THE MAHARAJA SAYAJIRAO UNIVERSITY OF BARODA

DOCTORAL THESIS

Study of nuclear reactions for structural materials and investigation of reactor shielding

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Executive Summary

Besides the theoretical interests, there is a growing need for nuclear data libraries for various applications in the energy span of 1 to 22 MeV. Neutron-induced reactions are of prime interest from the point of nuclear reaction theory, fission and fusion reactor technology, fast reactor design and control calculations, neutron fluence monitoring, safeguards, neutron therapy, medical physics, activation, and prompt radiation analysis, radionuclides production and applications of data in dosimetry [1]. Advanced reactor systems, such as ADSs and ITER, are being developed by different research and development groups to meet the criteria for clean energy production. The aim is to develop future generation of fission and fusion reactors with upgraded-safety features and economics enhanced resource-use with a minimum amount [2].

To understand and regenerate the operation and performance of fission based power plants, fusion devices, and accelerators, the concerned simulation codes must have a wide range of nuclear data, like cross-section and decay properties for all the materials of interest in the device. Hence, the part of the present proposed work focuses on the nuclear cross-section data where they need to be improved for an application like structural materials for fission reactor and future fusion devices [3]. Structural materials are a very important part of any nuclear reactor. Because they must have the capability of radiation hardness and long durability. As these materials are used for a reactor structure, the neutrons that are produced by fission or fusion mechanism are irradiating the reactor materials. However, in many cases, the data related to the structural materials of interest in reactor applications are still incomplete and have large discrepancies. Therefore, a complete and appropriate amount of data in the cross-section data library is needed. The data is important for ADSs and ITER development and the details about these two major projects are given in the following sections.

By considering the listed facts, it is realized that there is a strong need for the reaction cross-sections data of reactor structural materials based on advanced technologies. 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 a 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, a high-energy proton beam (approximately 1 GeV) strikes a heavy element target like tungsten, lead, or bismuth target, which yields copious neutrons by (p, xn) spallation reaction. Therefore, the spallation target becomes a source of neutrons, which can achieve a self-terminating fission chain in a sub-critical core. Therefore, neutron, as well as proton cross-section data, is required to design different components of the advanced reactor, i.e. structural material, shielding design, waste estimation, and estimation of radiation damage, nuclear heating, transmutation effects, and radiation dose.

The materials selected for the cladding, duct, and core structure must retain their identity in the core environment and also must have low neutron-absorption cross-sections. The later requirement limits the choice of materials to a very few, namely, Indium, Tantalum, Terbium, aluminum, magnesium, zirconium, beryllium, graphite, and thin stainless steel for thermal reactors. In fast reactors, however, the neutron cross-sections are lower, and stainless steels and nickel alloys are used extensively. Control rods are used in nuclear reactors to control the fission rate of uranium and plutonium. Silver-based alloy, cadmium, Europium Hexaboride, hafnium are extensively used for this purpose. The cladding is the outer layer of the fuel rods, standing between the coolant and the nuclear fuel. It is made of corrosion-resistant material with a low absorption cross-section for thermal neutrons. Usually, aluminum alloys (with Cu, Mn, Si, Mg, Mg-Si, Zn, and Li), Zircaloy, Fe-Cr-Al alloys are used for this purpose. Refractory metals are a class of metals that are extraordinarily resistant to heat and wear. Refractory Alloys for High-Temperature Applications are W, Ta, Nb, Mo, V which can be used as nuclear reaction control rods. Neutron-induced reaction cross-sections data for these materials (like Zr, Rh, Gd, Sr, Ce, Ag, Cd, W, Mo, Ta, Ni, Fe, Al, Pb, etc.) are basic quantities for evaluation of the processes in materials under irradiation in nuclear reactors. The neutron-induced reaction cross-section depends on neutron energy, target nucleus, and also a type of reaction (capture, fission, etc.). So for the particular selected neutron-induced reaction, it is required to measure the cross-section with various neutron energies in the reactor operational energy range. The predictive power of nuclear model codes can be validated and improved in comparison with good quality experimental data and turn, the model calculation will provide estimation where no experimental data is available.

Except the fission Reactor (ADSs) systems, International Thermonuclear Experimental Reactor (ITER), is also another option for green energy production. In a fusion reactor like ITER, during the plasma shot, DT fusion reaction will produce around 14.1 MeV neutrons. These neutrons will irradiate the structural materials of the reactor. In ITER, niobium, tin, indium, tungsten, terbium, etc. elements are selected for making different ITER components. 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 [6-11]. 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 crucial to estimate the cross-section of all possible reactions around 14 MeV on different isotopes of the selected materials.

Ever since the beginning of the development of reactor technology by using a variety of radioactive sources, radiation shielding has become an important field of research. This is because radiation is very harmful to living organisms and it should be protected. This can be fixed by three types namely time, distance, and shielding. The most significant type of radiation for which shielding is required in a nuclear reactor is primary γ-rays and neutron, originating within the core itself, and secondary γ-rays produced by neutron interactions with materials external to the reactor core. The materials to be used for shielding design should have homogeneity of density and composition. Many researchers reported that concrete is one of the most common and suitable materials used for reactor shielding as well as for other nuclear facilities like; particle accelerators, medical hospitals containing radioactive isotopes, nuclear power stations, and storing radioactive waste[12-14].

Shielding properties of concrete directly rely on its composition. In addition to this, the additives play an essential role in modifying the properties of concrete, such as structural strength and its radiation shielding capacity. Thus, attenuation increases as the atomic number of the absorber increases because photoelectric interactions are increased in high-Z materials especially for low energy γ, and high-Z materials yield more pair production interactions for high-energy γ. Because of the high-Z effect, lead and concrete are often used to line the walls of X-ray rooms and places mentioned above, and boron-containing compounds such as boron nitride, boron carbide are also incorporated into concrete to increase its effectiveness as a γ and neutron shield [15]. Another interesting point about the popularity of concrete is its hydrogen content, which is most suitable for neutron shielding. Concrete blocks absorb neutrons because of their hydrogen content (approximately 1% unit weight). There are several studies about radiation shield processes using concrete. The kind and quantity of shielding material fluctuate by radiation type, the activity of source, and the dose rate. There are several other factors for the choice of shielding materials such as their fabrication, cost, and weight. Also, materials used for this purpose must be available in the country. In this respect, the studies of the absorption of radiation in materials that are locally available have become an important issue and thus it is desirable to know the effective materials for γ-ray and neutron shielding.

From the above discussion, it is understandable that the data of nuclear reaction cross-section is a primary input in reactor/accelerator technology. Apart from this, the improvement in materials in aspects of γ-ray and neutron shielding is also important. To generate nuclear reaction cross-section data precisely as per our requirement one needs to learn about nuclear reactions, for which, a detailed description is provided in the following section.

Concrete is the most popular material for the shielding of ionizing radiation. It is broadly used in nuclear facilities like reactors, spent nuclear fuel repositories, particle accelerators, radiotherapy rooms, and many more. As a shielding material, concrete is very popular because of its attenuation properties, which can be conveniently changed by changing its chemical composition. Moreover, the fabrication cost is low and can be easily cast in many complex forms with good structural and mechanical properties. These aspects of concrete make it a suitable material for the aforementioned shielding applications. In the past, extensive work has been carried out for the optimization of the key properties of concrete shielding in nuclear applications focused on the improvement of radiation shielding properties by adding suitable admixtures. They are ingredients, added a small fraction to improve the structural strength and the radiation capacity of concrete. Therefore, every effort the study radiation shielding in concrete modified with these types of additives give benefits to the reactor shielding. Studying the effect of low-Z and high-Z additives on the properties of concrete is a vast topic of research. To choose the objectives of this work more achievable, this study is concerned with the γ-rays and neutron parameters of adding low-Z and high-Z additives on the ordinary Portland cement. The study was performed considering both the experimental and simulation techniques. The experimental techniques were used to determine the attenuation parameters of the samples. Whereas the simulations were performed to make an analogy with the experimental measurements and predict the attenuation properties of the studied samples.

Motivation / Objectives

A vast number of experimental work has been performed to examine the neutron-induced reactions of structural materials in low to moderate energy regions. Precise nuclear data about the structural materials are in demand for the prediction of the sustainability upcoming rector technology (ADSs and ITER). The neutron data plays a prominent role in all branches of nuclear science and technology like cancer treatment, positron emission tomography, single-photon emission computed tomography, and many more. The data also plays a crucial role to analyze the experimental parameters which may be used for the lesser long-lived actinides production in an ADSs or fast reactor. Hence, neutron-induced reaction data are crucial to upgrade the present reactor and accelerator technologies. The literature survey on the neutron-induced reaction cross-sections for structural/cladding material like terbium, indium, and tantalum using EXFOR data library [16] shows that only a few measurements are available at higher neutron energies, whereas, the available data have large uncertainties. Moreover, most of the data reported around 14 MeV are due to mono-energetic neutron sources. Apart from 14 MeV [16], the data were reported using quasi mono-energetic sources from the relative measurement method (monitor cross-section). The data thus contains errors that should include the uncertainties from the monitor reaction, which can easily be calculated from covariance analysis. Therefore, the neutron-induced reaction cross-sections were examined for the specific materials useful in the reactor-based cladding, structural, and shielding materials in the energy span of 5 to 20 MeV with the uncertainties and correlation coefficients measurement from covariance analysis.

In addition to this, utilization and generation of radiation are prominent in various nuclear applications, like in fission/fusion reactors for clean energy, therapeutic nuclear medicine, and radioisotopes handling for disease diagnosis and treatment, and space radiation research. During functioning, these fields require an appropriate shielding material to assure the safety and protection of radiation workers from the deleterious effect of undesired radiation exposure. Among all the radiation fields, present work is focused on the improvement of reactor shielding material. Customarily, concrete is widely used for reactor shielding due to its chemical composition holds both light and heavy nuclei, low fabrication cost, abundance, ease of construction, and superior γ-rays and neutron attenuation capability. Also, it has adaptability with the other additive compounds to enhance the shielding performance. In this context, attention has been paid to the production of low-cost, lightweight, and efficient shielding material. So far, many authors have analyzed concrete containing, barite, different hematite-serpentine, and Ilmenite-limonite, rock and concrete, zeolite, blast furnace slag, silica fume, different lime/silica ratio, and many more. Among these, only a few studies have investigated both the γ-rays and neutron shielding parameters for concrete compositions. Taking this as a motivation, we have investigated the shielding properties of the concrete samples with additives of both light and heavy nuclei.

Objective of the Thesis

The following objectives have been taken into considerations in the present work,

- To measure the reaction cross-sections from medium to fast neutron-induced reaction related to reactor structural materials. We have also planned to measure the (n, γ), (n, n') and (n, 2n) reaction cross-sections of a few of the structural and nuclear medicine such as Tb, In, and Ta induced by fast neutrons. The mono-energetic fast neutrons can be produced using $^{7}Li(p, n)$ reaction at BARC-TIFR Pelletron and $^{3}H(^{2}H, n)^{4}He$ reaction at Purnima accelerator facilities.
- The experimental work consists of the irradiation of structural materials (Tb and In) with quasi mono-energetic neutrons within 5-20 MeV energies and of Ta with proton beam at 14.78 MeV energy. The measurements were carried out using neutron activation analysis followed by the offline γ-ray spectrometric technique. The irradiated samples were counted by using HPGe detectors.
- The covariance analysis was also performed to measure the uncertainties and correlations for the neutron-induced reaction cross-sections.
- To analyze the γ-ray and neutron shielding parameters for reactor by using prepared concretes samples with WC and B_4C additives.
- The experimental work consists of attenuation measurements of γ and neutron with ^{60}Co and ^{252}Cf sources at Defence Laboratory Jodhpur.
- The γ-ray and neutron shielding parameters mass attenuation coefficient (μ_m), effective atomic number (Zeff), effective electron density (Neff), half-value layer (HVL), tenth-value layer (TVL), mean free path (MFP), and effective removal cross-section (Σ_R) were calculated using theoretical prediction codes XCOM, MCNP, Auto-Zeff, and NXcom.

Structure of Present Thesis

The thesis work is organized into five chapters. The contents of each chapter are summarized below.

Chapter 1: Introduction

The chapter will provide a preamble that gains insight into the viewpoint of the study of cross-sections on the upcoming reactor structural materials. It gives the reader an idea of the importance of structural materials utilization in the future and the development of underlined accelerator technology. This chapter also provides the need for an essential improvement in reactor shielding materials. The increasing use of accelerators in industry, research, and medical fields demands a deeper knowledge of different aspects of accelerator safety and radiation protection. Later this chapter gives us the profound ideas and motivations essential for the completeness of this work. Lastly, the organization of the thesis is provided.

Chapter 2: Computational Codes

Chapter two deals with the computational aspects of this thesis, it will provide a complete understanding of the various nuclear codes used in the present reaction cross-sections and reactor shielding analysis. A brief discussion about each code will help the reader to find the specific details regarding the codes.

Chapter 3: Neutron Induced Reaction Cross-sections for Structural Materials

The chapter presents all the detailed information regarding the study of reaction cross-sections for structural materials. It provides an outlook on nuclear reactor-generated energy and the importance of structural materials. It gives the reader an idea of the importance of structural materials in upcoming fission and fusion reactor development. This chapter also gives the details of the work being done by different authors/groups in previous years. The chapter also puts light on different nuclear reaction processes useful to understand the current work. At mid-journey, this chapter incorporates all the necessary detailed information regarding the experimental work carried out to measure the reaction cross-sections with a neutron as the incident particle. Later it gives a brief discussion about the measurement techniques used in the present work. With a detailed derivation of the reaction cross-section formalism used for calculations together with the complete details about the error propagation methods in the form of covariance analysis, used to measure the uncertainties in the detector efficiencies and the experimentally measured data. At the end, the present experimental work along with the literature data have been compared with the different nuclear model codes for complete understanding of the underlying of the reaction mechanism. A much-specified discussion will be added about each code which will help the reader to find the specific details regarding the study of neutron-induced reaction cross-sections. Complete calculations is present in the Appendix for better understanding.

Chapter 4: Improvement of Reactor Materials

Similar to chapter 3, this chapter presents all the detailed information regarding the improvement of shielding materials. Firstly, it introduces different forms of radiation, their interaction effects in a various matters, and their shielding aspects. Along with the details of the previously published work being done by different authors/groups. The chapter also puts light on different γ-rays neutron shielding parameters useful to understand the present work. At mid-journey, this chapter incorporates all the necessary detailed information regarding the manufacturing of concrete samples by adding a suitable amount of additives to it. Further, to investigate the γ-rays and neutron radiation parameters on prepared concretes, experimental work was carried out using two different sources ^{60}Co and ^{252}Cf. With a detailed derivation of the γ-rays and neutrons shielding parameters formalism used for calculations together with the complete details about different nuclear codes provides a backbone to the present work. The end of this chapter provided a complete understanding of the different nuclear codes used in the present analysis. A much-specified discussion will be added about each code which will help the reader to find the specific details regarding the study of different parameters used to investigate γ-rays and neutron shielding.

Chapter 5: Summary & Conclusions

This will be the final chapter which will contain the summary and conclusions obtained through the present work. There are still some unexplored areas in this field that need some investigation. The chapter will also describe, in brief, the need and scope for such kind of work for other reactions and materials to improve the nuclear data libraries and shielding materials.

Summary & Conclusions

The measurement of neutron-induced reactions for reactor structural materials is reported in the present thesis. The experimental work has been performed at BARC-TIFR Pelletron and BARC-Purnima accelerator facilities. The neutrons of required energies were generated by using ^{nat}Li(p, n) and ^{3}H(^{2}H, n) reactions at BARC-TIFR Pelletron and BARC-Purnima accelerator, respectively. The generated neutrons from these methods have been found very much suitable for this type of experimental measurement. Further, the off-line γ-ray spectroscopy is adopted for the measurement of the γ-activity of irradiated samples. The irradiated samples were counted after the irradiation, using a p-type single crystal HPGe detector. Here, the γ-detection and detector efficiency of the samples were measured from a common detector setup, hence, the covariance analysis was adopted to measure the correlations and uncertainties in the neutron reaction cross-sections. The overall uncertainties in neutron-induced reaction data were found to be below 19%. The reported data were found to be consistent with the literature data for the entire range of consideration, however, at few energies, negligible variation can be noticed. The outcomes of this work present a better data set for the measured reactions, especially where the literature data contained discrepancies. Moreover, the results were also found to be in agreement with the evaluated and theoretical data from the nuclear code. Another main objective of this thesis was to improve reactor shielding materials by adding low-Z and high-Z additives in concrete. The samples were prepared according to the local manufacturing standards. Total, twenty-one concrete samples were prepared to investigate the shielding parameters of γ-rays and neutron. The experimental work has been performed using ^{60}Co and ^{252}Cf sources at Defence Laboratory Jodhpur. The γ-rays and neutron attenuation parameter were measured using NaI(TI) scintillation and BF_3 detectors. This experimental measurement compared with various theoretical prediction codes, like MCNP, XCOM, Auto-Zeff, and NXcom. The parameters μ_m, Zeff, Neff, HVL, TVL, MFP, and Σ_R were calculated to investigate the present work. The outcomes of the present work show that the modified samples have more advantages as compared to the pristine concrete. These also revealed that shielding parameters strongly rely on the atomic composition and density of the prepared samples. Moreover, the results of the parameters were also found to be in good agreement with the theoretical codes. The specific details for both the study are provided in the following sections, respectively.

Neutron Induced Reactions

The excitation function for the ^{159}Tb(n, γ)^{160}Tb, ^{113}In(n, n')^{113m}In, ^{115}In(n, 2n)^{114m}In and ^{115}In(n, n')^{115m}In, and ^{181}Ta(n, 2n)^{180}Ta reactions were measured for the incident quasi-monoenergetic neutron energies in the span of 5 to 20 MeV varying differently for each case. Basically, the present work pointing that the uncertainties in the monitor reaction data are crucial in the uncertainty calculations. Moreover, The conclusions drawn from each reaction measurement are as follows:

- The $^{159}Tb(n, \gamma)^{160}Tb$ reaction cross-sections were measured relative to the $^{232}Th(n, f)^{97}Zr$ monitor reaction at the neutron energies of 5.08 ± 0.165, 12.47 ±0.825, and 16.63 ±0.95 MeV. The measured cross-sections match well with the literature as well as theoretical data. The uncertainties in the data were obtained with the help of covariance analysis was found to be within the span of 11-14%. Among the total, the major contribution was coming from the counting statistics of the monitor reaction. The results were compared and found to be in agreement with the evaluated data from ENDF/B-VII.1 and JENDL-4.0 libraries. The theoretical prediction from TALYS-1.9 using different level density model, reproduced the trend of the data very well, except for 5.08 ±0.165 MeV, which was found lower compared to the theoretical predictions.
- The $^{113}In(n, n')^{113m}In$, $^{115}In(n, 2n)^{114m}In$ and $^{115}In(n, n')^{115m}In$ reaction cross-sections were measured at neutron energies of 10.95 ±0.67, 13.97 ±0.97, 16.99 ±0.88 and 20.00 ±0.94 MeV. The uncertainties measured using covariance analysis were found to be in the span of 4-19%. The results were found to be in agreement with the literature, evaluated data from JENDL/AD-2017 and the theoretical prediction by using different as well as by adjusting level density models in TALYS-1.9. Default level density model (collective + effective) of TALYS were found unable to emulate the experimental as well as the literature data. It surpassed the trend of data in the threshold to 22 MeV for $^{113}In(n, n')$ and 12 to 22 MeV range for $^{115}In(n, 2n)$ reaction. The results were found to be satisfactory for $^{115}In(n, n')^{115m}In$ reaction.
- The production cross-section for ^{181}Ta is measured at the neutron energy of 14.78 ± 0.20. The $^{197}Au(n, 2n)^{196}Au$ reaction were used as monitor for (n, 2n) reaction cross-section. The uncertainty in the present measurement calculated with the help of covariance analysis was found to be 15%. The present results were found in accordance with the both literature and the evaluated data from JEFF-3.32, ENDF/B-VII.1 libraries. Similar to the above measurement, the theoretical calculations reproduced the datum satisfactorily.

Shielding Material

The investigation of WC and B_4C additives in ordinary concrete for γ-rays and neutron shielding have been performed through parameters μ_m, Zeff, Neff, HVL, TVL, MFP, and Σ_R.

- The measured γ-ray attenuation parameter μ_m data for all the investigated samples were found to be in good agreement with the theoretical prediction XCOM and MCNP.
- The measured value of μ_m is further used to calculate Zeff, Neff, HVL, TVL, and MFP. Among these, the value of Zeff, and Neff are predicted from Auto-Zeff software. The calculated and predicted (Auto-Zeff) trend of Neff are closely related to Zeff and for all the samples their variations with respect to energy are also found similar.
- The results of HVL and TVL parameters show the prepared M1-M5 sample has required a lower volume of the material compared to M6 and M7. Analyzing all these parameters suggested that among the prepared samples M1-M5 has a superior for γ-ray shielding.

- The experimental findings of neutron attenuation were compared with the NXcom prediction and MCNP. The comparison revel that the prepared M1-M6 samples are more effective for neutron attenuation relative to OC.
- The both γ and neutron study emphasized that the compounds of tungsten (WC) and boron B_4C as additive may be a good choice that can be used for the shielding.
- It is suggested that, for radiation containing γ and/or neutron, the materials M1-M5 provide an alternative source/option with ordinary concrete. Which can minimize the required space as well as the effective cost of shielding materials.

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