

CHAPTER 1

INTRODUCTION

1.1 Introduction

1.2 Literature survey

1.3 Accelerator driven sub-critical system (ADSs)

1.4 Nuclear reactions

1.4.1 Neutron induced reactions

1.5 Motivation

1.6 Objective

1.7 Content of the present thesis

References

1.1 Introduction

Nuclear data such as fission/reaction cross-sections and the isotopic production yields as well as the nuclear spectroscopic data are important for their application in various fields and research work. In particular, the nuclear fission and reaction cross-sections of actinides and structural materials induced by photon, neutron and charged particles (proton) are of special interest for their applications in the present and modern accelerator technology for the production of more efficient nuclear energy. Among these, the fast photon, neutron and proton induced reaction/fission cross-sections in the Fast Breeder Reactor (FBR), Advanced Heavy Water Reactor (AHWR) and Accelerator Driven Sub-critical system (ADSs) are the immense interests of present time to enhance the nuclear energy production to fulfil the increasing energy demand. Thus, several countries all over the world have different programs for the production of the nuclear energy. Some of them are developing the advanced technology for the design of fast reactor, AHWR and ADSs, to utilize the potential of fertile ^{238}U and ^{232}Th [1-3]. In December 1999, a co-ordinator committee was formed in India for the research, development and construction of ADSs [2, 4, 5]. Primarily, the advancement of the technology, which is mandatory for building ADSs was discussed in details. The construction of the linear proton accelerator (up to 20 MeV proton energy and 30 mA current) and study of the necessary maps and drafts of subcritical reactor as well as understanding of codes and development of befitting nuclear model codes, all these aspects are taken care in the first phase [1-11]. Actual blueprint and assembly of ADSs was looked after in the later phase.

Some of the striking features of ADSs are superior safety characteristics, its potential to incinerate long lived actinides, transmutation of long lived fission products and, of course, nuclear energy production (electricity generation plus research and development). Rich reservoirs of thorium in India, add an additional dimension to this advanced technology. Also, as compared to uranium, thorium produces much less amount of the long-lived radio actinides. However, thorium is a fertile element and not the fissile one. This fertile thorium can be turned into fissile isotope ^{233}U by using the transmutation process, in which fertile ^{232}Th captures a neutron and gets converted into fissile isotope ^{233}U by neutron capture and two successive beta decay. This ^{233}U can be used as a reactor fuel using ADSs, which has been estimated to be $\approx 20\%$ more efficient than conventional ^{238}U as reactor fuel. A part of the energy generated, which can be employed to govern the accelerator connected with the sub-critical reactor using a feedback loop. The energy feedback to the accelerator makes the

whole process more energy efficient and self-sustainable. The construction of ADSs was divided in several stages. These stages are discussed below in details:

1. Reactor physics experiments using AHWR reactors and 14 MeV neutron generators-building, well designed cyclotron (up to 350 MeV) and Linear Accelerator (LINAC) of energies (up to 100 MeV) were taken care in first stage.
2. Second stage, if decided and few other sub-stages in order to test the modules of ADSs, spallation target and fission power.
3. Final stage will have the appropriate designing of a proto-type of ADSs with proton beam up to 1 GeV. Proper testing and further research and development will be done under this stage.

Neutron cross-section data are required for the fabrication of different components of an advanced reactor, i.e., shielding design, estimation of waste and radiation damage, nuclear heating, transmutation effects and radiation dose. The fast neutron induced cross-section data of (n, γ), (n,2n), (n,p) and (n, α) reactions with the spallation target (^{nat}Pb and/or ^{209}Bi) and structural materials (e.g., Zr, Nb, Fe, Ni, Cr, Mn, Y and Ag) are needed to reduce the risk related to radiation leakage. At high energies, nuclear data of structural materials (e.g., Zr, Nb, Fe, Ni, Cr, Y, Mn and Ag) are required as hydrogen and helium production occurs during the irradiation, which may lead to swelling of the fuel pellets as well as surrounding structural material and can cause the failure of the mechanical assembly. Besides this, the neutron induced reaction products are important for the use of medical diagnosis such as positron emission tomography (PET) and single photon emission computed tomography (SPECT). However, the needed database related to the production and dose estimation of medical isotopes is scarce. In view of the above discussion, the study of the fast neutron induced reaction/fission cross-sections of stable fertile actinides (e.g., ^{232}Th and ^{238}U), structural and cladding materials (e.g., Zr, Nb, Fe, Ni, Cr, Mn, Y and Ag) [1-5] is vital for the advancement and development of the present reactor and/or accelerator technology and to build nuclear data libraries which would be helpful in order to calculate certain parameters for research in physical as well as medical sciences.

1.2 Literature survey

Literature survey shows that a lot of experimental work has been performed for the neutron induced reactions, in the lower energy region of structural as well as fuel materials. There are no data available for these materials at neutron energy regions above 1 MeV. Also

there are discrepancies among the available data of numerous authors in the measured as well as computed data [7-11].

On the basis of rigorous literature survey, the author has found that for $^{197}\text{Au}(n,\gamma)$ reaction there are measured cross-section data compiled in the EXFOR data base [7]. This shows that the $^{197}\text{Au}(n,\gamma)$ reaction cross-section data are available for the neutron energies of 0.024 to 3 MeV and 14.7 MeV. Except these, there are no experimental data available within the neutron energy range of 3 to 14.7 MeV [12-24].

For $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ reaction, the measured cross-sections are available within 4 MeV neutron energy range and around 13.4 to 15 MeV [25-34]. From 0.97 to 19.4 MeV energy range, only one data set is reported by Menlove et al [35]. Within the neutron energies of thermal to 4 MeV, there are discrepancies among the available data from various authors.

The detailed literature survey on $^{232}\text{Th}(n,\gamma)$ and $^{238}\text{U}(n,\gamma)$ reaction cross-sections indicates that different experimentally measured data are available for the $^{232}\text{Th}(n,\gamma)$ and $^{238}\text{U}(n,\gamma)$ reactions over vast range of neutron energies from thermal to 3 MeV based on physical measurements and activation technique. Beyond 3 MeV, only few measured data of the $^{232}\text{Th}(n,\gamma)$ reaction are available at the neutron energies of 3.7, 5.9, 8.04, 9.85, 11.9, 13.5, 14.55, 14.8, 15.5 and 17.28 MeV [36-47] and in the case of $^{238}\text{U}(n,\gamma)$ reaction, measured data are available at the neutron energies of 3.033, 3.5, 3.7, 4, 5, 5.9, 6, 7, 7.2, 7.6, 8.04, 8.2, 9.2, 9.85, 10.2, 11.2, 11.9, 12.2, 13.2, 14, 14.2, 14.5, 13.5, 14.8, 15.5, 17, 17.28, 18, 19 and 20 MeV [48-62].

1.3 Accelerator Driven Sub-critical system (ADSs)

Most of the countries in the world are inclining towards the development of the ADSs (Accelerator Driven Sub-critical system). In past few years there has been so much of progress in this concept. ADSs is an advance reactor system, in which thorium and long lived minor actinides can play considerable role as the nuclear fuel and can also be incinerated without any risks related to criticality. Bowman [3] is the one who has initiated to show the great advantages of this type of advanced reactor. In his paper, he has explained how the commercial reactor of adequate power can be built with a sub-critical core. The only condition to initiate the nuclear reaction in this reactor is to feed high energy neutron beam externally based on the required intensity of accelerated proton.

In ADSs, the high-energy proton beam strikes a heavy element target like $^{\text{nat}}\text{Pb}$ and ^{209}Bi . This interaction generates plentiful amount of neutrons by (p,xn) spallation reaction. This spallation target is the greatest source of neutrons, which also can drive the self-

terminating fission chain reactions in a sub-critical core. Thus, each energetic proton of 1 GeV can generate around 20-30 neutrons by bombarding on a high z-elements. A schematic of the ADSs is given in figure 1.1 and the principal sub-systems of ADSs are given below:

1. Proton accelerator with 1 GeV energy and current ≥ 10 mA
2. Spallation targets for the generation of beam power ≥ 10 MW
3. Sub-critical reactor system

The ADSs has so many interesting features for the generation of nuclear power by nuclear energy. However, the most challenging part in the construction of ADSs is the high energy and high current accelerator with adequate power, which should be able to deliver the average proton beam power of 10 MW or greater. The development work of such accelerator is going on a large scale and it will be useful in nuclear sciences and technological applications. Thermal power of ADSs depends on the multiplication factor k of the fuel (sub-critical) and also on the strength of primary n^0 source. In ADSs, the energy generated by spallation is quite higher than the neutron energy of the conventional reactor due to high energy incident proton beam.

ADSs can be very useful for incinerating the heavy long-lived actinide isotopes (^{237}Np , ^{240}Pu , $^{241,243}\text{Am}$, ^{244}Cm) and transmuting long-lived fission products (^{93}Zr , ^{99g}Tc , ^{107}Pd , ^{129}I , ^{135}Cs). In the blanket assembly, there is actinide fuel or spent nuclear fuel. One can start with the conventional reactors spent fuel in the outer blanket region and gradually moving it inwards. After that it can be removed or reprocessed. At the end, one will be left with the recyclable uranium and separate fission product waste. After that actinides can be reused for further fission.

When the fuel contains odd numbered isotopic atoms, heavier than ^{232}Th , there is a higher probability of neutron absorption, which will lead to fission process. Thus, energy will be liberated and multiplication process will go on. This is because the fast neutron causes maximum fission of even-even, odd-even actinides. When the fuel outside the core and blanket contains even-even actinide isotopes, neutron will be captured and will subsequently cause the beta decay to produce a fissile isotope, which is shown in the figure 1.2 given below.

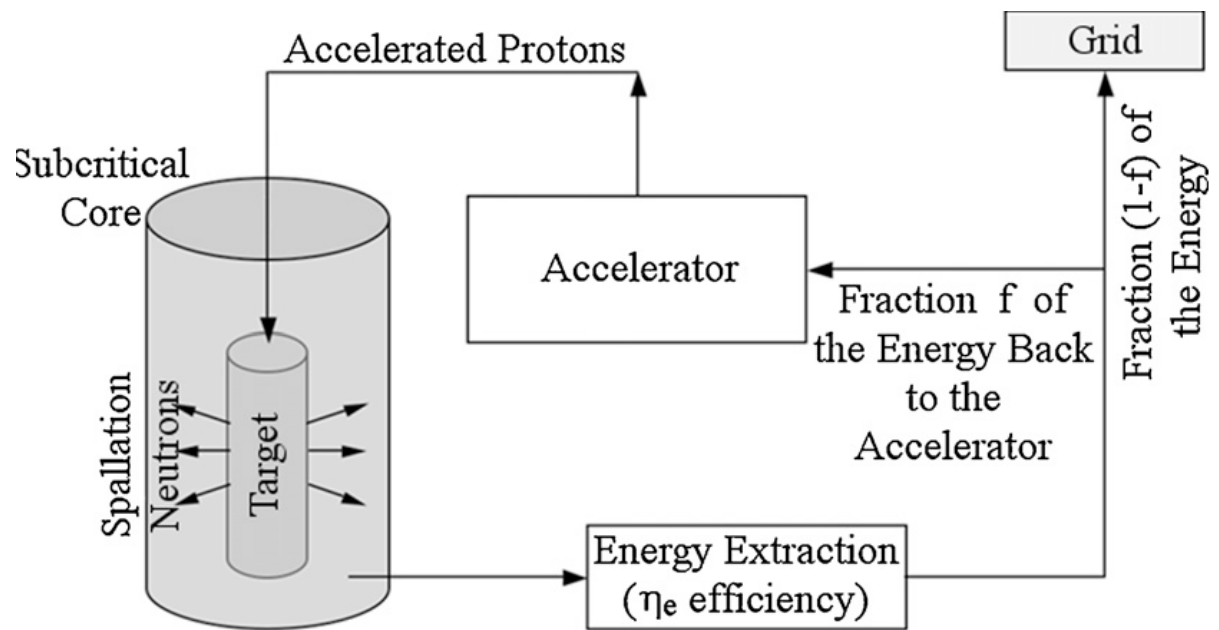


Figure 1.1: Schematic diagram of ADSs [63]

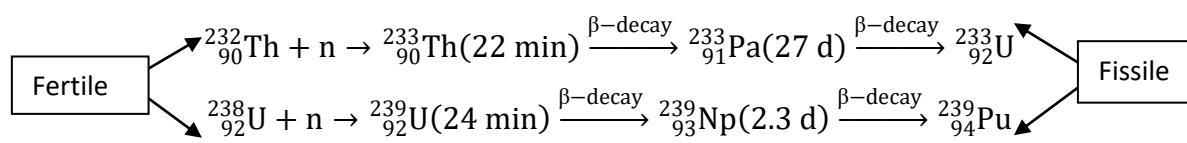


Figure 1.2: Production of fissile isotopes from the fertile isotopes in ADSs

1.4 Nuclear reactions

When a projectile is bombarded on a target nucleus, the interaction is known as a nuclear reaction. For the occurrence of different reactions, the energy of the projectile as well as the charge (Z) of target and projectile are responsible. If the ejectile is same as the projectile and the nucleus doesn't break, the process is known as scattering. It has two types elastic and inelastic.

1.4.1 Neutron induced reactions

These are the types of nuclear reactions where nucleus of target element interacts with the neutrons, which are projected towards it. As the neutrons do not possess any charge, there is no Coulomb barrier between neutron and target nucleus and that is the reason why neutrons of any energy interacts with all the nuclides from the nuclear element chart.

There are two types of nuclear reactions:

1. Endoergic Nuclear Reaction
2. Exoergic Nuclear Reaction

If energy is liberated during nuclear reaction, it is called exoergic nuclear reaction and if energy is required to initiate the nuclear reaction, it is called endoergic nuclear reaction. Neutron with zero energy also can start the reaction. For the endoergic reactions to occur, incident neutrons must be holding higher kinetic energy than the threshold energy of that particular reaction.

There are numerous reactions that happen when projectile interacts with the target. i.e., scattering, direct reaction, pre-equilibrium, compound reaction, fission reaction, etc. There are different channels through which a neutron induced reactions may result such as (n,γ) , (n,p) , (n,n') , $(n,2n)$, $(n,3n)$, (n,α) , (n,f) , etc with different excitation functions. In the present work, I have studied (n,γ) reactions for various targets i.e., ^{197}Au , ^{55}Mn , ^{232}Th and ^{238}U . All (n,γ) reactions are exoergic with positive Q value and there are no threshold bars. As can be seen from the excitation function of $^{55}\text{Mn}(n,\gamma)$ reaction given in chapter 5 figure 5.1, the (n,γ) reactions have higher cross-section values at low neutron energies. As the neutron energy increases the (n,γ) reaction cross-section decreases.

1.5 Motivation

Demonstration of accelerator driven sub-critical reactor system (ADSs) indicates that a commercial nuclear power plant, which is able to generate just enough power can be built around a sub critical reactor, with the condition of feeding it externally with the help of

adequate amount of neutrons from the accelerator [1]. The ADSs have potential to diminish the troublesome long-lived minor actinides and fission products generated in the spent fuel and it can also generate nuclear energy utilizing the thorium as a fuel. From India's perspective, ADSs is admissible because one can utilize its potential to the full extent to fabricate hybrid reactor that can produce nuclear power with the use of abundant reservoirs of thorium, which can be used as a main fuel. ADSs based hybrid reactors, which uses thorium fuel may need only small amount of thorium, whereas AHWR needs small amount of uranium and plutonium to start the loop. In general, the additional degree of freedom provided by the external source in ADSs can enable one to design reactor system which primarily burn thorium fuel as well as make a more efficient use of natural uranium fuel. Thus, ADSs seems to possess the ability of providing an additional path, for an effective and economic power generation using the rich thorium resources in India. Structural Materials used in reactors, need to fulfil two objectives. (i) Even after the bombardment with the high neutron fluxes, these materials must hold their mechanical properties. (ii) Activation induced by neutron must not generate the long-lived radioactive waste.

In view of above facts, it is important to study the fast neutron induced reaction/fission cross-sections of actinides (^{232}Th and ^{238}U), long-lived minor actinides (^{237}Np , ^{240}Pu , $^{241,243}\text{Am}$, ^{244}Cm), spallation targets ($^{\text{nat}}\text{Pb}$, ^{209}Bi), structural and cladding materials (e.g., Zr, Nb, Fe, Cr, Mn, Ni, Y and Ag). Because of their very low absorption cross-sections for thermal neutrons and resistance to corrosion, these materials are frequently used as cladding of fuel rods. The cross-sections database for the same is rare [7-8]. Neutron induced reaction cross-section data are required for the design of different components of advanced reactor. i.e., shielding design, estimation of waste and radiation damage, nuclear heating, transmutation effects and radiation dose.

The detailed literature survey shows that considerable amount of experimental work has been executed in the low energy neutron induced fission/reactions of actinides. However, experimental nuclear data in the medium to high-energy neutron induced fission of actinides is so much lean [36-62]. Most of the nuclear data in the neutron-induced fission/reactions of actinides available the compilation are based on average neutron spectrum of reactor. Therefore, there is strong need to measure thereaction cross-sections of Th and U in the medium energy region (1-18 MeV) with mono-energetic neutrons apart from reactor neutrons. Therefore, measurements of the different types of reaction cross-sections in the above mentioned neutron energy region will help us to understand the relation between the

energy and the activation cross-sections in detail. This will provide a total database, which will be utterly helpful in the better understanding of the nuclear reaction process.

Reaction cross-sections such as (n,γ) , $(n,2n)$, (n,p) and (n,α) of the spallation target (Bi) and structural materials (e.g. Mn, Y and Ag) induced by fast neutrons are required for the safety purposes for the power plants as well as research reactors. Besides this, the neutron induced reaction products are important for the use of medical diagnosis such as positron emission tomography (PET).

1.6 Objective

In view of the above facts, we have measured the reaction cross-sections in fast neutron induced reaction of ^{232}Th and ^{238}U isotopes related to ADSs. We have also measured the reaction cross-section of neutron flux monitor (Au) and structural material (Mn) induced by fast neutrons.

The experimental work consists of irradiation of the actinides (Th, U), structural materials (Mn) and flux monitor (Au) in the above mentioned neutron sources. After that off-line gamma ray spectrometric analysis was done for the irradiated samples by using HPGe detector connected to a PC based 4096 channel analyser.

Theoretical data was computed using TALYS [9] and EMPIRE [11] nuclear model codes. Analysed nuclear data were compared to the theoretically computed data with necessary graphs drawn in ORIGIN. These data were published in national/international conferences as well as renowned journals.

1.7 Content of the present thesis

The contents of the present work are divided in the seven chapters. In which the first chapter throws light on the introduction, literature survey, motivation and objective of the present work. The second chapter covers the experimental methodology used for the present work. Theoretical aspects related to the present study were taken care of in the third chapter. Fourth, Fifth and sixth chapters are dedicated to the research work done for the Au, Mn, Th and U targets, respectively. They contain the detailed information extended with necessary tables and graphs regarding the experimental and theoretical work for the present thesis. Seventh chapter summaries the complete work done by me and the future outlooks of the present work are given at the end of that chapter.

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