Neutron Induced Cross Section and Fission Yield Measurements for Thorium and Uranium

Sadhana Mukerji
Reactor PhysicsPhysics Division, Bhabha Atomic Research Centre, Mumbai - 400085, INDIA
. * *email: mukerjis@barc.gov.in*

The thesis presents the original contribution from the author towards the measurements of basic nuclear data related to neutron induced reaction cross sections and fission fragment yields for thorium and uranium. Experiments were carried out with the quasi-mono-energetic neutrons of 2.18-17.28 MeV from the ^{7}Li(p,n) reaction. The neutron beam was produced by using the 5-20 MeV proton beam from main line at 6 meter height irradiation set up at the 14UD BARC-TIFR Pelletron facility at TIFR, Mumbai, India.[1] The measurements reported here for the 2.18, 3.15, 13.5, 15.5 and 17.24 MeV neutron energies are done for the first time. Neutron energies above 3.15 MeV reported in the thesis correspond to the average neutron flux weighted energy in ^{7}Li(p,,n) reaction. In the present work, measurements of neutron induced reactions are reported for neutrons energy range of 2.18-17.28 MeV. Gamma ray spectroscopy was carried out for a range of fission and reaction products in the neutron induced fission reactions of ^{232}Th and ^{238}U. Contemporary higher multi-group analyses of current reactors, GEN-IV reactors and ADSS demand improvements of the nuclear data for ^{238}U and ^{232}Th and especially precision measurements at higher neutron energies

A. Measurement of reaction cross section

The experiments were carried out at TIFR-BARC Pelletron facility at the 6 meter height irradiation set up of main line. A stack of Ta-Li-Ta was made for the production of neutron beam range of 2.18-17.28 MeV. Gamma ray spectroscopy was carried out for a range of fission and reaction using the ^{7}Li(p, n) reaction. About 1.0 cm^2 of ^{232}Th and ^{238}U metal foils with thickness 344.1-634.2 mg/cm^2 doubly wrapped with 0.025 mm thick Al foil was mounted at zero degree with respect to the beam direction at a distance of 2.1 cm from the location of the Ta-Li-Ta stack. The ^{232}Th and ^{238}U metal foils were irradiated for 9-12 hours with 2.18-17.28 MeV quasi-mono-energetic neutrons generated from the ^{7}Li(p, n) reaction using the 5-20 MeV proton beam. The proton current during the irradiations was around 100-200 nA. After irradiation, the samples were cooled for one hour. Then the irradiated target of ^{232}Th and ^{238}U along with Al wrapper was mounted on Perspex plate and taken for γ-ray spectrometry. The γ-rays of reaction/fission products from the irradiated ^{232}Th and ^{238}U sample were counted in energy and efficiency calibrated 80 c.c. HPGe detector coupled to a PC-based 4K channel analyzer. The counting dead time was kept always less than 5 % by placing the irradiated ^{232}Th and ^{238}U sample at a suitable distance from the detector to avoid pileup effects. The gamma ray counting of the irradiated sample was carried out in live time mode and was followed as a function of time. The ^{238}U(n,γ)^{239}U reaction cross-section at the average neutron energies of 3.15, 13.5, 15.5 and 17.28 MeV and ^{238}U(n, 2n)^{237}U reaction cross-section at the average neutron energies of 13.5, 15.5 and 17.28 MeV have been determined using activation and off-line γ-ray spectrometric technique. Since the neutrons are not mono-energetic, appropriate corrections to the cross sections are incorporated. [2] The experimentally determined ^{238}U(n, γ)^{239}U and ^{238}U(n, 2n)^{237}U reaction cross-sections from present work were compared with the evaluated data of ENDF/B-VII.1(Evaluated Nuclear Data File), JENDL-4.0(Japanese Evaluated Nuclear Data Library), JEFF-3.1(Joint Evaluated Fission and Fusion File) and CENDL-3.1(Chinese Evaluated Nuclear Data Library) nuclear data library. The experimental values were found to be in general agreement with the evaluated values of ENDF/B-VII.1 and JENDL4.0 but different from the values of JEFF-3.1 and CENDL-3.1. It is observed that, the ^{238}U(n, γ) reaction cross-section decreases in the energy range of 100 keV to 8 MeV with a dip at 6-8 MeV. It then increases above the neutron energy of 8.0 MeV and thereafter remains constant up to the neutron energy of 14 MeV. However, the ^{238}U(n,2n) reaction cross-section increases from its threshold to 8MeV. There after remains almost constant up to 14 MeV. Beyond 14 MeV both ^{238}U(n, γ) and ^{238}U(n, 2n) reaction cross-sections show decreasing trend due to opening of (n, 3n) and (n, 2nf) reaction channels. The present data along with literature data in a wide range of neutron energies were interpreted in terms of competition between ^{238}U(n, γ), (n, f), (n, nf) and (n, xn) reactions channels. The ^{238}U(n, γ) and ^{238}U(n, 2n) reaction cross-sections were also compared with theoretical results of TALYS[3] computer code. The TALYS results for ^{238}U(n, γ) reaction cross-section over predict the experimental data within the neutron energy of 1 keV to 3 MeV and thereafter it is in agreement with the experimental data. However, the TALYS results for ^{238}U(n, 2n) reaction cross-sections are in good agreement with the experimental data up to 8 MeV. Above 8 MeV, the ^{238}U(n, 2n) reaction cross-sections remain constant up to 14 MeV. There after it shows decreasing trend due to opening of (n, 3n) and (n, 2nf) reaction channels. The ^{232}Th(n, 2n) cross section for 17.28 MeV has also been determined using the same technique. The experimentally determined ^{232}Th(n, γ) and ^{232}Th(n, 2n) reaction cross-sections from the present work were compared with the evaluated data of ENDF/B-VII.1 and JENDL-4.0 and were found to be in good agreement. The present data along with literature data in a wide range of neutron energies were interpreted in terms of competition between ^{232}Th(n, γ), (n, f), (n, nf) and (n, xn) reactions channels. The ^{232}Th(n, γ) and ^{232}Th(n, 2n) reaction cross-sections were also calculated theoretically using the TALYS computer code and were found to be in good agreement with the experimental data of present work but are slightly higher than the literature data at lower neutron energies. The ^{232}Th(n, γ) reaction cross section shows decreasing trend up to the neutron energy of 6-8 MeV. then increasing and remaining constant up to 14 MeV. The ^{232}Th(n,2n) reaction cross section increases from its threshold energy to 8 MeV and then remains constant up to 14 MeV. After 14 MeV both ^{232}Th(n, γ) and ^{232}Th(n,2n)[4] reaction cross sections decrease with neutron energy due to opening of (n, 3n) and (n, 2nf) reaction channels.

B. Measurement of Fission Product Yield data

At present, the available experimental data of fission product yields at different incident neutron energies is insufficient for the development of fission systematics, whereas the theoretical models of fission are still not well understood. Therefore, measurements are needed for high energy neutron-induced fission of ^{232}Th and ^{238}U[5]. Further, not many experimental data of fission product yields are available in the neutron energies range of 16-20 MeV. In the present study, fission product yield measurement has been carried out by neutron irradiation at 9.32 MeV and 12.52 MeV energies on ^{232}Th [6]and ^{238}U[7] targets by using off-line gamma ray spectroscopy. Activated targets are counted in highly shielded HPGe detectors over a period of several weeks to identify decaying fission products. The fission yields values are reported for twelve fission products. The results obtained from present work at 12.53 MeV have been compared with the similar data of mono-energetic neutrons of comparable energy of 14.1 MeV from literature and are found to be in good agreement. The fission products yields data at the neutron energy of 9.25 MeV from the present work are determined for the first time. There are no data available in literature around the neutron energy of 9.25 MeV to compare with the data of present work. The mass yield distribution in the symmetric valley region shows the presence of a third peak.

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