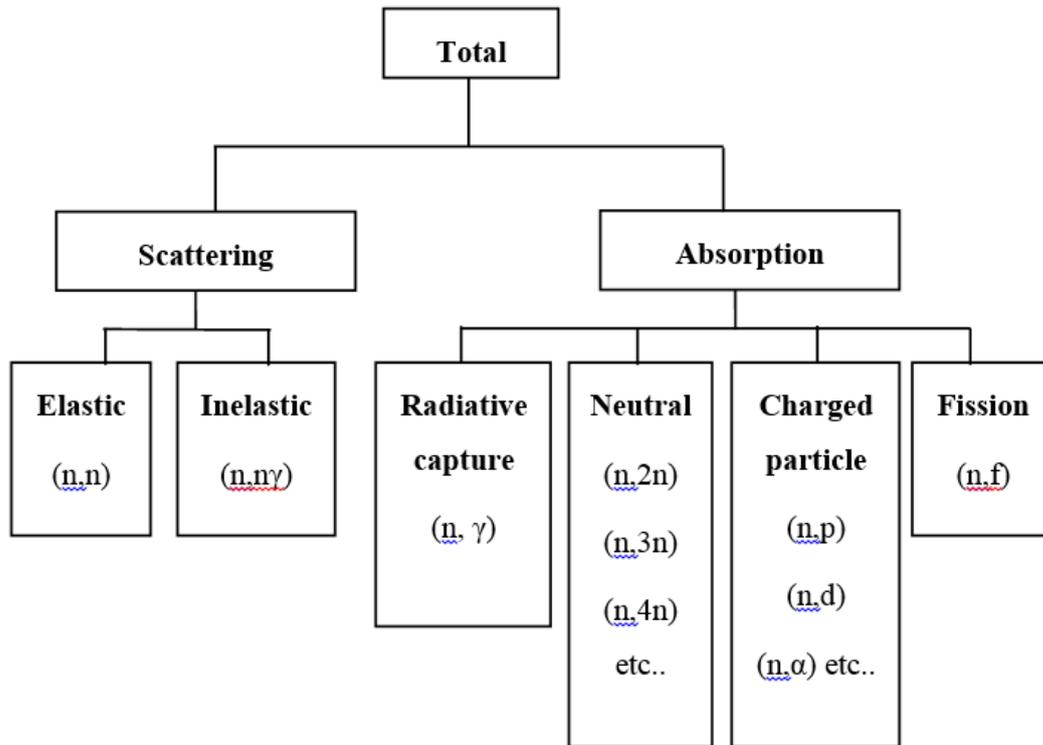


Figure 1.6 Types of reactions induced by neutron interaction

1.4 Requirement of Nuclear data

The accurate nuclear data are fundamentally important for the study of operation and performance of fission-fusion reactors and accelerators. Therefore a wide range of nuclear data such as cross section and decay data of reactor materials are required in a wide range of energies (10^{-5} eV to 20 MeV standard range but extended to 200 MeV) to study the neutron and particles interactions mechanism, transmutation studies, material analysis useful for reactor applications and other general applications like nuclear medicine, radiation therapy, nuclear waste management studies etc. [19-21]. Moreover the quality of the reported data is enumerated by the uncertainties of the parameters involved in the measurements and hence researchers should be aware of all the sources of uncertainties and the correlation between them. In data evaluation, researchers find difficulties due to scarcity and lack of information of the uncertainties in the experiments [22, 23]. In present thesis work, the (n,γ) , (n,p) and $(n,2n)$ reactions cross section were determined for different materials (Tungsten, Zirconium, Niobium, Strontium and Rubidium) in the fast neutron energy range used in fission-fusion

reactors and accelerators applications. A detailed uncertainty calculation of all the parameters involved were presented using covariance analysis method.

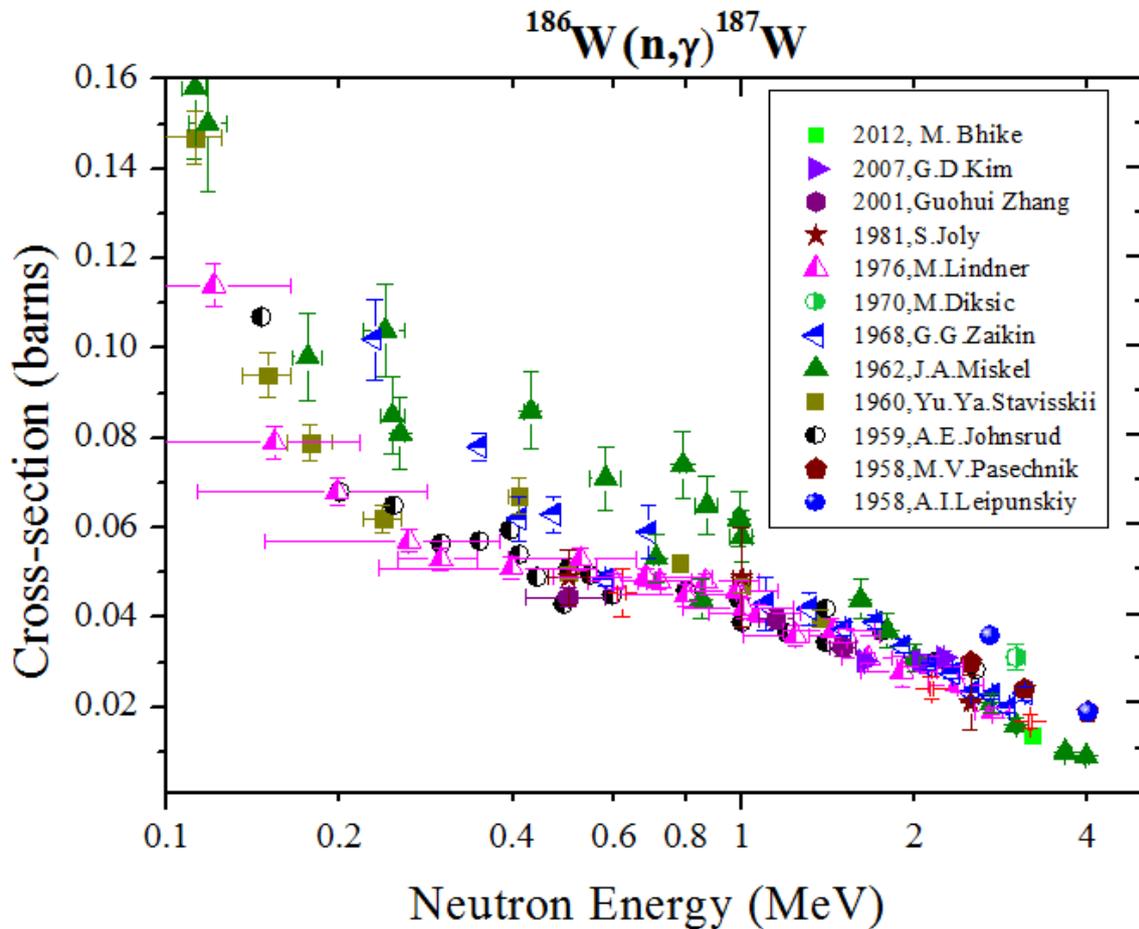


Figure 1.7 Reported radiative capture cross section data for $^{186}\text{W}(n,\gamma)^{187}\text{W}$ reaction

The neutron radiative capture reaction cross section for tungsten isotope in the fast neutron energy range is presented in Figure 1.7 with previously published data retrieved from EXFOR database [24-35]. The database shows that the multiple cross section data available up to 3 MeV have discrepancies whereas in the neutron energy range from 4 to 20 MeV there is a scarcity in the reaction cross section data.

The (n,2n) and (n,p) reactions cross section for isotope of zirconium [36-52] was and rubidium [53-67] were presented in Figure 1.8 and Figure 1.9 and the (n,2n) reaction cross section for isotope of niobium [68-74] and strontium [75-84] was presented in Figure 1.10 in the neutron energy range from threshold to 20 MeV.

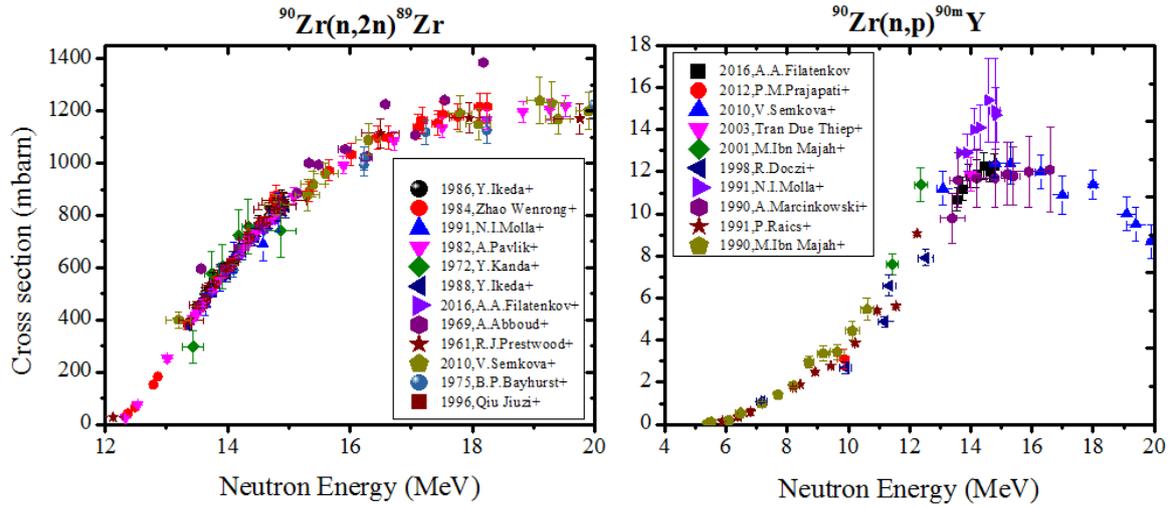


Figure 1.8 Reported cross section data for $^{90}\text{Zr}(n,2n)$ and $^{90}\text{Zr}(n,p)$ reactions

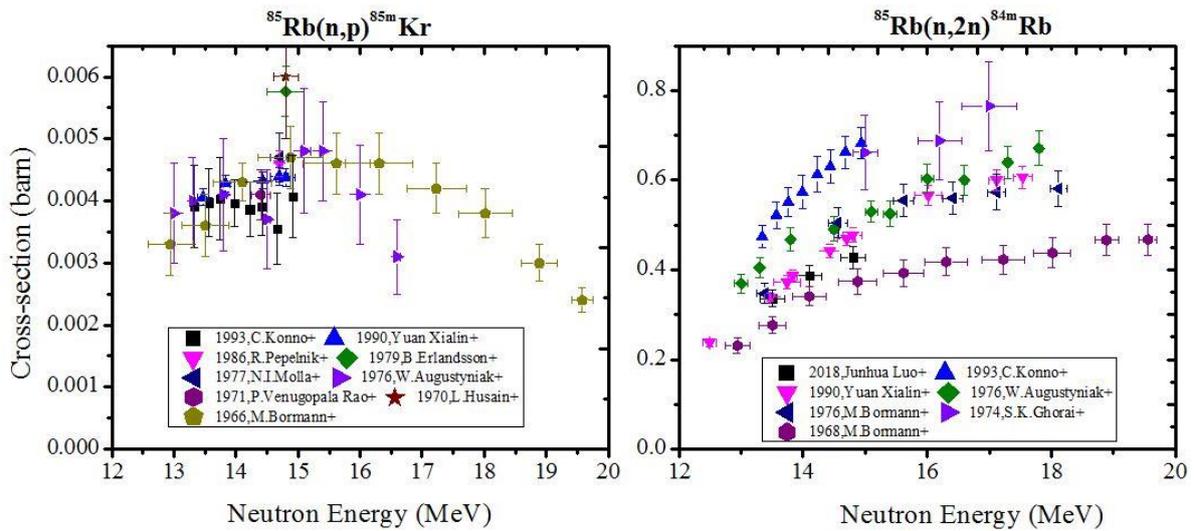


Figure 1.9 Reported cross section data for $^{85}\text{Rb}(n,2n)$ and $^{85}\text{Rb}(n,p)$ reactions

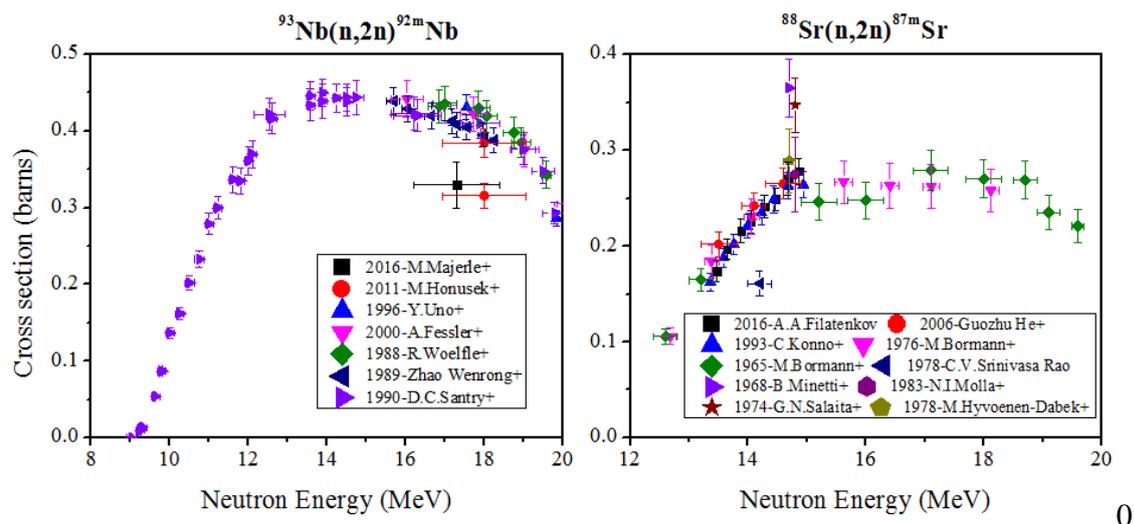


Figure 1.10 Reported cross section data for $^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$ and $^{88}\text{Sr}(n,2n)^{87m}\text{Sr}$ reactions

1.5 Objectives of present thesis

The objectives fulfilled in this doctoral thesis are as follow:

- To measure (n,γ) reaction cross section of ^{186}W isotope in the fast energy range at FOTIA and BARC-TIFR facilities. The analysis was carried out by offline γ -ray spectroscopic technique. The mono-energetic neutrons were produced by $^7\text{Li}(p,n)^7\text{Be}$ reaction.
- To measure the $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ and $^{90}\text{Zr}(n,p)^{90m}\text{Y}$ reactions cross section in the neutron energy range from 10 to 20 MeV. The covariance analysis was also carried out to calculate the uncertainties and correlations.
- To measure the $^{85}\text{Rb}(n,2n)^{84m}\text{Rb}$ and $^{85}\text{Rb}(n,p)^{85m}\text{Kr}$ reaction cross section in the neutron energy range 10 to 20 MeV. The absolute flux value during the experiments was determined from the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ monitor reaction.
- The reactions cross section of selected materials were predicted by using TALYS and EMPIRE codes by using different input parameters like level density models, γ -strength function and other nuclear models.

References

- [1]. OECD, Nuclear Energy Agency, Nuclear Energy in a Sustainable Development Perspective, Paris (France), 2000.
- [2]. WNA Pocket Guide on Reactors, 2009.
- [3]. PRIS-Home, <https://pris.iaea.org/pris>
- [4]. W. Oldekop, (1982), Electricity and Heat from Thermal Nuclear Reactors, Primary Energy, Berlin, Heidelberg: Springer Berlin Heidelberg, pp. 66–91.
- [5]. DOE Fundamentals Handbook: Nuclear Physics and Reactor Theory (PDF). US Department of Energy. Archived from the original (PDF) on 23 April 2008. Retrieved 24 September 2008.
- [6]. P. Kaw and I. Bandyopadhyay, Fusion Physics, Chapter 1: The Case for Fusion. International Atomic Energy Agency, 2012, ch. 1, 125 pp. 1–58. [Online]. Available: <http://www-pub.iaea.org/books/IAEABooks/8879/Fusion-Physics>
- [7]. Elmer E. Lewis, Fundamentals of Nuclear Reactor Physics 2008, Pages 1-27, Chapter 1- Nuclear Reactions.
- [8]. J. Ongena and Y. Ogawa, Nuclear fusion: Status report and future prospects, Energy Policy, 96 (2016) 770-778. Available online at <http://www.sciencedirect.com/science/article/pii/S0301421516302658>
- [9]. Alejandro Garcia, Experimental Tools II: Particle Accelerators, Encyclopedia of Nuclear Energy (2021) 146-152.
- [10]. Helmut Wiedemann, Particle Accelerator Physics, Springer Berlin, Heidelberg , eBook ISBN: 978-3-540-49045-6, 2007, <https://doi.org/10.1007/978-3-540-49045-6>
- [11]. G. S. Was, Journal of Nuclear mater, 276 (2000) 123.
- [12]. Retrieved online site from <https://www.iter.org/>
- [13]. Steven J Zinkle, J.T. Busby, Structural Materials for Fission & Fusion Energy Structural Materials Represent the Key for Containment of Nuclear Fuel. Materials Today 12 (11) (2009) 12-19.

- [14]. S. J. Zinkle, G. S. Was, Materials challenges in nuclear energy, *Acta Materialia* 61 (2013) 735-758.
- [15]. Organisation for Economic Co-Operation and Development, Nuclear Energy Agency, 75 - Paris (France) (2002). Accelerator-driven systems (ADS) and fast reactors (FR) in advanced nuclear fuel cycles. Nuclear Energy Agency of the OECD (NEA): Organization for Economic Co-Operation and Development - Nuclear Energy Agency. <https://www.oecd-nea.org/ndd/reports/2002/nea3109-ads.pdf>.
- [16]. Accelerator Driven System mechanism, <http://www/iket.kit.edu/221.php>
- [17]. Hamid Aït Abderrahim, Didier De Bruyn, Gert Van den Eynde, Rafaël Fernandez , Accelerator Driven Subcritical Systems, *Encyclopedia of Nuclear Energy*, Elsevier, 2021, Pages 191-202, <https://doi.org/10.1016/B978-0-12-819725-7.00093-3>.
- [18]. <https://sites.ifi.unicamp.br/mabernal/files/2014/09/NeutronInteractions.pdf>
- [19]. R. A. Forrest, *Energy Procedia* 7 (2011) 540-552.
- [20]. R. A. Forrest, *Fusion Engineering and Design* 81 (18) (2006) 2143-2156.
- [21]. S. Ganesan, *Pramana Journal of Physics* Vol. 68 (2) (2007) 257-268.
- [22]. N. Otuka et al., *Radiation Physics and Chemistry* 140 (2017) 502.
- [23]. Rebecca Pachuau, A. Gandhi, Namrata Singh et al., *Chinese Physics C* 47 (7) (2023) 074001.
- [24]. M. Bhike, B. J. Roy, A. Saxena, R. K. Choudhury, S. Ganesan, *Nuclear Science and Engineering*, 170 (2012) 44.
- [25]. G. D. Kim, H. J. Woo, H. W. Choi, N. B. Kim, T. K. Yang, J. H. Chang, K. S. Park, *Journal of Radioanalytical and Nuclear Chemistry*, 271 (2007) 553.
- [26]. Guohui Zhang, Zhaomin Shi, Guoyou Tang, Jinxiang Chen, Guangzhi Liu, Hanlin Lu, *Nuclear Science and Engineering*, 137 (2001) 107.
- [27]. J. Voignier, S. Joly, G. Grenier, D.M. Drake, L. Nilsson, Report: Centre d'Etudes Nucleaires, Saclay Reports No.5089, NSR-Key No: 1981VOZW (1981).
- [28]. M. Lindner, R. J. Nagle, J. H. Landrum, *Nuclear Science and Engineering* 59 (1976) 381.
- [29]. M. Diksic, P. Strohal, G. Peto, P. Bornemisza-Pauspertl, I. Hunyadi, J. Karolyi, *Acta Physica Hungarica*, 28 (1970) 257.

- [30]. G. G. Zaikin, I. A. Korzh, N. T. Sklyar, I. A. Totskii, Soviet Atomic Energy 25 (1968) 1362.
- [31]. J. A. Miskel, K. V. Marsh, M. Lindner, R. J. Nagle, Physical Review 128 (1932) 2717.
- [32]. Yu.Ya. Stavisskii, V.A. Tolstikov, Journal of Nuclear Energy A&B (Reactor Sci. and Technol.) 16 (1960) 496.
- [33]. A. E. Johnsrud, M. G. Silbert, H. H. Barschall, Physical Review 11 (1959) 927.
- [34]. M. V. Pasechnik, I. F. Barchuk, I. A. Totskiy, V. I. Strizhak, A. M. Korolev, Yu. V. Gofman, G. N. Lovchikova, E. A. Koltypin, G. B. Yankov, Conf.: Second Internat. At. En. Conf., Geneva, Vol.15 (1958) 18.
- [35]. A. I. Leipunskiy, O. D. Kazachkovskiy, G. Ja. Artyukhov, A. I. Baryshnikov, T. S. Belanova, V. I. Galkov, Yu. Ja. Stavisskiy, E. A. Stumbur, L. E. Sherman, Conf.: Second Internat. At. En. Conf., Geneva, Vol.15 (1958) 50.
- [36]. A. A. Filatenkov, Neutron activation cross sections measured at KRI in neutron energy region 13.4 - 14.9 MeV, Report: USSR report to the I.N.D.C.No.0460, 2016.
- [37]. V. Semkova, E. Bauge, A.J.M. Plompen, et al., Nuclear Physics A 832 (2010) 149.
- [38]. Qiu Jiuzi, Yuan Junqian, Yang Jingkan, Journal of Lanzhou Univ., Natural Science Ed., 32 (1997) 57.
- [39]. A. Pavlik, G. Winkler, H. Vonach, et al., Jour. of Physics, Part G (Nucl. and Part. Phys.) 8 (1982) 1283.
- [40]. Y. Kanda, Nuclear Physics A, 185 (1972) 177.
- [41]. Zhao Wenrong, Lu Hanlin, Fan Peiguo, Chinese J. of Nuclear Physics (Beijing), 6(1) (1984) 80.
- [42]. N. I. Molla, R. U. Miah, M. Rahman, et al., Conf. on Nucl. Data for Sci. and Technol., Juelich (1991) 355.
- [43]. Y. Ikeda, C. Konno, K. Oishi, et al., Report: JAERI Reports No.1312 (1988).
- [44]. Y. Ikeda, H. Maekawa, T. Nakamura, et al., Radiation Effects 92 (1986) 175.
- [45]. B. P. Bayhurst, J. S. Gilmore, R. J. Prestwood, et al., Physical Review C 1 (1975) 451.
- [46]. A. Abboud, P. Decowski, W. Grochulski, et al., Nuclear Physics A 139 (1969) 42.

- [47]. P. Raics, S. Nagy, S. Szegedi, N.V. Kornilov, A.B. Kagalenko, Conf. on Nucl. Data for Sci. and Technol., (Juelich 1991) 660.
- [48]. A. Marcinkowski, U. Garuska, H.M. Hoang, et al., Nuclear Physics A 510 (1990) 93.
- [49]. M. IbnMajah, A. Chiadli, S. Sudar, et al., Applied Radiation and Isotopes 54 (2001) 655.
- [50]. R. Doczi, V. Semkova, A. Fenyvesi, et al., Nuclear Science and Engineering 129 (1998) 164.
- [51]. M. IbnMajah, S. M. Qaim, Nuclear Science and Engineering, 104 (1990) 271.
- [52]. P. M. Prajapati, S. Mukherjee, H. Naik, et al., Nuclear Science and Engineering 171 (2012) 78.
- [53]. C. Konno, Y. Ikeda, K. Oishi, K. Kawade, H. Yamamoto, H. Maekawa Activation Cross section measurements at neutron energy from 13.3 to 14.9 MeV Report: JAERI Reports No.1329, 1993.
- [54]. R. Pepelnik, B. Anders, B. M. Bahal, M. Farooq, 14 MeV neutron activation cross sections-, Report: Ges.Kernen.-Verwertung, Schiffbau and Schifffahrt No.86-E-29, (1986).
- [55]. B. Erlandsson, A. Marcinkowski, K. Nilson, Physica Scripta 19 (3) (1979) 251.
- [56]. N.I. Molla, S.M. Qaim, Nuclear Physics, Section A 283 (1977) 269.
- [57]. L. Husain, A. Bari, P.K. Kuroda, Physical Review C, Nuclear Physics 1 (1970) 1233.
- [58]. M. Bormann, F. Dreyer, H. Neuert, I. Riehle, U. Zielinski, Measurements of some fast neutron cross sections with the activation method, Nuclear Data For Reactors Conf., Paris 1966 Vol.1 (196) 225.
- [59]. Yuan Xialin, Zhao Wenrong, Yu Weixiang, Lu Hanlin, Chinese J. of Nuclear Physics (Beijing), 12 (4) (1990) 289.
- [60]. W. Augustyniak, M. Herman, A. Marcinkowski, Acta Physica Polonica, Part B 7 (1976) 347.
- [61]. P. Venugopala Rao, R.E. Wood, J.M. Palms, R.W. Fink, Physical Review C, Nuclear Physics 3 (1971) 629.
- [62]. Junhua Luo, Li Jiang, Long He, Radiochim Acta, 106(9) (2018) 709-717.

- [63]. C. Konno, Y. Ikeda, K. Oishi, K. Kawade, H. Yamamoto, H. Maekawa, Activation Cross section measurements at neutron energy from 13.3 to 14.9 MeV, Report: JAERI Reports No.1329 (1993).
- [64]. M. Bormann, H-K. Feddersen, H.-H. Holscher, W. Scobel, H. Wagner, Zeitschrift fuer Physik A, Hadrons and Nuclei 277 (1976) 203.
- [65]. S. K. Ghorai, R. Vos, J. R. Cooper, W. L. Alford, Nuclear Physics, Section A 223 (1974) 118.
- [66]. M. Bormann, A. Behrend, I. Riehle, O. Vogel, Nuclear Physics, Section A 115 (1968) 309.
- [67]. B. Minetti, A. Pasquarelli, Nuclear Physics Section A 118 (1968) 449.
- [68]. Imran Pasha, Rudraswamy Basavanna, et al., Journal of Radioanalytical and Nuclear Chemistry 320 (2019) 561-568.
- [69]. M. Majerle, P. Bem, J. Novak, E. Simeckova, et al., Nuclear Physics, Section A 953 (2016)139.
- [70]. M. Honusek, P. Bem, V. Burjan, et al., Journal of the Korean Physical Society 59 (2011) 1374.
- [71]. Y. Uno, S. Meigo, S. Chiba, T. Fukahori, et al., Measurements of activation cross sections for the neutron dosimetry at an energy range from 17.5 to 30 MeV by using the ${}^7\text{Li}(p,n)$ quasi-mono-energetic neutron source, Conf.: 9th Internat. Symposium on Reactor Dosimetry, Prague (1996) 465.
- [72]. A. Fessler, et al., Nuclear Science and Engineering, 134 (2) (2000) 171.
- [73]. Zhao Wenrong, et al., Compilation of measurements and evaluations of nuclear activation cross sections for nuclear data application, Report: Chinese report to the I.N.D.C.No.16, 1989.
- [74]. D. C. Santry, R. D. Werner, Canadian Journal of Physics, 68 (1990) 582.
- [75]. A. A. Filatenkov, Neutron activation cross sections measured at KRI in neutron energy region 13.4-14.9 MeV, Report: USSR report to the I.N.D.C.No.0460, 2016.
- [76]. He. Guozhu, et al., Annals of Nuclear Energy 33 (2006) 37.

- [77]. C. Konno, Activation Cross section measurements at neutron energy from 13.3 to 14.9 MeV, Report: JAERI Reports No.1329, 1993.
- [78]. M. Bormann, et al., Zeitschrift fuer Physik A, Hadrons and Nuclei, 277 (1976) 203.
- [79]. M. Bormann, Nuclear Physics, 63 (1965) 438.
- [80]. C.V. Srinivasa Rao, et al., Fluctuations in the systematics of (n,2n) reaction cross sections in the low mass region; Fast neutron cross sections of fusion reactor interest, Conf.: 21. Nucl. Phys. and Solid State Phys. Symp-1978., Bombay Vol.2 (1978) 113.
- [81]. B. Minetti, A. Pasquarelli, Nuclear Physics Section A 118 (1968) 449.
- [82]. N. I. Molla, Measurement of cross section for neutron induced reactions at 14 MeV via activation technique, Prog. Rep.: Bangladesh report to the I.N.D.C.No.002, 1983, pp.1.
- [83]. G. N Salaita, P. K. Eapen, Production of Isomers by the Cyclic Activation Method, Conf. on Nucl.Meth.in Envir. Res., Columbia-1974, 95.
- [84]. M. Hyvoenen-Dabek, Tarvainen, P. Holmberg, Journal of Radioanalytical Chemistry 46 (1978) 357.