Determining ^{234}Pa(n,f) cross sections using the surrogate method

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(Received 19 November 2013; revised manuscript received 15 January 2014; published 13 February 2014)

The fission decay probabilities of the ^{235}Pa and ^{236}U compound nuclei produced in a single experiment in ^{232}Th(^{7}Li, αf)^{235}Pa and ^{232}Th(^{7}Li, tf)^{236}U transfer induced fission reaction channels, have been measured at $E_{\rm lab} = 39.5$ MeV in the excitation energy range of 14–20 MeV. The ^{234}Pa(n,f) cross sections are then deduced from the measured fission decay probability ratios of ^{235}Pa and ^{236}U compound systems in the equivalent neutron energy range of 8–14 MeV within the framework of the hybrid surrogate ratio method, considering the well-measured ^{235}U(n,f) cross sections as the reference. The experimental data on ^{234}Pa(n,f) cross sections have been compared with the calculated fission cross sections using EMPIRE-3.1 code with the fission barrier height values obtained from barrier formula (BF) as well as RIPL-3 [24]. The present experimental results are found to be in very good agreement with the EMPIRE-3.1 predictions for the fission barrier heights predicted by the BF.

DOI: 10.1103/PhysRevC.89.024606 PACS number(s): 24.50.+g, 24.87.+y, 25.85.Ec, 28.20.−v

I. INTRODUCTION

^{234}Pa is one of the nuclide present in the Th-U fuel cycle [1,2], for which no experimental neutron induced fission cross sections data are available for the fast neutron induced compound nuclear fission reactions. The ^{234}Pa$(n,\ f)$ cross sections are considered to be difficult to measure directly, due to the very short half-life (6.7 h) of the ^{234}Pa nucleus. Surrogate reaction methods [3,4] obviate this issue which is a specific indirect method, that combines experiment with reaction theory to obtain cross sections for compound nuclear reactions involving short-lived radioactive targets. In this method, the desired compound nucleus is populated through a direct reaction involving a stable target and projectile and the decay probability of the compound nucleus is experimentally measured. The neutron induced fission cross section is inferred by using the compound nuclear capture cross section value from the theory and fission decay probability value from experiment with the assumption that the decay of the compound nucleus is independent of the angular momentum and parity of the populated state which is known as the Weisskopf-Ewing limit of Hauser-Feshbach theory [5,6]. The surrogate reaction method in its various forms, such as absolute surrogate method [3,4,7], surrogate ratio method (SRM) [8–13], and hybrid surrogate ratio method (HSRM) [14,15], have been used in the past to determine neutron induced fission cross sections for the reactions involving unstable targets. In our earlier works, we have determined the neutron induced fission cross sections for the ^{233}Pa [14],^{239}Np, and ^{240}Np [15] nuclei using the HSRM. In the present work the ^{234}Pa(n,f) cross sections have been determined by employing the HSRM where the ^{232}Th(^{7}Li, αf)^{235}Pa reaction has been used as a surrogate of $n +^{234}$ Pa $\rightarrow$ fission and similarly the ^{232}Th(^{7}Li, tf)^{236}U reaction as a surrogate of the $n +^{235}$ U $\rightarrow$ fission reaction. More detailed discussions on hybrid surrogate ratio methods can be found in [14,15]. Here we only present the experimental details and the essential steps of the data analysis adopted to determine ^{234}Pa(n,f) cross sections.

II. EXPERIMENTAL DETAILS AND DATA ANALYSIS

The experiment was performed at Bhabha Atomic Research Centre - Tata Institute of Fundamental Research (BARC–TIFR) 14-MV Pelletron accelerator facility at Mumbai. A self-supporting ^{232}Th target of thickness 1.3 mg/cm^2 was bombarded with ^{7}Li beam of energy 39.5 MeV. The two silicon surface barrier ΔE-E detector telescopes T_1 and T_2 with ΔE detectors of thicknesses 150 μm and 100 μm and identical E detectors of thicknesses 1 mm were mounted in a reaction plane at angles of 85° and 105° with respect to the beam direction to identify the projectile-like fragments (PLFs). A aluminum foil of thickness 4.0 mg/cm^2 was placed in front of the detector telescopes to stop the fission fragments and thereby protect the ΔE detectors from radiation damage. A large area (450 mm^2) solid state detector was kept at an angle of 160° with respect to the beam direction and subtended a solid angle of 63 msr with an angular opening of 16° to detect the fission fragments in coincident with PLFs. The telescopes were energy calibrated by using a ^{228,229}Th α source and an in-beam experiment that made use of the discrete α-particle peaks corresponding to ^{15}N* states in the ^{12}C(^{7}Li, α)^{15}N* reaction at ^{7}Li beam energy of 18.0 MeV.

The compound nuclei ^{235}Pa and ^{236}U are formed *in situ* in ^{232}Th(^{7}Li,α)^{235}Pa and ^{232}Th(^{7}Li,t)^{236}U transfer reactions are identified by outgoing α and triton PLFs, respectively. The ground state Q values (Q_{gg}) for the above reactions are 5.642 and −7.040 MeV, respectively. The ^{7}Li beam energy of $E_{\rm lab} =$ 39.5 MeV was chosen so that the ^{235}Pa and ^{236}U compound systems were populated at overlapping excitation energies. The excitation energy spectra for the ^{235}Pa and ^{236}U compound systems are obtained by employing two-body kinematics for the outgoing PLFs α and triton, respectively. The excitation energy spectra of ^{235}Pa and ^{236}U compound systems for PLF-fission coincidence and PLF singles are shown in Fig. 1.

The fission decay probabilities of ^{235}Pa and ^{236}U compound systems are determined in steps of 1.0 MeV excitation energy

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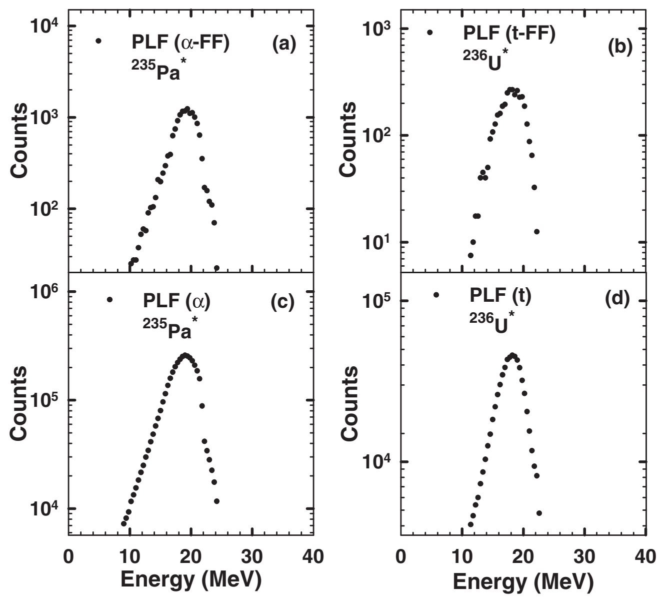

FIG. 1. Excitation energy spectra of compound systems ^{235}Pa and ^{236}U produced in ^{232}Th(^{7}Li, αf)^{235}Pa and ^{232}Th(^{7}Li, tf)^{236}U reactions, for PLF-fission coincidence (a) and (b) and PLF singles (c) and (d), respectively.

bin by taking the ratio of the number of coincidence events between the outgoing PLF-fission fragment coincidence to the number of PLF singles, using the relation

$$P_f^{CN}(E_{\rm ex}) = \frac{N_{i-f}(E_{\rm ex})}{N_i(E_{\rm ex})}, \quad (1)$$

where i denotes the α or triton PLF channels. For each excitation energy bin of 1.0 MeV, the ratio of the fission decay probability of the ^{235}Pa and ^{236}U compound nuclei is determined. The relative fission probabilities of the compound nuclei are then multiplied with the ratio of the corresponding neutron induced compound nucleus formation cross section $\sigma^{CN}_{n+^{234}\mathrm{Pa}}(E_{ex})$ and $\sigma^{CN}_{n+^{235}\mathrm{U}}(E_{ex})$, to obtain the ratio of the compound nuclear reaction cross section at the excitation energies of $n + ^{234}$Pa $\rightarrow$ ^{235}Pa $\rightarrow$ fission and $n + ^{235}$U $\rightarrow$ ^{236}U $\rightarrow$ fission reactions as follows [14]:

$$\frac{\sigma_f^{n+^{234}\mathrm{Pa}\rightarrow^{235}\mathrm{Pa}}(E_{ex})}{\sigma_f^{n+^{235}\mathrm{U}\rightarrow^{236}\mathrm{U}}(E_{ex})} = R(E_{ex}) = \frac{\sigma^{CN}_{n+^{234}\mathrm{Pa}}(E_{ex})}{\sigma^{CN}_{n+^{235}\mathrm{U}}(E_{ex})}\frac{P_f^{^{235}\mathrm{Pa}}(E_{ex})}{P_f^{^{236}\mathrm{U}}(E_{ex})}. \quad (2)$$

The well-measured $n + ^{235}$U $\rightarrow$ ^{236}U $\rightarrow$ fission cross sections in the neutron energy range of 0–30.0 MeV are taken from ENDF/B-VII.1 [16] and used as the reference reaction with energy scale converted to excitation energy by adding the neutron separation energy of ^{236}U ($S_n = 6.545$ MeV). The neutron induced compound nuclear formation cross sections for ^{234}Pa and ^{235}U nuclei have been calculated at corresponding excitation energy using the EMPIRE-3.1 code [17] with optical model potential parameters (OMP) taken from RIPL catalog no. 2408 for Capote *et al.* [18] and the ratio $\frac{\sigma^{CN}_{n+^{234}\mathrm{Pa}}(E_{ex})}{\sigma^{CN}_{n+^{235}\mathrm{U}}(E_{ex})}$ is determined. The sensitivity of the compound nucleus formation cross section ratio, on the various OMP inputs has been investigated for OMP inputs taken from Reference Input Parameter Library (RIPL) catalog nos. 2408 for Capote *et al.* [18], 600 for Vladuca *et al.* [19], 604 for Maslov *et al.* [20], 2303 for Ignatyuk *et al.* [21], and 2601 for Soukhovitskii *et al.* [22]. It is found that the ratio of compound nucleus formation cross sections, calculated using various OMP inputs, can introduce an uncertainty in the range of 2–3%. It may be noted that there is a possibility of ^{7}Li breaking up into the α and triton in the Coulomb field of the target nuclei, with either α or triton getting detected in the ΔE-E telescope, without the complimentary part getting captured in the target nucleus. In the presence of projectile breakup the value of decay probabilities of ^{235}Pa and ^{236}U compound systems are affected. But in the present method, the ratio of decay probabilities is considered for the determination of the desired reaction cross section as given in Eq. (2). Therefore, the possible effect due to projectile breakup gets canceled if the fraction of the contribution is similar for both the transfer channels. In the past, this aspect also has been theoretically investigated by Chiba *et al.* [23] where the surrogate ratio method is shown to be a robust method in the presence of the breakup contribution to the PLF. The ^{234}Pa(n, f) cross sections as a function of excitation energy are obtained over the energy range of 14–20 MeV, using Eq. (2). Finally the excitation energies are scaled down by subtracting the ^{235}Pa neutron separation energy (S_n = 6.123) to obtain the ^{234}Pa(n,f) cross sections at the appropriate equivalent neutron energies. The present experimental results for the ^{234}Pa(n, f) cross sections in the equivalent neutron energy range of 8–14 MeV are shown in Fig. 2.

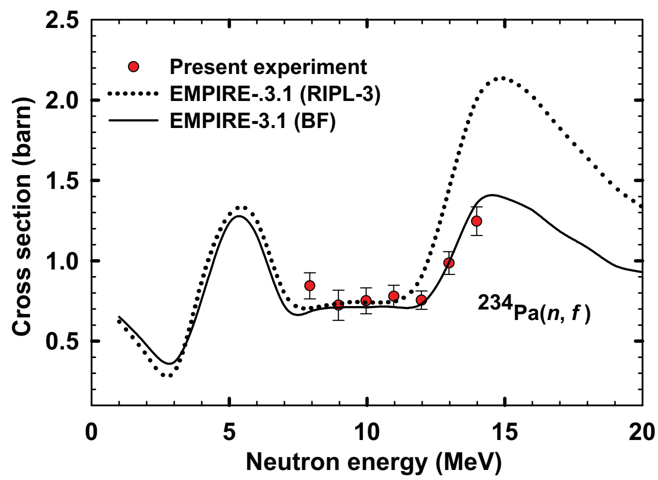

FIG. 2. (Color online) The present experimental ^{234}Pa(n,f) cross sections (solid circles). Calculated results using the EMPIRE-3.1 code for the fission barrier heights obtained from the barrier formula (BF) (solid line) and RIPL-3 [24] (dotted line) are also shown.

III. RESULT AND DISCUSSIONS

The statistical model calculations using the EMPIRE-3.1 [17] code are performed to quantitatively understand the

TABLE I. List of parameters for the barrier formula.

$i = a/b$	a	b
B_{si}	0.0317	0.1029
B_{ci}	−0.0165	−0.0626
π (MeV)	0.1199	0.2497
ν (MeV)	0.0132	0.0650
k (MeV)	−10.2761	−28.8118
δ_z(MeV)	−0.1553	−0.2088
δ_n (MeV)	−0.3224	−0.2183

present experimental results on ^{234}Pa(n, f) cross sections over the neutron energy of 1–20 MeV. For the transthorium elements, the fission process is governed by a double humped barrier transmission. The EMPIRE-3.1 predictions on neutron induced fission cross sections are very sensitive to the fission barriers of the fissioning nucleus. The value of inner- and outer-fission barrier heights of a double humped fission barrier for the various Pa isotopes used in the EMPIRE-3.1 calculations considering a contribution up to third chance fission are taken from the Reference Input Parameter Library (RIPL)-3 [24] and also predicted by the barrier fFormula (BF) [25,26] as given below:

$$V_{i=a/b} = a_s B_{si} A^{2/3} + a_c B_{ci} \frac{Z^2}{A^{1/3}} + \pi Z + \nu N + k + 0.5[1 + (-1)^Z]\delta_z + 0.5[1 + (-1)^N]\delta_n, \quad (3)$$

where $V_{i=a/b}$ denotes the inner barrier (V_a) and outer barrier (V_b) of the the fissioning nucleus. The first two terms in Eq. (3) are surface and Coulomb terms. The value of parameters a_s and a_c are given by $a_s = 19(1 - 2.84[\frac{N-Z}{A}]^2)$ MeV and $a_c = 0.72$ MeV. The changes of surface and Coulomb terms B_{si} and B_{ci} due to the deformation are taken from the work of Brack *et al.* [27] and their values are listed in Table I. The next three terms are the microscopic ones and satisfy the Hugenholtz–Van Hove theorem [28]. The last two terms denote the pairing effects. The associated five parameters π, ν, k, δ_z, and δ_n are obtained by least square fitting to the fission barrier data given by Bjornholm and Lynn [29] for $Z = 89$ to $Z = 98$ are also given in Table I. Using the above expression of BF, the fission barriers for various systems have been calculated in the present work.

The values of fission barrier heights obtained from RIPL-3 and the BF for various Pa isotopes, used in EMPIRE-3.1 calculations are listed in Table II. The values of fission barrier heights for ^{233}Pa isotopes are taken from the earlier work

TABLE II. The values of fission barrier heights of the various Pa isotopes, used in EMPIRE-3.1 calculations for BF and RIPL-3 calculations.

System	Inner barrier height (MeV)		Outer barrier height (MeV)	
	BF	RIPL–3	BF	RIPL–3
^{235}Pa	5.86	5.86	6.18	6.18
^{234}Pa	6.2	6.3	6.4	6.15
^{233}Pa	6.2	5.7	6.3	5.8

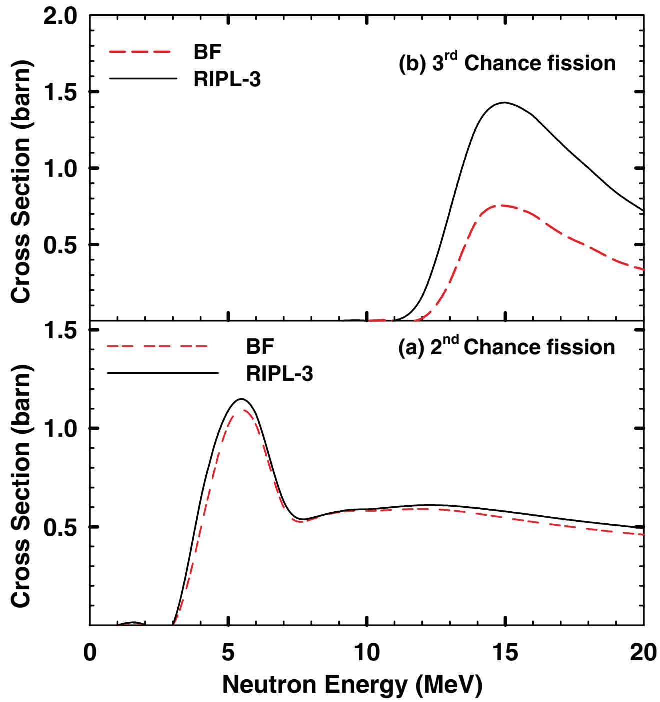

FIG. 3. (Color online) EMPIRE-3.1 predictions of fission excitation functions for BF (dashed line) and RIPL–3 (solid line): (a) second chance fission cross sections and (b) third chance fission cross sections.

[14], where the BF predicted inner barrier height 5.9 MeV corresponding to ^{233}Pa has been changed to 6.2 MeV, for the best fit of the ^{233}Pa(n,f) experimental data. The values of fission barriers for the ^{234}Pa and ^{235}Pa isotope are obtained from BF. The RIPL-3 does not have the fission barriers corresponding to ^{235}Pa isotope. Hence in the EMPIRE-3.1, the ^{235}Pa fission barrier values are kept the same for both BF and RIPL-3 calculations. The Fig. 2 also shows the calculated fission excitation function for ^{234}Pa(n, f) by EMPIRE-3.1 code for fission barriers corresponds to RIPL-3 and BF. The present experimental data agree very well with the calculated neutron induced fission cross sections for the fission barrier heights obtained from BF. The predictions of EMPIRE-3.1 for the RIPL-3 fission barriers are only in agreement with the data for the neutron energies below 12 MeV. For the neutron bombarding energies above 12 MeV, the EMPIRE-3.1 calculations for RIPL-3 overpredict the experimental data. Figure 3 shows the EMPIRE-3.1 predictions on fission excitation functions of second chance and third chance fission for BF and RIPL–3, contributing to the total ^{234}Pa(n, f) cross sections. The major difference has been observed in predictions of EMPIRE-3.1 for third chance fission excitation function between BF and RIPL-3 fission barrier parameters. The observed difference is consistent with the difference between the BF and RIPL-3 fission barrier values of the ^{233}Pa isotope as shown in Table II.

IV. EFFECT OF PREEQUILIBRIUM PARTICLE EMISSION ON NEUTRON INDUCED REACTION CROSS SECTIONS

The preequilibrium particle emission is likely to play some role in the equivalent incident neutron energy range

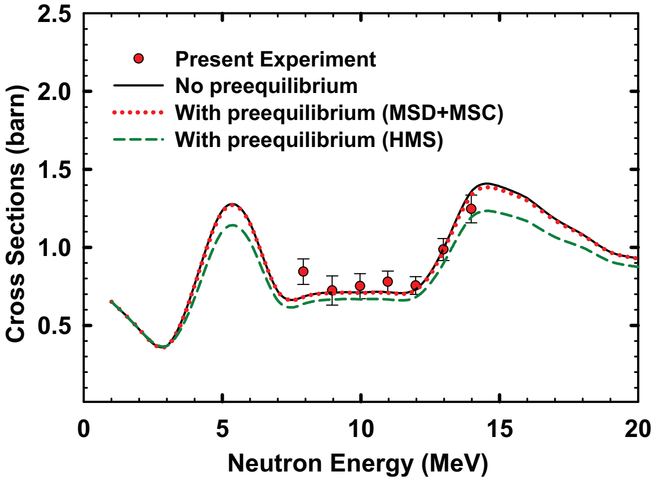

FIG. 4. (Color online) The EMPIRE-3.1 code predicted ^{234}Pa(n, f) cross sections with the pre-equilibrium contribution modeled using multistep direct + multistep compound (MSD+MSC) models (dotted line) and hybrid Monte Carlo simulation (HMS) model (dashed line). For reference purpose, the present experimental ^{234}Pa(n, f) cross sections (solid circles) and the EMPIRE-3.1 calculations with no pre-equilibrium effect (solid line) are also shown.

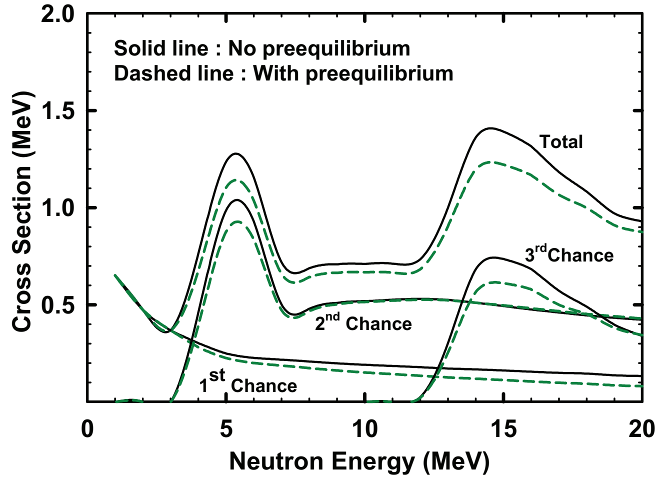

FIG. 5. (Color online) The EMPIRE-3.1 code predicted ^{234}Pa(n, f) cross sections with the pre-equilibrium contribution modeled using the hybrid Monte Carlo simulation (HMS) model (dashed line). For reference purpose, the EMPIRE-3.1 calculations with no pre-equilibrium effect (solid line) are also shown. Labels indicate the contributions from first-, second- and third- chance fission as well as the total.

for which ^{234}Pa(n, f) cross sections are measured in the present work. The extent of this effect on ^{234}Pa(n, f) cross sections has been investigated by invoking the preequilibrium models in the EMPIRE-3.1 calculations for fission barrier heights corresponding BF. The current version of EMPIRE-3.1 has four modules for the consideration of pre-equilibrium decay [17,30]; (i) exciton model (PCROSS), (ii) multistep direct (MSD), (iii) multistep compound (MSC), and (iv) hybrid Monte Carlo simulations (HMS). The EMPIRE-3.1 calculations have been carried out by invoking PCROSS, MSD+MSC, and HMS pre-equilibrium models. The effects of pre-equilibrium decay, modeled using PCROSS, do not show any effect on ^{234}Pa(n, f) cross sections in the measured equivalent neutron energy range. This complication is introduced by the using ECIS06 code in the EMPIRE-3.1 calculations, which calculates coupled-channel contributions to the collective discrete levels in the incident channel. In general, these contributions are so strong that adding those provided by the exciton model leave the results practically unchanged [30]. The pre-equilibrium effects on ^{234}Pa(n, f) cross sections calculated by invoking the MSD+MSC modules in the EMPIRE-3.1 calculations are shown in Fig. 4, which are not significant in the measured equivalent neutron energy range. The contribution of the pre-equilibrium processes modeled by invoking the HMS model is also shown in Fig. 4. The ^{234}Pa(n, f) calculated by invoking the HMS model differ by 8%–15% from the results without HMS in the considered neutron energy range. The contribution due to HMS is a maximum near the thresholds for second and third chance fission. Figure 5 shows the contributions from first-, second-, and third-chance fission as well as the total ^{234}Pa(n, f) cross sections with pre-equilibrium decay calculated using HMS (dashed line), for reference purpose, the similar calculations without preequilibrium decay invoked (solid line) are also shown.

V. SUMMARY

The transfer induced fission reactions ^{232}Th(^{7}Li, αf)^{235}Pa and ^{232}Th(^{7}Li, tf)^{236}U have been used as surrogate reactions of the ^{234}Pa(n, f) and ^{235}U(n, f) reactions. The compound nuclei ^{235}Pa and ^{236}U are populated at overlapping excitation energies, and their fission probabilities are measured in the excitation energy range of 14–20 MeV in steps of 1.0 MeV. The HSRM approach is then used to determine the ^{234}Pa(n, f) cross sections in the equivalent neutron energy range of 8–14 MeV, taking the ^{235}U(n, f) cross section values as reference. The experimental results on ^{234}Pa(n, f) cross sections have been compared with the calculated neutron induced fission cross sections using the EMPIRE-3.1 code for the barrier heights obtained from BF as well as RIPL-3. The EMPIRE-3.1 calculations are in good agreement with the present experimental data for the BF predicted fission barrier values. Whereas calculations performed using RIPL-3 fission barriers are consistent with the present experimental data only for the neutron energies below 12 MeV. The effect of pre-equilibrium neutron emission on the neutron-induced fission cross section of ^{234}Pa(n, f) has been investigated by invoking various pre-equilibrium models such as MSD, MSC, and HMS in the EMPIRE-3.1 calculations.

ACKNOWLEDGMENTS

The authors thank the operating staff of the BARC-TIFR Pelletron Accelerator facility, for smooth operation of the accelerator during the experiment.

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