Chapter 7

Summary & Conclusions

This chapter provides the summary and outcomes of the present work. The chapter is divided into three sections, in which the first two contain the conclusions drawn from the neutron-induced and proton-induced reaction cross-section measurements and the third section gives an insight into the future perspective of the present work. In summary, the data reported in the present thesis are vital for the advancement of the current reactor/accelerator technology, for dose estimation of nuclear waste disposal, in nuclear medicine etc.

The measurement of neutron and proton induced reactions for reactor based applications are reported in the present thesis. The experimental work has been carried out at BARC-TIFR Pelletron and BARC-FOTIA accelerator facilities. The neutrons of required energies were generated by using $^{nat}Li(p,n)$ reaction, which has been found very much suitable for the production of neutrons for such type of experimental measurements. Off-line γ-ray spectroscopy is utilized for post irradiation measurement of the γ-activity of activated samples. A p-type single crystal HPGe detector is used for counting of the samples for both n & p-induced reaction cross-section measurements. Since, a common detector setup was used for the γ-detection and counting, therefore, the correlations and uncertainties in measured (n, x) reaction cross-sections were calculated by using covariance analysis. Covariance analysis has not been done in case of (p, x) reactions as the flux was measured by using a Faraday cup directly, unlike by using a monitor reaction in case of (n, x) reactions, which in turn produces large uncertainties in measured data. The overall uncertainties in both the n & p-induced reaction data were found to be below 20% and 10%, respectively. The reported data were found to be consistent with the literature data for the entire range of consideration, however, at few points minor discrepancies can be noticed. The results from this work present a better data set for the measured reactions, especially where the literature data contained discrepancies. Overall, in addition to literature data, the results were found to be in agreement with the evaluated and theoretical data from nuclear codes, as well. The specific details for each case are provided in the following sections, respectively.

7.1 Neutron-Induced Reactions

The excitation function for the $^{232}Th(n,\gamma)^{233}Th$, $^{100}Mo(n,2n)^{99}Mo$, $^{58}Ni(n,p)^{59}Co$, and $^{58}Ni(n,2n)^{57}Ni$ reactions were measured for the incident quasi-monoenergetic neutron energies within the range of 2-20 MeV varying differently for each case. In general, the present work summarizes that the uncertainties in the monitor reaction data are important in the uncertainty calculations. Different reaction model codes may also produce varying results depending upon the sensitivity of the calculations on level density parameter, mean free parameter and PE process. Furthermore, The conclusions drawn from each reaction measurement are as follows:

- The $^{232}Th(n,\gamma)^{233}Th$ reaction cross-sections were measured relative to the production cross-sections of the fission product ^{97}Zr at average neutron energies of 10.95 ± 0.59, 13.97 ± 0.57, 14.98 ± 0.55, and 18.99 ± 0.65 MeV. The measured cross-sections agreed well with the literature and theoretical data. The uncertainties in the data were obtained by covariance analysis and found to be within the range of $10 - 20\%$. The major contribution were found be 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 results from TALYS-1.9 by using default parameters, successfully reproduced the trend of the data, however, a better fitting was done by modifying the level density parameter values. The TALYS-1.9 was also used for the literature data of $^{232}Th(n,2n)$ reaction with default and modified parameters. The $^{232}Th(n,\gamma)$ cross-section are also found to be suppressed by the $^{232}Th(n,2n)$ reaction cross-section at energies above 7 MeV.

- The $^{100}Mo(n,2n)^{99}Mo$ reaction cross-sections were measured at neutron energies of 10.95 ± 0.59, 13.97 ± 0.57, 14.98 ± 0.55, and 18.99 ± 0.65 MeV. The uncertainties with covariance analysis were found to be in the limit of $12 - 18\%$. The results were found to be in accordance with the literature, evaluated data from ENDF/B-VII.1, JENDL-4.0, JEFF-3.1, CENDL-3.2 and the theoretical data calculated by using different level density models in TALYS-1.9 and the default of ALICE-2014. The results from TALYS-1.9 were found to be satisfactory as compared to the ALICE-2014, which over-predicted the cross-section data for region where the cross-section starts to saturate. A better fit has been found by optimizing the input parameters. No significant PE contribution was observed for the production fo the ^{99}Mo isotope. The TALYS-1.9 is also used for pure CN and CN+PE calculations in order to find the A number of neutron systematics were also being tested alongside the model codes to predict the data at around 14.5 MeV. The results were found to be satisfactory for different isotopes of Mo.

- The production cross-sections for ^{58}Co and ^{57}Ni are measured for the incident neutron energies up to 20 MeV. The $^{197}Au(n,\gamma)^{198}Au$, $^{115}In(n,n')^{115m}In$ reaction were used as monitor for (n, p) reaction, whereas, the $^{27}Al(n,\alpha)^{24}Na$ reaction was used for (n, 2n) cross-sections. The uncertainties with covariance analysis were found within the range of $11 - 16\%$. The present results were found in agreement with the both literature and the evaluated data from ENDF/B-VII.1, JENDL-4.0, CENDL-3.1, and JEFF-3.2 libraries. TALYS-1.9 and EMPIRE-3.2.3-Malta were used for the comparison of the literature data, in which TALYS-1.9 was found suitable to fit the available data. Similar to the case above, the theoretical calculations reproduced the data satisfactorily without the involvement of the PE process.

7.2 Proton-Induced Reactions

The proton induced reaction cross-sections were measured for ^{nat}Nb, ^{nat}Ag, and ^{nat}Ti isotopes by using stack-foil technique degrading a 22 MeV p-beam following off-line γ-ray spectroscopy using a HPGe detector. The beam current was monitored with a Faraday cup and hence the error in the flux was limited to $\approx 3\%$. The overall uncertainty in the measured data has been calculated as the quadratic sum of both the statistical and systematic errors and is found to be $\leq 10\%$. The conclusions drawn from the analysis of the (p, x) reaction cross-sections data are as follows:

- The measured data for all the reported excitation functions were found to be in a good agreement with the present literature and the theoretical calculations using TALYS-1.9 and ALICE-2014 nuclear model codes.
- TALYS-1.9 has been found to be more suitable than the ALICE-2014 model code that was used to generate reaction cross-section data in the incident proton energy regime 10-20 MeV. The TALYS-1.9 produced overall better results as compared to ALICE. The level density model as well as the PE models were found to successful in order to fit the literature data properly.
- Large PE contribution has been noticed for reactions involving few particles in the outgoing channel. This may be attributed to the fact that there is always a greater probability for the nucleus to de-excite by emitting single particle during the thermal equilibrium process of the formed compound nucleus.
- The PE fraction was found to be higher with the increasing excitation energy. The PE fraction also depend on the associated Q-value of the reaction.
- The present data are vital for the charged particle monitoring purposes, to estimate thick target yields and to develop new routes for rare medical isotopes.

7.3 Future Plans

In the present work the experimental study for reaction cross-sections could only be performed up to 20 MeV incident neutron/proton energies as the accelerators used can not be operated above the mentioned energy range. Therefore, similar work can be done with higher energies to produce precise nuclear data and to investigate the energy dependence of reaction cross-section as energies above 20 MeV. Since at higher energies, more and more channels open up and start contributing in total cross-section of a particular reaction, therefore, these kind of studies become necessary to check the performance and adaptability of different nuclear model codes. Attention needs to be given specifically on the use of different neutron generators. It is also proposed to have neutron spectra generation by using simulation codes like MCNP and GEANT4 at higher at incident proton energies above 15 MeV. The neutron spectrum from the $^{7}Li(p,n)$ reaction can also be generated experimentally utilizing n-TOF technique. As the most important conclusion of the present work, the covariance analysis is suggested for the data which is measured relative to other data. Similarly, proton induced reaction can also be performed at energies above 20 MeV to calculate the contribution of different reaction mechanisms like, PE, direct reactions etc. on the production cross-sections of different residues. The study would be useful to refine the existing model codes and to extend them for further higher incident particle energies.

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