Determination of ^{241}Pu(n,f) cross sections by the surrogate-ratio method

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The ^{241}Pu(n,f) cross sections have been determined by the surrogate ratio method in the equivalent neutron energy range 11.0–16.0 MeV by using ^{238}U(^{6}Li,d)^{242}Pu and ^{232}Th(^{6}Li,d)^{236}U transfer reactions at E_{lab} = 39.6 and 39.0 MeV, respectively. Results have been compared with direct measurement of ^{241}Pu(n,f) cross-section data and predictions of statistical model code EMPIRE 3.1 for different sets of values of fission barrier heights from evaluated data libraries. The present ^{241}Pu(n, f) cross-section data are observed to be consistent with the direct measurements, suggesting the applicability of surrogate methods. However, the EMPIRE 3.1 predictions are not in complete agreement with the experimental data in the neutron energy range 2.0–20.0 MeV for any set of the fission barrier data libraries. The consistency of experimental results on ^{241}Pu(n,f) cross section data from direct and surrogate measurements suggests the need for fission barrier heights for Pu isotopes that are different from those used in standard libraries. The fission barrier heights for various Pu isotopes have been obtained for best fit to the experimental data in the neutron energy range 2.0–20.0 MeV.

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I. INTRODUCTION

Renewed interest in nuclear energy has led to design of new reactor systems based on fast neutron induced fission which take advantage of advances in nuclear technology [1]. These future reactors promise enhanced safety, reliability, sustainability, and waste reduction. An important component of the research and development for these reactor concepts is the improvement of the fundamental nuclear cross-section data. The transuranic nuclide produced in the nuclear fuel cycles by successive neutron capture plays a prominent role in these new designs. Accurate data sets, especially neutron induced reaction cross sections with these unstable isotopes taking place at energies from several keV to tens of MeV, are important for designing and engineering of these reactor systems [1,2]. Fast neutron reactions have also been proposed for the incineration of actinide materials, notably minor actinide isotopes which are produced in Th-U or U-Pu fuel cycles. The spent fuel will be burned in a dedicated reactor, where neutron reactions such as (n,f) or (n,2n) can be used to reduce the content of radio-toxic isotopes [3]. A number of either new or improved neutron induced reaction cross-section measurements are needed to determine the feasibility, effectiveness, and safety issues for the efforts proposed for these applications. Not all relevant data can be directly measured in the laboratory or accurately determined by calculations. Direct measurements may encounter a variety of difficulties: many of these nuclei are too difficult to produce with currently available experimental techniques or too short lived to serve as targets in a present day experimental setup [2,4]. Also, the sufficient flux of neutron beams of the required energy regime is often inaccessible.

The surrogate nuclear reaction technique [5,6] is an indirect method that allows the determination of cross sections for compound nuclear reactions involving difficult to produce targets. In this approach the compound nucleus (say B*) occurring in the reaction of interest (a + A → B* → c + C) is produced via an alternative reaction, called a "surrogate reaction" (d + D → B* + b), which involves a stable projectile-target combination (d + D) that is experimentally more accessible. In the actinide region, short-lived isotopes often have longer-lived neighbors that are suitable candidates for use in a surrogate experiment. The charged particle reaction on these neighboring isotopes can be used to form the same compound nucleus as that of the desired neutron induced reactions. The decay of the compound nuclear state is assumed to be independent of the production mechanism, allowing charged particle reactions with neighboring isotopes to be used as surrogates for the neutron induced reaction of interest. In recent years, the surrogate reaction method in various forms, such as (i) the absolute surrogate method [6,7], (ii) the surrogate ratio method (SRM) [8–10], and (iii) the hybrid surrogate ratio method [11], has been employed successfully to get the indirect estimate of the compound nuclear reaction cross sections in the actinide region.

Recently, Tovesson and Hill [12] have reported ^{241}Pu(n,f) cross sections by direct measurement using the neutron time-of-flight technique. Discrepancies of up to 30% between the experimental data and evaluation by the standard library are found in the neutron energy range 10–20 MeV [3]. The challenge of obtaining the fission cross section of ^{241}Pu lies in its relatively short half-life of 14.4 yr. The natural decay makes the sample difficult to handle and produces a large background component in the measurements. Because of the importance of ^{241}Pu in nuclear energy applications, it is suggested to carry out further confirmatory measurement on ^{241}Pu(n,f) cross sections,

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particularly in the fast neutron region [3]. With a motivation to probe the observed discrepancy between measured and evaluated data on ^{241}Pu(n,f) cross sections, we have carried out cross-section measurements by employing the SRM.

In the past, the neutron induced compound nuclear fission cross sections for unstable actinide targets have been accomplished by surrogate ratio methods, using light charged particle induced reactions like p, d, or t. In the present work, we have used a ^{6}Li beam to produce ^{242}Pu and ^{236}U compound nuclei at similar excitation energies by ^{238}U(^{6}Li,d)^{242}Pu and ^{232}Th(^{6}Li,d)^{236}U transfer reactions at $E_{lab} = 39.6$ and 39.0 MeV, respectively. The ^{242}Pu system is the surrogate of desired reaction $n + ^{241}$Pu, and similarly ^{236}U is the surrogate of $n + ^{235}$U and used as a reference monitor.

II. EXPERIMENTAL DETAILS AND DATA ANALYSIS

The self-supporting ^{238}U and ^{232}Th targets of thicknesses 2.3 and 1.3 mg/cm^2 were bombarded with ^{6}Li beams of energies 39.6 and 39.0 MeV, respectively, from the Bhabha Atomic research Centre – Tata Institute of Fundamental Research 14-MV Pelletron Accelerator at Mumbai. The ^{242}Pu and ^{236}U compound nuclei are formed in ^{238}U(^{6}Li,d)^{242}Pu and ^{232}Th(^{6}Li,d)^{236}U transfer reactions, which serve as surrogates to $n + ^{241}$Pu $\rightarrow$ ^{242}Pu and $n + ^{235}$U $\rightarrow$ ^{236}U, respectively. A solid-state ΔE-E detector telescope, with ΔE being the detector of thickness 150 μm and E being the detector of thickness 1.0 mm, was kept at $\theta_{lab} = 85^\circ$ around the transfer grazing angle to identify projectilelike fragments (PLFs). A 3.86-mg/cm^2-thick aluminum foil was placed in front of the particle telescope to stop the fission fragments and thereby protect the ΔE detector from damage. The proton, deuteron, triton, and α are uniquely identified by plotting ΔE against the residual energy in the E detector (E_{res}). This plot was transformed to create an effective particle identification (PI) [13] versus total-energy plot, which was generated using the linearization function (PI $= (bE_{tot}^{1.73} - E_{res}^{1.73})$, where E_{tot} is the total particle energy, E_{res} is the energy deposited in the E detector, and b is a constant. Figure 1 shows a typical PI versus total-energy plot, where all the PLFs are clearly identified. A large area solid-state detector of 450 mm^2 area was centered at angle 160° with respect to beam direction and subtended a solid angle of 63 msr with an angular opening of 16° to detect the fission fragment in coincidence with PLFs. A typical fission-fragment spectrum in coincidence with deuteron PLFs, obtained in ^{238}U(^{6}Li,d)^{242}Pu* $\rightarrow$ fission, is shown in Fig. 2. The time correlation between PLFs and fission fragments is recorded through a time-to-amplitude converter (TAC). The deuteron gated correlation between the fission-fragment pulse height and TAC is shown in Fig. 3. The ground-state Q values (Q_{gg}) for ^{238}U(^{6}Li,d)^{242}Pu and ^{232}Th(^{6}Li,d)^{236}U transfer reactions are -6.458 and -6.046 MeV, respectively. Hence, the ^{242}Pu and ^{236}U compound systems are populated at overlapping excitation energies in ^{6}Li + ^{238}U and ^{6}Li + ^{232}Th transfer reactions. The ratios of PLF and fission coincidence to deuteron singles counts in ^{232}Th(^{6}Li,d)^{236}U and ^{238}U(^{6}Li,d)^{242}Pu transfer reactions correspond to the fission decay probability of the compound systems ^{236}U and ^{242}Pu, respectively, by the following relation:

$$\Gamma_f^{CN} = \frac{N_{d-f}}{N_d}. \quad (1)$$

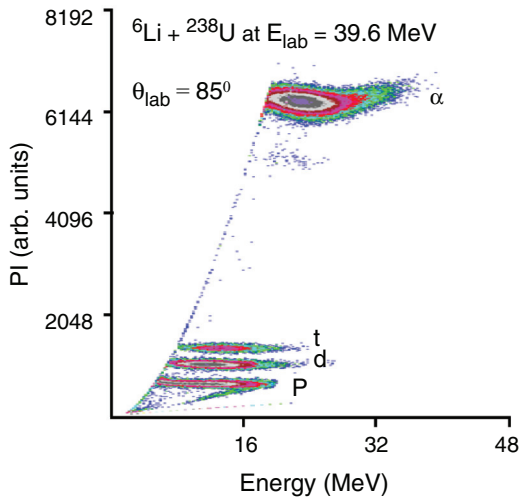

FIG. 1. (Color online) Typical PI vs E plot for particle identification.

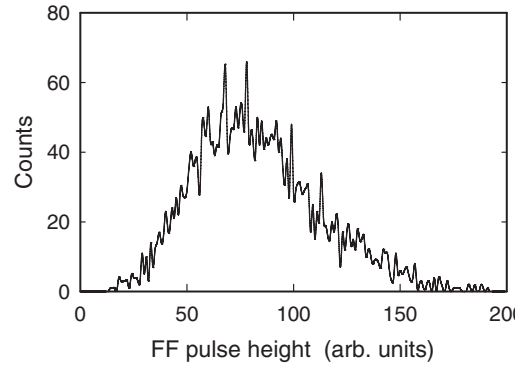

FIG. 2. Typical fission-fragment (FF) pulse height spectrum obtained in coincidence with deuteron in the ^{238}U(^{6}Li,d)^{242}Pu $\rightarrow$ fission reaction.

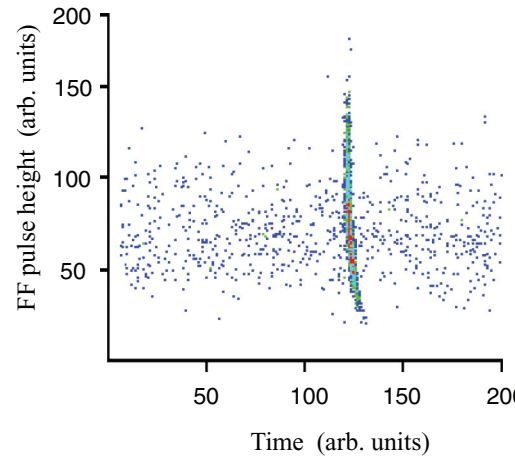

FIG. 3. (Color online) The fission-fragment (FF) pulse height vs time correlation between deuteron and FFs.

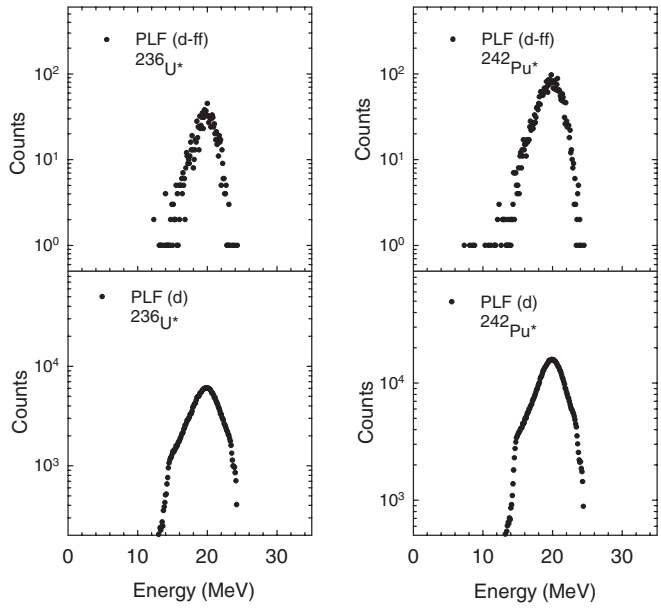

FIG. 4. Excitation energy spectra of compound systems ^{236}U and ^{242}Pu produced in ^{232}Th(^{6}Li,d)^{236}U and ^{238}U(^{6}Li,d)^{242}Pu reactions with (upper) and without (bottom) coincidence with fission fragments.

The ΔE and E silicon detectors were energy calibrated by using a ^{228,229}Th source and in an in-beam experiment that made use of the discrete α-particle peaks corresponding to ^{15}N* states from the ^{7}Li(^{12}C, α)^{15}N* reaction at a ^{7}Li beam energy of 18.0 MeV. The excitation energies of the targetlike residues, ^{242}Pu and ^{236}U populated in ^{6}Li + ^{232}Th and ^{6}Li + ^{238}U, were determined by employing two-body kinematics corresponding to outgoing deuteron PLFs channels. The excitation energy spectra so obtained for ^{242}Pu and ^{236}U nuclei are shown in Fig. 4. The ratios of coincidence to singles counts were determined in steps of 1.0-MeV excitation energy bins in the excitation energy range 17.0–22.0 MeV for ^{242}Pu and ^{236}U nuclei. The relative fission probabilities of the compound nuclei are then multiplied with the relative neutron induced compound nuclear formation cross sections of $\sigma^{\rm CN}_{n+^{241}{\rm Pu}}$ and $\sigma^{\rm CN}_{n+^{235}{\rm U}}$ to obtain the ratio of compound nuclear reaction cross sections at the same excitation energies of $n + {}^{241}$Pu $\rightarrow$ ^{242}Pu $\rightarrow$ fission and $n + {}^{235}$U $\rightarrow$ ^{236}U $\rightarrow$ fission reactions, as follows:

$$\frac{\sigma_f^{n+^{241}{\rm Pu}\rightarrow^{242}{\rm Pu}}(E_{\rm ex})}{\sigma_f^{n+^{235}{\rm U}\rightarrow^{236}{\rm U}}(E_{\rm ex})} = R(E_{\rm ex}) = \frac{\sigma^{\rm CN}_{n+^{241}{\rm Pu}}(E_{\rm ex})}{\sigma^{\rm CN}_{n+^{235}{\rm U}}(E_{\rm ex})}\frac{\Gamma_f^{^{242}{\rm Pu}}(E_{\rm ex})}{\Gamma_f^{^{236}{\rm U}}(E_{\rm ex})}. \quad (2)$$

At high neutron kinetic energies, generally 8.0 MeV or more, since the resonance scattering is negligible, the total cross section for absorption plus inelastic scattering approaches the geometrical cross section of the nucleus [14,15]. Therefore, the ratio of neutron induced compound nucleus formation cross sections for ^{242}Pu and ^{236}U in Eq. (2) is taken as the ratio of $A^{2/3}$ values of corresponding target nuclei. The $n + {}^{235}$U $\rightarrow$ ^{236}U $\rightarrow$ fission reaction cross-section values as functions of excitation energy were used as the reference reactions, which were derived from Evaluated Nuclear Data File (ENDF)/B-VII.0 [16] using the neutron separation energy of ^{236}U ($S_n = 6.545$ MeV). Using Eqs. (1) and (2) the ^{241}Pu(n,f) cross sections as functions of the excitation energy of ^{242}Pu were obtained over the excitation energy range 17.0–22.0 MeV. The excitation energy was scaled down to the equivalent neutron energy range 11.0–16.0 MeV in the laboratory frame by using Eq. (3) [7], where S_n is the neutron separation energy of ^{242}Pu ($S_n = 6.309$ MeV) and A is the atomic mass number of ^{241}Pu:

$$E_n = \frac{A+1}{A}(E_{\rm ex} - S_n). \quad (3)$$

III. RESULTS AND DISCUSSION

The EMPIRE 3.1 [17] calculations have been carried out to quantitatively understand the ^{241}Pu(n,f) cross section over the neutron energy range 1.0–20.0 MeV. The inner and outer fission barrier heights of a double humped fission barrier for the Pu isotopes required for the EMPIRE 3.1 calculations were taken from the Reference Input Parameter Library (RIPL)-1 [18] and RIPL-3 [19] standard library of fission barrier heights for actinides as given in Table I. The ^{241}Pu(n,f) cross section as a function of neutron kinetic energy along with the directly measured ^{241}Pu(n,f) cross section by Tovesson and Hill [12] and the calculated cross section by the EMPIRE 3.1 code for RIPL-1 and RIPL-3 are shown in Fig. 5. Also shown in Fig. 5 are the predictions of the EMPIRE 3.1 code for the fission barrier heights obtained from the barrier formula (BF) [20], which has been fitted to reproduce the fission barriers given by Bjornholm and Lynn [21]. The present experimental results on ^{241}Pu(n,f) cross sections by the surrogate ratio method follow closely the

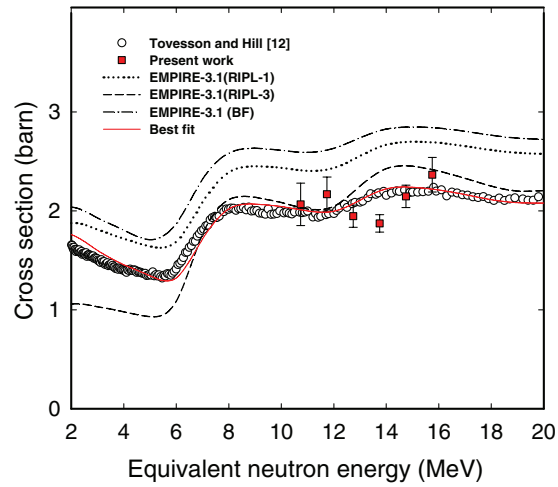

FIG. 5. (Color online) Experimental ^{241}Pu(n,f) cross sections, present measurement (solid squares) and from direct measurement work of Tovesson and Hill [12] (open circles). Results are calculated using the EMPIRE 3.1 code for fission barrier libraries RIPL-1 (dotted line) and RIPL-3 (short-dashed line), the barrier formula (BF) (dash-dotted line), and the best fit (solid line).

TABLE I. Fission barrier heights used in EMPIRE 3.1 calculations.

System	Inner barrier height (MeV)				Outer barrier height(MeV)			
	RIPL-1	RIPL-3	BF	Best fit	RIPL-1	RIPL-3	BF	Best fit
^{242}Pu	5.85	6.02	5.67	5.35	5.05	5.61	5.04	5.40
^{241}Pu	6.15	5.85	6.00	6.22	5.50	5.81	5.24	5.85
^{240}Pu	6.05	5.89	5.68	6.30	5.15	5.73	4.99	5.82
^{239}Pu	6.20	5.96	6.00	6.30	5.70	5.86	5.17	5.96

reported direct measurement by Tovesson and Hill [12]. This supports the applicability of the surrogate technique for fast neutron induced fission cross sections in the neutron energy range 11.0–16.0 MeV as measured in the present experiment. A large discrepancy is observed between experimental and calculated cross sections by the EMPIRE 3.1 code, for RIPL-1 library and fission barriers obtained from BF. Similar behavior has also been reported by Tovesson and Hill [12]. However, if one performs EMPIRE 3.1 calculations for fission barriers obtained from RIPL-3, which is a recent fission barrier library for actinides, the calculated fission cross sections give reasonably good agreement with experimental data at higher energies, but significant discrepancy is observed at energies below 7.0 MeV, as shown in Fig. 5. The RIPL-3 inner and outer barrier heights were further fine tuned to obtain a best fit to the experimental data. This fit required the inner and outer barrier heights for first chance fission to be reduced and those for second and third chances to be increased. The values of barrier heights for the best fit to the experimental data are also given in Table I.

IV. SUMMARY

In summary, we have measured the ^{241}Pu(n,f) cross section by employing the surrogate ratio method. The ^{6}Li + ^{238}U $\rightarrow$ ^{242}Pu* $\rightarrow$ fission and ^{6}Li + ^{232}Th $\rightarrow$ ^{236}U* $\rightarrow$ fission reactions were used as surrogates of desired n + ^{241}Pu $\rightarrow$ ^{242}Pu* $\rightarrow$ fission and n + ^{235}U $\rightarrow$ ^{236}U* $\rightarrow$ fission reactions. Compound nuclei ^{242}Pu* and ^{236}U* were populated at overlapping excitation energies, and fission decay probabilities were measured in the excitation energy range 17–22 MeV. The SRM approach has been used to determine the ^{241}Pu(n,f) cross section in the equivalent neutron energy range 11.0–16.0 MeV by taking the corresponding energy ^{235}U(n,f) cross-section values as the reference reaction taken from ENDF/B-VII. The experimental results on ^{241}Pu(n,f) cross sections by the surrogate reaction technique compare well with reported direct measurements [12]. The observed discrepancy between the experimental ^{241}Pu(n,f) cross-section values and calculated cross sections using EMPIRE 3.1 suggests the need for fission barrier heights for Pu isotopes that are different from those used in the RIPL-1 and RIPL-3 standard libraries or derived from BF. The RIPL-3 fission barrier heights have been adjusted to obtain a best fit to the experimental data in the neutron energy range 2.0–20.0 MeV.

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