Synopsis on

Study of Nuclear Reaction Cross-Sections for Reactor Applications

By

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### 1 Introduction

For an efficient economic and social development, a basic necessity is to access an affordable energy. In addition to food and water, electricity in particular is a major factor in the quality of life. It is clear that the world is facing considerable energy and environmental challenges keeping in mind that the current energy systems, particularly in the underdeveloped and the developing countries, are highly dependent on fossil fuels, whose combustion accounts for global greenhouse gas emission [1]. This trend will continue in the future, unless more affordable and environmental friendly source of electricity could be supplied. With all the energy production systems there are environmental issues to be considered, risks to be assessed, and challenges to be addressed. It needs to be emphasized that nuclear power is currently the only technology with a secure base load electricity supply and no greenhouse gas emissions that has the potential to expand to a large scale, and efficiently replace fossil fuel while it has a fifty year history of operation. However, there are issues that need to be addressed in the conventional nuclear reactors and advanced reactor designs. Most of the countries around the globe are using conventional nuclear reactors using Uranium and Plutonium as nuclear fuel. There are several challenges comes to existence with these kind of reactors, like, the troublesome issue of disposal of the nuclear waste, in particular long-lived transuranic elements (TRU), e.g. <sup>237</sup>Np, <sup>240</sup>Pu, <sup>241</sup>Am, <sup>243</sup>Am, <sup>244</sup>Cm etc. and fission products (FP), e.g.  $^{129}I$ ,  $^{135}Cs$ ,  $^{99}Tc$ ,  $^{93}Zr$ ,  $^{107}Pd$  etc. The nuclear waste disposal is certainly an urgent and important issue to be tackled to ensure future growth of nuclear power.

The fuel cycle based on thorium can address both these issues of global warming as well as nuclear waste management, owing to a number of favorable material characteristics which makes thorium a better fertile host [2]. The  $^{232}Th - ^{233}U$  fuel cycle has an advantage over the present reactor based on uranium fuel from the point of thousand times less radiotoxic waste production. Further, Thorium in its natural form consist almost all of  $^{232}Th$  with some trace amounts of  $^{230}Th$ . Thorium has a longer half-life, and hence, its occurrence on the earth's crust is nearly three times as that of uranium. Thus, Thorium is an attractive fuel option for contributing to large-scale global deployment of nuclear energy to meet rising demands [2]. Thorium is a fertile material and has to be converted into  $^{233}U$ , a fissile isotope. The cross-section for capture of thermal neutrons in  $^{232}Th$  is typically 2.47 times that in  $^{238}U$ . Therefore, thorium offers a greater competition to capture of the neutrons and lower losses to structural and other parasitic materials leading to an improvement in conversion of

 $^{232}Th$  to  $^{233}U$ . In addition, of the three fissile species ( $^{233}U$ ,  $^{235}U$  and  $^{239}Pu$ ),  $^{233}U$  has the highest value of ' $\eta$ ' value (number of neutrons released per neutron absorbed) that remains nearly constant over a wide energy range, in thermal as well as epithermal regions, unlike  $^{235}U$  and  $^{239}Pu$ . This facilitates achievement of high conversion ratios with thorium utilization in reactors operating in the thermal/epithermal spectrum.

By considering the above fact, it has been realized that there is a strong need for the development of thorium based technologies for the entire fuel cycle. The Advanced Heavy Water Reactor (AHWR) has been designed to fulfill this need and maximum power generation from thorium in India [3]. Bowman [4] and C. Rubbia et al [5] have proposed the concept of Accelerator-Driven Sub-critical Systems (ADSs) which demonstrate that a commercial nuclear power plant of adequate power can also be built around a sub-critical reactor, provided it can be fed externally with required intensity of accelerator-produced neutrons. The ADSs have attractive features for the elimination of troublesome long-lived minor actinides and fission products of the spent fuel, as well as for nuclear energy generation utilizing thorium as fuel. In ADSs, high energy proton beam (around 1 GeV) strikes a heavy element target like W, Pb or Bi target, which yields copious neutrons by (p, xn) spallation reaction. Therefore, the spallation target becomes a source of neutrons, which can achieve selfterminating fission chain in a sub-critical core. Therefore, neutron as well as proton cross-section data is required to design different components of advanced reactor, i.e. shielding design, waste estimation, and estimation of radiation damage, nuclear heating, transmutation effects, and radiation dose.

In addition to the ADS systems, International Thermonuclear Experimental Reactor (ITER), is also another option for green energy production. In fusion reactor like ITER, during the plasma shot DT fusion reaction will produce 14.1 MeV neutrons. These neutrons will irradiate structural materials of the reactor. In ITER  $Nb_3Sn$  conductor is selected for making ITER toroidal field coils [6], are able to carry higher current and produce stronger magnetic field. They also consume less power and are cheaper to operate. Ten thousand tons of magnets produce the magnetic field that will initiate, confine shape and control the ITER plasma. As these field coils are located just after the blanket, these will get exposed to high energy neutrons produced from the fusion. Therefore, it is very necessary to estimate the cross-section of all possible reactions around 14 MeV on different isotopes of these carefully selected magnetic materials. The precise data for the reaction cross section are of prime importance from the view point of its nuclear applications, transmutation, shielding effect, radiation damage, long term activation etc. It is found from the Exchange Format (EXFOR) [7] database that there is lack of measured cross section data of (n, p) and (n, 2n) reactions for Tin (Sn) and its isotopes.

The proton induced reaction cross-section data are also important for the dose estimation, radiation safety and to monitor other parameters which are primarily useful for the development of advanced reactor and medical accelerators. Keeping this aim, proton induced reaction data were measured for Nb, Ag, and Ti isotopes. These isotopes are important to produce some of the important medical isotopes and are used for cladding and surrounding structural material alloys.

The uncertainties in the measured neutron induced reaction data were calculated by error propagation method or covariance analysis. The advantage of using this method is that it combines the errors from all the sources and propagate it into the final uncertainty of the data depending on the nature of the error. In the case of proton induced reaction data, uncertainties were measured directly by conventional method of taking quadratic sum of both statistical and systematic errors. However, the statistical errors and the error due to flux was reduced by counting the sample for appropriate time and by measuring the flux directly by using a Faraday's cup.

### 2 Literature Survey

The detailed literature survey shows that exhaustive experimental work has been carried out in the low energy neutron induced reactions of structural as well as fuel materials. However, the data are usually scarce above the incident neutron energies of 10 MeV. In addition, there are so much discrepancy in the available data of various authors in the measured as well as computed data. The EXFOR compilation on the neutron induced data also indicates that there could be an involvement of pre-equilibrium (PE) process in the formation of the reaction residues at these energies, therefore, different nuclear model code were used to find the PE contribution in the measured data. It has also been noticed that most of the authors have reported the uncertainties without considering the error from the monitor reaction data. Since, the monitor reaction data also contain significant uncertainties, thus it becomes important to use a proper error propagation method to involve the errors coming from each parameter used in the calculations.

The literature survey of proton induced reaction data for the selected materials show that sufficient work has been done in order to understand the reaction mechanism up to 20 MeV incident particle energies. The re-

ported data contain minor discrepancies and errors which may come from the use of different monitor reactions. Since, the irradiation is carried out by using protons which is a charged particle, therefore, the flux estimation can be done by monitoring the beam directly from a Faraday cup. The use of a Faraday cup reduces the errors due to beam fluctuations and also the cross-sections can be measured directly, unlike relative measurement in case of the use of monitor reaction. Also, the PE process dominates over the compound nucleus process at higher energies. To investigate the fact, PE contribution was measured with the help of theoretical nuclear codes and is plotted with different systematics formulas.

### 3 Motivation

Enormous amount of experimental work has been carried out in low energy neutron induced reactions of actinides [8-19, 7] and nuclear structural materials. However, experimental nuclear data in medium to high-energy neutron or proton induced reactions are rare and very much limited. Therefore, there is a strong need to measure reaction cross sections of Th and reactor cladding and shielding materials to the medium energy region (up to 25 MeV) with mono-energetic neutrons and protons. Therefore, measurements of the different types of reaction cross sections in the above mentioned energy region will help us to understand the energy dependence of the activation cross-sections in detail. This will provide a complete database that will lead to better understanding of mechanisms of the nuclear reactions and in future reactor technology development.

Since, International Thermonuclear Experimental Reactor (ITER) can also be a sustainable energy source in future. Therefore, it is necessary to study the neutron capture cross-sections for its magnetic material Sn, since the magnets used for holding the plasma get irradiated with secondary neutrons and protons which are produced in the operation.

In view of the above we have measured the reaction cross section in fast neutron and proton induced reaction of Th and Mo, Ni, Sn, Nb, Ag, and Ti related to ADSs and ITER. The Tables 1 and 2 below summarize the reactions studied by using both the neutron and proton beams in the present work.

## 4 Objectives

The following objectives have been fulfilled in the present work,

r spectroscopic data for the isotopes used in the measurement of the $^{232}Th(n, \gamma)$ , $^{100}Mo(n, 2n)$ and $^{58}Ni(n, x)$ reaction	
Table 1: Nuclear spectroscopic data for the is	cross-sections.

Reaction	Threshold (Q-value) (MeV)	$T_{1/2}$	Decay Mode $E_{\gamma}$ (%) (keV	$E_{\gamma}$ (keV)	$I_{\gamma}^{(0/0)}$
$^{232}Th(n,f)^{97}Zr$ (m)	I	$16.749 \pm 0.008  \mathrm{h}$	$eta^-(100)$	743.36	$93.09\pm0.01$
$^{232}Th(n,\gamma)^{233}Th$	(4.78)	$21.83 \pm 0.04 \text{ min}$	$eta^-(100)$	I	I
$^{233}Th \xrightarrow{\beta^-} ^{233}Pa$	I	$26.975 \pm 0.013$ d	$eta^{-}(100)$	311.90	$38.5\pm0.4$
$^{27}Al(n, \alpha)^{24}Na \ (m)$	3.249(3.13)	$14.997 \pm 0.012 ~ m h$	$eta^-(100\%)$	1368.68	$89.43\pm0.23$
$^{100}Mo(n, 2n)^{99}Mo$	8.37 (8.29)	$65.976 \pm 0.024 \ { m h}$	$\beta^{-}(100\%)$	140.5	$9.9936 \pm 0.0015$
$^{197}Au(n,\gamma)^{198}Au$ (m)	(0.65)	$2.6941 \pm 0.0002 \text{ d}$	$eta^{-}(100\%)$	411.8	95.62
$^{115}In(n,n')^{115m}In$ (m)	I	$4.486\pm0.004~\mathrm{h}$	IT(95.00%)	336.2	$45.9\pm0.1$
			$eta^-(5.00\%)$		
$^{58}Ni(n, p)^{58}Co$	(0.4)	$70.86\pm0.06~{ m d}$	$\epsilon(100\%)$	810.7	99.45
$^{58}Ni(n,2n)^{57}Ni$	12.428 (12.21)	$35.60\pm0.06~\mathrm{h}$	$\varepsilon(100\%)$	1377.6	$81.70\pm0.24$

 $(m) \rightarrow$  monitor reaction,  $min \rightarrow minute$ ,  $h \rightarrow hour$ ,  $d \rightarrow day$ ,  $y \rightarrow year$ 

Nuclide	<i>T</i> <sub>1/2</sub>	Deacy Mode	$E_{\gamma}$ (keV)	$I_{\gamma}$ (%)	Channel	$E_{Th}$ (MeV)
<sup>93m</sup> Mo	6.85 h	IT (99.88 %)	263.05	$57.40\pm0.11$	$^{93}Nb(p,n)$	1.20
		$\epsilon(0.12\%)$	684.69	$99.9 \pm 0.8$	(,,,,,)	
			1477.14	$99.10\pm0.11$		
<sup>93</sup> gMo	$4.0  imes 10^3  ext{ y}$	$\epsilon(100\%)$	-	-	-	-
$^{92m}Nb$	10.15 d	$\epsilon(100\%)$	934.44	99.15	$^{93}Nb(p,pn)$	9.06
<sup>92</sup> gNb	$3.47 \times 10^7 \text{ y}$	$\epsilon(100\%)$	934.5	$74.0\pm0.11$		
	5	$\beta^+(< 0.05\%)$				
<sup>89m</sup> Zr	4.16 m	IT(93.77%)	587.83	89.62	$^{93}Nb(p,\alpha n)$	5.60
<sup>89</sup> gZr	78.41 h	$\epsilon(100\%)$	909.15	99.14		
<sup>107</sup> Cd	6.50 h	$\varepsilon(100\%)$	93.12	$4.8\pm0.3$	$^{107}Ag(p,n)$	2.21
					$^{109}Ag(p, 3n)$	18.82
$^{106g}Ag$	23.96 m	$\epsilon(99.5\%)$	-	-	-	-
0		$\beta^{-}(<1\%)$	-	-	-	-
<sup>106m</sup> Ag	8.28 d	$\epsilon(100\%)$	450.98	$28.2\pm0.7$	$^{107}Ag(p,d)$	7.37
			616.17	$21.6\pm0.6$	$^{107}Ag(p, pn)$	9.62
			717.24	$28.9\pm0.8$		
10			748.44	$20.6\pm0.6$	17	
$^{48}V$	15.97 d	$\epsilon(100\%)$	983.52	99.98	$\frac{47}{10}Ti(p,\gamma)$	
			1312.1	97.5	$\frac{48}{10}Ti(p,n)$	4.8
47					$\frac{49}{10}Ti(p,2n)$	13.20
<sup>47</sup> Sc	3.349 d	$eta^-(100\%)$	159.38	68.3	$\frac{48}{10}Ti(p,2p)$	11.68
					$^{49}Ti(p,^{3}He)$	12.11
16					$\frac{50}{17}Ti(p,\alpha)$	2.28
<sup>46</sup> Sc	83.79 d	$eta^-(100\%)$	889.28	99.98	$^{47}Ti(p,2p)$	10.69
			1120.55	99.99	$^{48}Ti(p,^{3}He)$	14.67
					$\frac{49}{50}Ti(p,\alpha)$	1.98
11		(			$50 Ti(p, n\alpha)$	13.13
44gSc	3.93 h	$\varepsilon(100\%)$		_	$\frac{47}{18}Ti(p,\alpha)$	2.30
$^{44m}Sc$	58.61 h	IT(98.8%)	271.1	86.7	$^{48}Ti(p,n\alpha)$	14.17
		$\epsilon(1.2\%)$				

**Table 2:** List of the identified residues in the proton induced  $^{nat}Nb$ ,  $^{nat}Ag$  and  $^{nat}Ti$  reactions with their spectroscopic data.

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

- To measure the reaction cross sections  $[(n, \gamma), (n, 2n)]$  from medium to fast neutron induced reaction of <sup>232</sup>*Th* related to fast reactor and ADSs. We have also planned to measure the (n, p) and (n, 2n) reaction cross sections of few of the structural and cladding materials such as Mo, Ni, and Sn induced by fast neutrons. The monoenergetic fast neutrons can be produced using <sup>7</sup>*Li*(*p*, *n*) reaction at BARC-TIFR Pelletron and at FOTIA facilities.
- To measure the (p, x) reaction cross-sections for the structural materials like Nb, Ag, and Ti. The proton beams were taken for irradiation purposes from BARC-TIFR Pelletron accelerator.
- The experimental work consists of the irradiation of Th, structural materials (e.g. Mo, Ni, Sn, Nb, Ag, Ti) with proton beam at 10- 22 MeV energy range, and quasi mono-energetic neutrons within 2-20 MeV energies. This was followed by their gamma-ray spectrometric analysis. The off-line *γ*-ray counting of the irradiated samples was carried out by using HPGe detectors.
- The covariance analysis was also carried out for the neutron induced reaction cross-sections to calculate the uncertainties and correlations among the different data.

## **5** Outline of the Thesis

The thesis is divided into seven chapters. A summarized detail of each chapter is as follows;

**Chapter 1-** It provides an outlook on the world's demand for energy and our dependence over the nuclear reactor generated energy. It gives the reader an idea of the importance of the thorium utilization in future and the development of underlined accelerator technology. This chapter also gives the details of the work being done by different authors/groups in previous years. It gives an insight to the ADS and ITER working. The chapter also puts light on different nuclear reaction processes useful to understand the presented work.

**Chapter 2-** The chapter contains all the necessary information and the details regarding the experimental work carried out to measure the reaction cross-sections with neutron as well as proton as the incident particle. The chapter contains brief discussion about the measurement techniques used in the present work.

**Chapter 3-** The third chapter gives a contains a detailed derivation of the cross-section formalism used for calculations together with the complete details about the error propagation methods in the form of covariance analysis, used to find the uncertainties in the detector efficiencies and the measured data.

**Chapter 4-** This chapter will provide a complete understanding of the different nuclear model codes used in the present analysis. A very brief discussion will be added about each code which will help the reader to find the specific details regarding the present work only.

**Chapter 5-** This chapter will present the complete experimental work carried out for neutron induced reaction cross-section data for Th, Mo, and Ni isotopes. The chapter is designed to present the experimental procedure followed by the data analysis and the measurement of cross-sections. Later it gives an insight into the uncertainty calculations using covariance analysis for  $^{232}Th(n, \gamma)$  reaction. Complete step by step calculations are present in Appendix for better readability.

**Chapter 6-** The sixth chapter contains the experimental findings of the proton induced reaction cross-section data. The chapter is designed similarly as chapter 5.

**Chapter 7-** In the end, the chapter 7 will be containing the summary, outcomes of the present work including a short description of the future perspective of the present work.

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### **List of Publications**

## **A.** Publications in Peer-Reviewed Journals

#### **Publications Related to Thesis**

1. Effect of neutron generator and monitor reactions on the  $(n, \gamma)$  and (n, 2n) cross-sections of fertile materials within 0.1-25 MeV

Siddharth Parashari and S. Mukherjee, *Under Review in European Physical Journal Plus.* (Review Article)

2. Measurement of the  ${}^{58}Ni(n,p){}^{59}Co$  and  ${}^{58}Ni(n,2n){}^{57}Ni$  reaction crosssections for the fast neutron energies upto 18 MeV

Siddharth Parashari, S. Mukherjee, H. Naik, S.V. Suryanarayana, R. Makwana, B.K. Nayak, and N.L. Singh *Eur. Phys. Jour. A* (2019) 55: 51.

3. Systematic analysis of the neutron induced reaction cross sections for <sup>nat</sup> Mo isotopes within 10-20 MeV

Siddharth Parashari, S. Mukherjee, S.V. Suryanarayana, B.K. Nayak, R. Makwana, N.L. Singh, and H. Naik *Phys. Rev. C* 99, 044602 (2019).

4. Excitation function of the  $^{nat}Ti(p, x)^{48}V$ ,  $^{47,46,44m}Sc$  reactions within the energy range of 10-22 MeV

Siddharth Parashari, S. Mukherjee, B.K. Nayak, H. Naik, S.V. Suryanarayana, R. Makwana, N.L. Singh *Nucl. Phys. A.* 987 (2019) 128–143.

5. Excitation function of the  $p + {}^{nat} Ag$  reactions in the energy range 10-22 MeV

Siddharth Parashari, S. Mukherjee, S.V. Suryanarayana, R. Makwana, B.K. Nayak, A. Shanbhag, H. Naik *Nucl. Phys. A* 979 (2018) 102-112.

6. Excitation functions of the  $p + {}^{93}Nb$  reaction in the energy range 10-22 MeV

Siddharth Parashari, S. Mukherjee, B.K. Nayak, R. Makwana, S.V. Suryanarayana, H. Naik, S.C. Sharma *Nucl. Phys. A* 978 (2018) 160–172.

7. Measurement of  $^{232}Th(n, \gamma)$  reaction cross sections in the neutron energy range of 11–19 MeV

Siddharth Parashari, S. Mukherjee, A. P. Singh, Vibha Vansola, H. Naik, B. K. Nayak, Rajnikant Makwana, S.V. Suryanarayana, N. L. Singh, Mayur Mehta, Y. S. Sheela, M. Karkera, R. D. Chauhan, and S. C. Sharma *Phys. Rev. C* 98, 014625 (2018).

8. Investigation of (n, p), (n, 2n) reaction cross sections for Sn isotopes for fusion reactor applications

Siddharth Parashari, S. Mukherjee, Vibha Vansola, Rajnikant Makwana, Nand Lal Singh, Bhawna Pandey, *Applied Radiation and Isotopes* 133 (2018) 31–37.

#### **Other Publications**

9. Measurement of (n, xn) reaction cross-sections on <sup>113,115</sup>*In* isotopes using quasi-monoenergetic neutrons within 10-20 MeV

Bhargav Soni, Siddharth Parashari, S. Mukherjee, Rajnikant Makwana, S. V. Suryanarayana, B. K. Nayak, H. Naik, and K. Katovsky *Under Review in European Physical Journal Plus.*.

10. 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, Siddharth Parashari, R. Singh, H. Naik, S.C. Sharma, S. Ayyala, B. Soni, R. Chauhan *App. Rad. iso.* **154** (2019) 108866.

11. Systematic study of the break-up fusion process in the  ${}^{12}C + {}^{165}Ho$  system and interplay of entrance channel parameters

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12. Systematic study of low energy incomplete-fusion dynamics in the <sup>16</sup>O +<sup>148</sup> Nd system: Role of target deformation

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17. Measurement of excitation functions of evaporation residues in the  ${}^{16}O + {}^{124}$ Sn reaction and investigation of the dependence of incomplete fusion dynamics on entrance channel parameters

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## 18. Sensitivity of low-energy incomplete fusion to various entrance-channel parameters

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## **B.** Publications in the Proceedings of the International Conferences

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