

# A re-calculation of criticality property of $^{231}\text{Pa}$ using new nuclear data

S. Ganesan\*<sup>†</sup>, Umasankari Kannan\*\*\*, P. D. Krishnani<sup>†</sup>, V. Jagannathan<sup>†</sup>,  
R. P. Jain<sup>†</sup> and R. Karthikeyan<sup>†</sup>

<sup>†</sup>Reactor Design and Nuclear Data Section, Theoretical Physics Division, Central Complex and

\*\*Reactor Physics and Design Section, Engineering Hall No. 7, Bhabha Atomic Research Centre, Trombay, Mumbai 400 085, India

An interesting paper published by an earlier Bhabha Atomic Research Centre (BARC) team in *Current Science*<sup>1</sup> and elsewhere<sup>2-7</sup> dealt with data of criticality of minor actinide nuclides and their relevance to the long-lived-fission-waste problem. The purpose of the present article is to communicate the changes in the results of criticality calculation of  $^{231}\text{Pa}$  due to the use of more recent and improved neutron-nuclear interaction cross-section data. The new results of criticality for  $^{231}\text{Pa}$  based on improved basic data differ significantly from those published earlier by the BARC team.

installed capacity<sup>7</sup>. The isotope  $^{231}\text{Pa}$  has a long half-life of  $32760 \pm 110$  years for alpha activity. It is produced primarily by fast neutrons through the  $(n, 2n)$  reaction in  $^{232}\text{Th}$  followed by beta decay. It constitutes a considerable source of radio-toxicity. In advanced concepts such as the Energy Amplifier (EA) proposed by Carlo Rubbia *et al.*<sup>9</sup>, production of  $^{231}\text{Pa}$  takes place in significant quantities and a net stockpile of the order of 5 kg of  $^{231}\text{Pa}$  will persist during the whole life time of an EA plant. Criticality experiment has not been conducted with pure individual actinide isotope such as  $^{231}\text{Pa}$  thus far as earlier noted by Clayton<sup>10</sup>.

## Comments on the earlier BARC calculations of criticality

The earlier study<sup>1</sup> employed the multigroup transport theory approach using the DTF-IV code<sup>11</sup> to solve the one-dimensional Boltzmann neutron transport equation using the discrete ordinates method to give neutron density as a function of position, angle and energy inside a given medium. This code was used to compute the infinite medium multiplication factors and bare critical masses for spherical fast systems of actinides, using a 35 group cross-section set<sup>12</sup>. Further, it may be noted from Srinivasan *et al.*<sup>7</sup> that for  $^{231}\text{Pa}$  a special BARC-assembled nuclear data file was used. Srinivasan *et al.*<sup>7</sup> stated that the cross-section set for  $^{231}\text{Pa}$  was derived by combining BARC-evaluated data in the 1 to 20 MeV energy region with the  $^{233}\text{Pa}$  data of the JENDL-2 file below 1 MeV. Srinivasan *et al.*<sup>7</sup> also stated that the nuclear data of  $^{231}\text{Pa}$  above 1 MeV was based<sup>13</sup> on BARC calculations of theoretical nuclear models and a complete file for  $^{231}\text{Pa}$  was assembled artificially by taking the data of  $^{233}\text{Pa}$  from JENDL-2 below 1 MeV. In our opinion, this approach of constructing a complete data file by merging data of one isotope above 1 MeV with that of another isotope below 1 MeV is fundamentally unacceptable. The neutron-nuclear interaction cross-sections change drastically as we go from one isotope to another of the same element. The discussions by the BARC team indicate that the earlier BARC study<sup>7</sup> had an awareness that the calculated critical mass for  $^{231}\text{Pa}$  could be very uncertain due to uncertain

AN interesting study of criticality properties of minor actinide nuclides and their relevance to the long-lived-fission-waste problem has been published in the period 1989-1991 in several journals and conferences<sup>1-7</sup> by Bhabha Atomic Research Centre (BARC). In this paper, we re-assess the value of critical radius due to the use of improved input nuclear data of  $^{231}\text{Pa}$ . It may be noted that in the case of  $^{231}\text{Pa}$ , a value of 162.31 kg corresponding to a calculated critical radius of 13.61 cm (density =  $15.37 \text{ g/cm}^3$ ) has been presented in earlier reports<sup>1-7</sup>. Our finding which is drastically different from the earlier BARC result is that with the use of the new Japanese nuclear data<sup>8</sup> even the infinite medium of  $^{231}\text{Pa}$  is sub-critical.

Using improved nuclear data and methods in simulation of nuclear systems is an important aspect of any serious nuclear programme. The generation and use of accurate nuclear data are considered fundamentally important, as accurate nuclear data are essential inputs to simulate nuclear interactions to obtain engineering parameters. New concepts can be studied with greater confidence if the scientific basis is sound. In the case of minor actinides, such as  $^{231}\text{Pa}$  studied in this paper, the nuclear data status is very uncertain compared to the major isotopes of the uranium fuel cycle. The isotope  $^{231}\text{Pa}$  occurs in nature due to decay of  $^{235}\text{U}$  actinide ( $4n + 3$ ) series with a natural total world inventory of about 120 g. In thorium-fuelled reactors, production of  $^{231}\text{Pa}$  takes place at several hundred grams per year per GWe of

\*For correspondence. (e-mail: ganesan@magnum.barc.ernet.in)



data used at that time. The discussion<sup>7</sup> also indicates that the American National Standard Institute (ANSI)<sup>14</sup> critical mass value is 750 kg for <sup>231</sup>Pa, 'based on three group diffusion theory calculations by Wu and Ruby<sup>15</sup> and not so carefully evaluated data' (as quoted in Srinivasan *et al.*<sup>7</sup>) and the Trombay criticality formula-based value is 463 kg. We note further that the ANSI critical mass is presented for <sup>231</sup>Pa by the American Nuclear Society (ANS)-based value reported by Wu and Ruby<sup>15</sup>. In the ANSI report<sup>14</sup>, it is mentioned as '750 kg (indicated)' expressing some caution that the ANS value may need a re-assessment as Wu and Ruby<sup>15</sup> used a diffusion calculation and less experimental data were then available. The revision by BARC of the ANS result is not substantiated by our improved and rigorous investigations presented in this paper. Here, we are not questioning the use of DTF code or the way the code was used, but only the error due to the use of wrong nuclear data.

### New calculations of criticality of <sup>231</sup>Pa

We used the JENDL-3.2 basic evaluated data file<sup>8</sup> released in 1994 and available over the internet<sup>16</sup> from the Nuclear Data Section of the International Atomic Energy Agency (IAEA), Vienna. This database provides a comprehensive neutron transport cross-section library for all neutron-induced reactions in 10<sup>-5</sup> eV to 20 MeV, in ENDF/B-VI format<sup>17</sup>. The material identification number in the file is MAT = 9131. These computerized data files contain recommended values for use in application calculations and are coded in the ENDF/B format. The

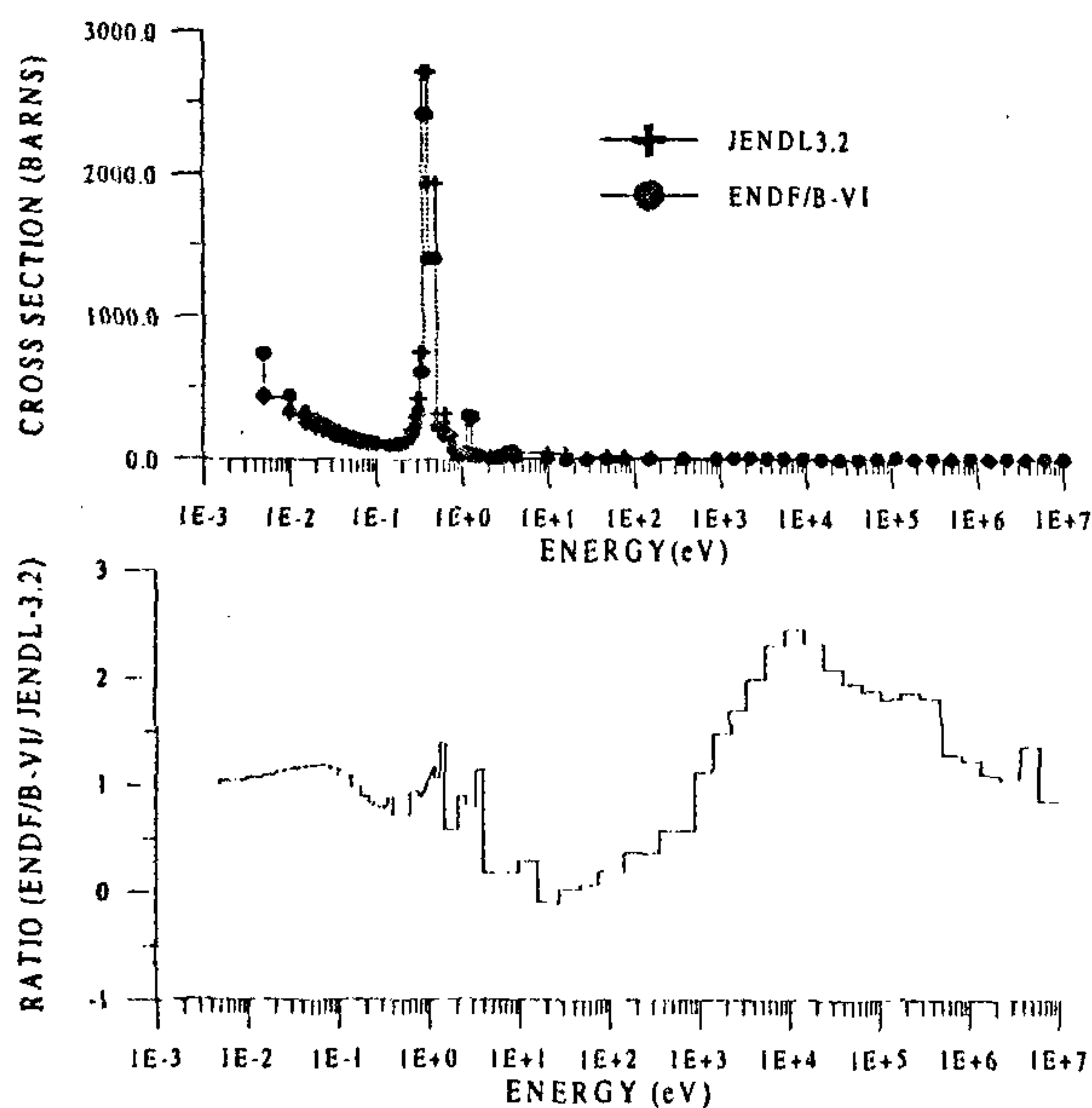


Figure 1. Comparison of neutron-induced absorption cross-sections in 69 energy groups for <sup>231</sup>Pa.

numerical values are obtained based on an evaluation of existing experimental data supplemented by theoretical model-based predictions and systematics<sup>8,18</sup> and thus represent the best data available in the world in electronic form for application calculations.

A graphical inter-comparison of all the neutron reaction data of JENDL-3 and ENDF/B-VI for all main isotopes of thorium fuel (<sup>230</sup>Th, <sup>232</sup>Th, <sup>231</sup>Pa, <sup>233</sup>Pa, <sup>232</sup>U, <sup>233</sup>U, and <sup>234</sup>U) are available in the handbook by Ganesan and MacLaughlin<sup>19</sup>. Generally, for these isotopes, the Japanese data, JENDL-3.2 is considered superior because the evaluations of nuclear data for these isotopes were funded well in Japan and significant efforts were made to incorporate the current status of theoretical predictions and available experiments in the creation of JENDL-3.2. On the other hand, as mentioned in the comments section of the electronic file of ENDF/B-VI for these isotopes, the USA database in ENDF/B-VI for these isotopes is a carry over from their earlier ENDF/B-V created around 1981.

Our calculations were performed as follows: A multi-group cross-section set in 69 groups in the WIMS format<sup>20</sup> was generated in collaboration with the IAEA Nuclear Data Section using the NJOY97 code system<sup>21</sup>. The multi-group cross-section set was generated following our specifications by McLaughlin from the IAEA Nuclear Data Section, using NJOY97. This task was successfully carried out within the scope of the IAEA Coordinated Research Programme (CRP) entitled, 'Final Stage of the WIMS Library Update Project'. The interested reader can get more details on this IAEA-CRP at

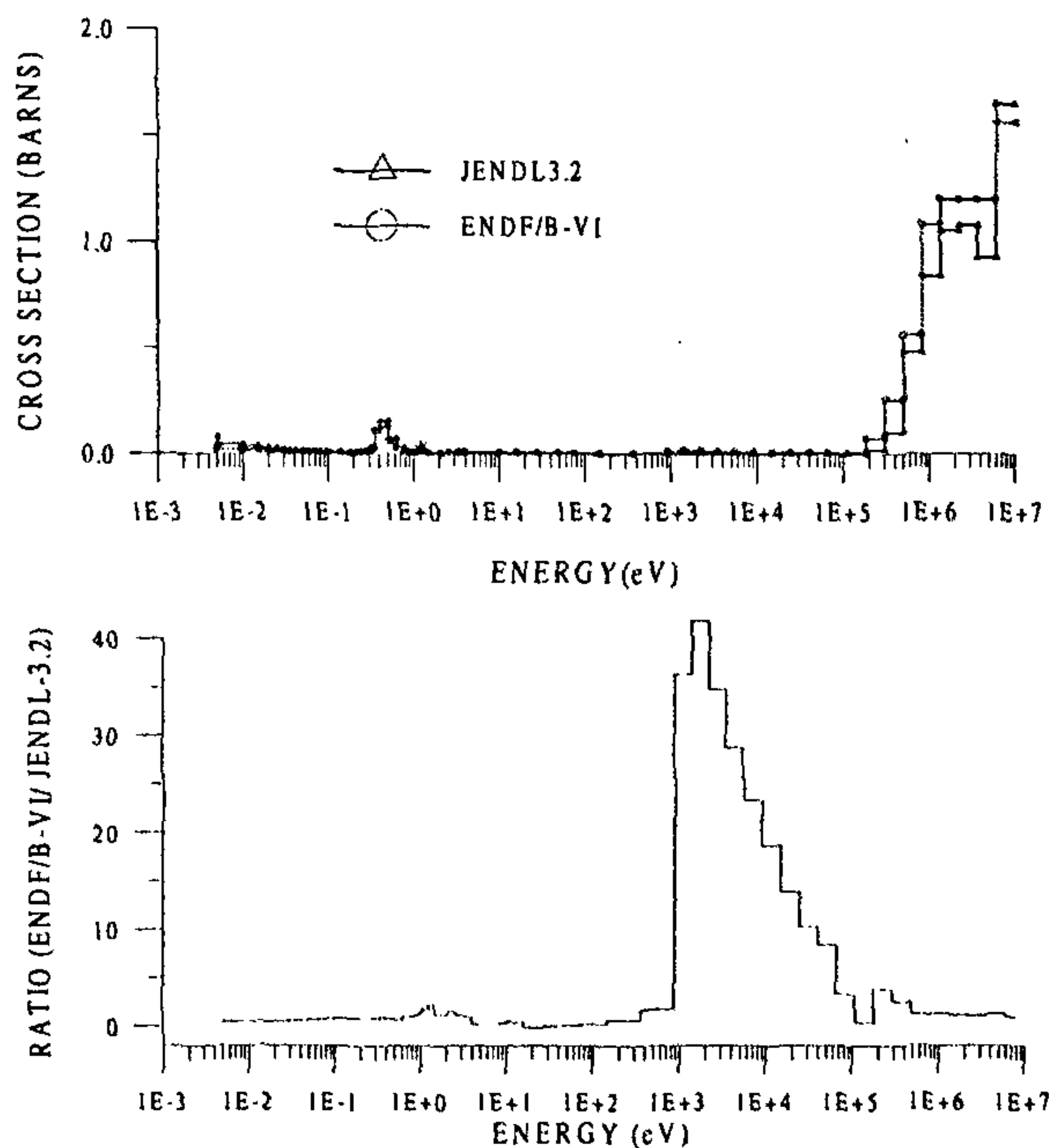


Figure 2. Comparison of neutron-induced fission cross-sections in 69 energy groups for <sup>231</sup>Pa.

the web-site <http://www-rcp.ijs.si/~wlup>. The generation of complete WIMS library for  $^{231}\text{Pa}$  involves several steps, viz. resonance reconstruction, Doppler broadening of neutron cross-section curves for all reaction channels, calculation of self-shielding factors in the resolved and unresolved resonance region for various dilutions, generation of self-shielded multigroup cross-sections, and finally, multigroup transfer matrices for elastic and inelastic cross-sections. These multigroup data are in the WIMS format in the 69 groups covering the energy region  $10^{-5}$  eV to 10 MeV. These state-of-art calculations were all performed strictly in accordance with the internationally established ENDF/B conventions and procedures for  $^{231}\text{Pa}$  for the first time.

The detailed intermediate outputs and the complete 69 group cross-section set in WIMS format derived from JENDL-3.2 are available free of cost upon request from Ganesan. These calculations were also performed using the American evaluated nuclear data file ENDF/B-VI available from the IAEA Nuclear Data Section<sup>22</sup>. Figures 1–3 present a graphical comparison of the cross-sections in the two files in multigroup form for fission, absorption and 'nubar', the total number of neutrons released per fission for the infinite medium taking self-shielding into account. For clarity, the ratio has also been shown in the graphs separately.

The calculations of infinite medium multiplication factor ( $K_{\infty}$ ) were performed using three different computer codes ITRAN1D (ref. 23), DTF-IV (ref. 11) and NEWMURLI (refs 24, 25) using the 69 group cross-sections for  $^{231}\text{Pa}$ . The ITRAN1D is a multigroup integral

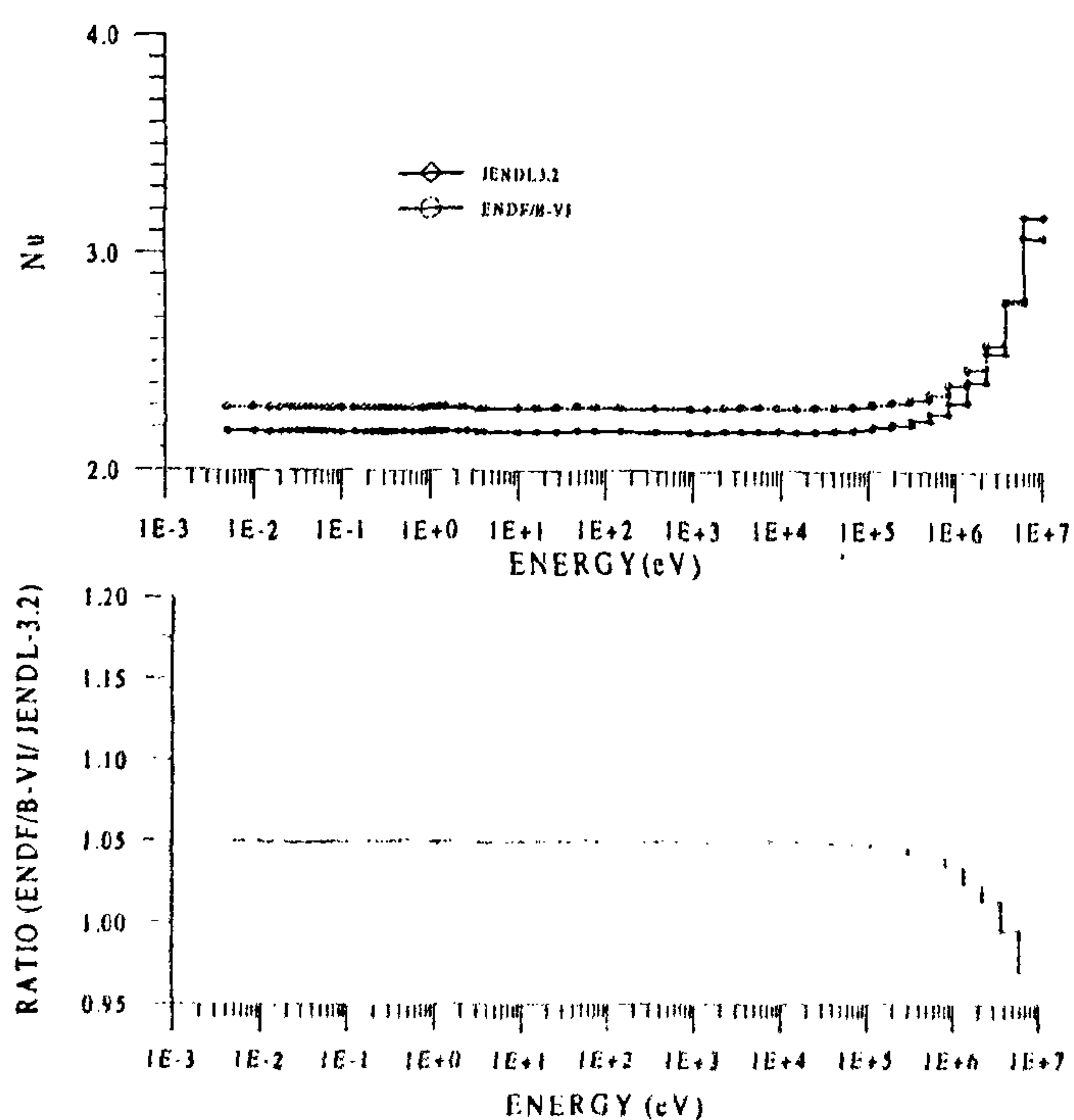


Figure 3. Comparison of effective total neutrons released per fission 'nubar' in 69 energy groups for  $^{231}\text{Pa}$ .

transport theory code based on interface current approach for three (sphere, slab, cylinder) geometries. The computer code DTF-IV is a multigroup integro-differential transport theory code based on  $S_N$  method. The code NEWMURLI is based on first flight collision probability method. The results were identical with all the above mentioned codes. We believe that improvements in processing approximations such as increasing the number of energy groups or Legendre order for the scattering matrices will not change the conclusions of this paper qualitatively.

Some additional discussions are presented in this section to provide further insight into our study. With the new 69 group cross-section data derived from JENDL-3.2, our calculated  $K_{\text{eff}}$  for a sphere of radius 13.61 cm is 0.6029 and the calculated infinite medium multiplication factor is 0.9729. These new results differ significantly from earlier results<sup>1-7</sup> in which the critical radius was 13.61 cm. We repeated, with the 69 group cross-section data derived from ENDF/B-VI, calculations of infinite medium multiplication factor using the ITRAN/DTF-IV/NEWMURLI codes. The effective one group cross-sections were also obtained by collapsing the 69 group effective cross-sections including self-shielding with the calculated fluxes obtained by transport calculations in the infinite medium of  $^{231}\text{Pa}$ . The effective nubar is obtained by collapsing with the product of fission cross-section and the flux. Table 1 compares the effective one group values for infinite medium of  $^{231}\text{Pa}$  and break-up into components.

The infinite medium multiplication factor,  $K_{\infty}$ , obtained in the earlier BARC studies<sup>1-7</sup>, is 2.199, which is significantly different from our present values of 0.9729 (JENDL-3.2) and 0.9410 (ENDF/B-VI) presented in Table 1. Note that in the JENDL-3.2 file, the value of fission spectrum average of eta is 1.88, which is much less than the value of eta (the infinite medium multiplication factor) in earlier studies of 2.199. Figure 4 compares the normalized neutron spectra in an infinite medium of  $^{231}\text{Pa}$  with that of virgin fission neutron spectrum. It should be further stressed, as seen in Figure 4, that the infinite medium neutron spectrum, due to moderation by mainly neutron inelastic scattering in  $^{231}\text{Pa}$ , is softer than the

Table 1. Values for infinite medium of  $^{231}\text{Pa}$

Quantity	Basic file	
	JENDL-3.2	ENDF/B-VI
Fission cross-section $\sigma_f$ (barns)	0.4270	0.5846
Capture $\sigma_c$ (barns)	0.6465	0.9724
$\sigma_{n,2n}$ (barns)	0.0018	0.0043
Absorption [capture + fission - ( $n, 2n$ )] $\sigma_a$ (barns)	1.072	1.553
Number of neutrons per fission $\nu$	2.442	2.499
Infinite medium multiplication factor $K_{\infty} = \nu\sigma_f/\sigma_a$	0.9729	0.9410



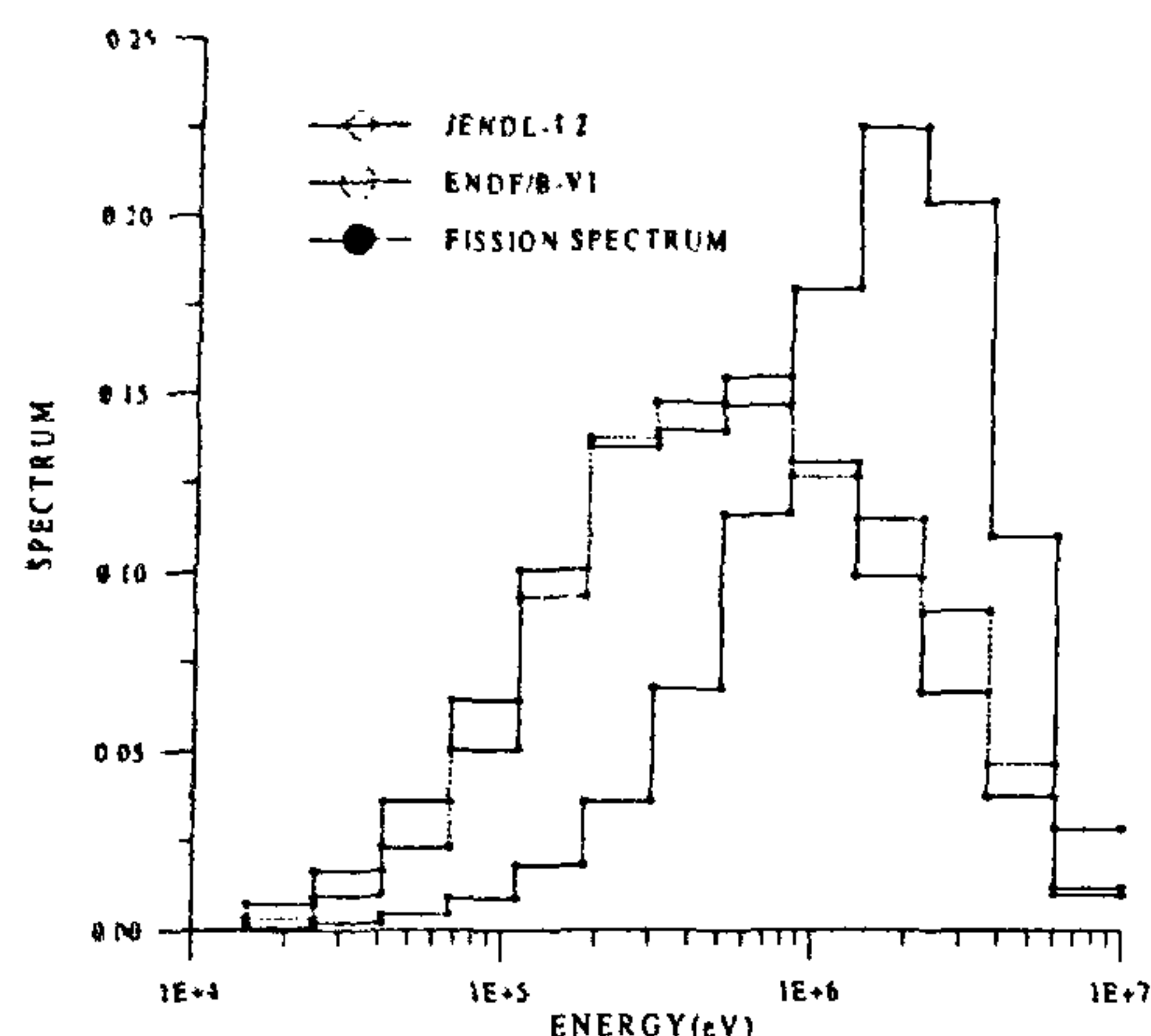


Figure 4. Comparison of normalized multigroup neutron spectra in infinite medium of  $^{231}\text{Pa}$  with normalized fission spectrum.

virgin fission neutron spectrum. The value of 2.199 for infinite medium multiplication factor obtained in earlier studies<sup>1-7</sup> indicates that the cross-sections used there for  $^{231}\text{Pa}$  are very different from the values given in JENDL-3.2 and ENDF/B-VI.

For a sphere with radius of 13.61 cm, we performed Monte Carlo calculations of  $K_{\text{eff}}$  employing the JENDL-3.2-based continuous energy data and using the MCNP code<sup>26</sup>. For consistency, the WIMS data derived by us from the same JENDL-3.2 was used in the multigroup transport theory calculations using the DTF code. The DTF calculations gave a  $K_{\text{eff}}$  of 0.60288 while the Monte Carlo calculations resulted in a value of  $0.60137 \pm 0.0034$ .  $K_{\infty}$  was also calculated with the JENDL-3.2 file using the MCNP code and was found to be  $0.9727 \pm 0.00114$ . The error quoted in the case of the MCNP calculations is the statistical uncertainty in the Monte Carlo simulation. These results which are summarized in Table 2 provide additional confidence in our calculations.

### Conclusions and recommendations

A summary of the results is presented in Tables 1 and 2. Unfortunately, the breakup of the earlier BARC result for  $K_{\infty}$  of 2.199, into components of fission, absorption, etc. or the detailed basic data actually used there are not available. The only basic data that is available from the previous study is the 'nubar' value given in Table 1 of Srinivasan *et al.*<sup>1</sup>. In that table, the total number of neutrons released per fission, nubar, is given as 2.598 corresponding to the neutron energy range 0.6 to 1.1 MeV (9th energy group of the 35 group set). The values in the basic evaluated data files JENDL-3.2 and ENDF/B-VI are seen by us to be 2.296 and 2.380 at the energy of 0.85 MeV. It may be noted that the evaluation of 'nubar' is based on Bois-Frehaut's semi-empirical formula<sup>27</sup> in JENDL-3.2 and on Howerton's semi-empirical formula<sup>28</sup>

Table 2. Summary of results for  $^{231}\text{Pa}$

Description	Earlier BARC study	Present BARC study based on JENDL-3.2
$K_{\text{eff}}$ of a sphere of radius of 13.61 cm	1.000 (using the DTF and 35 group data)	0.60288 (using the DTF and 69 group data)
$K_{\text{eff}}$ of a sphere of radius of 13.61 cm using Monte Carlo method (MCNP)	Not available	$0.6014 \pm 0.0034$
Infinite medium multiplication factor, $K_{\infty}$	2.199 (DTF)	0.9729 (DTF) $0.9727 \pm 0.00114$ (MCNP)

in ENDF/B-VI. It is not clear how the high value of 2.598 for nubar was arrived at in the earlier study.

We conclude that  $^{231}\text{Pa}$  does not become critical even for infinite mass. This conclusion is based upon rigorous calculations and analyses using improved data files JENDL-3.2 and ENDF/B-VI and improved processing of these files. These data files provide the state-of-the-art nuclear data based on more recent measurements and state-of-the-art theoretical calculations of cross-sections that are based on improved models and systematics.

In the earlier BARC study<sup>5</sup>, it is stated (within quotes): 'Although  $^{231}\text{Pa}$  produced in large quantities in  $\text{Th}/^{233}\text{U}$  reactors would be classified as a fissile nuclide on account of even number of neutrons, surprisingly, its  $K_{\infty}$  value is as high as 2.2 (higher than that of  $^{235}\text{U}$ ), indicating that  $^{231}\text{Pa}$  is a very good nuclear fuel.' We do not agree with this earlier conclusion in view of our results.

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## Molecular basis of antifungal toxin production by fluorescent *Pseudomonas* sp. strain EM 85 – A biological control agent

K. N. Anith<sup>\*†</sup>, K. V. B. R. Tilak<sup>\*\*</sup> and S. P. S. Khanuja<sup>‡</sup>

<sup>†</sup>Regional Agricultural Research Station, Ambalavayal, Wayanad 673 593, India

<sup>\*\*</sup>Division of Microbiology, Indian Agricultural Research Institute, Pusa, New Delhi 110 012, India

<sup>‡</sup>Biotechnology and Genetic Research Division, Central Institute of Medicinal and Aromatic Plants, Lucknow 226 015, India

A fluorescent *Pseudomonas* sp. (strain EM 85) was found to inhibit growth of many soil-borne plant pathogenic fungi and effectively control fungal diseases. The genetics governing this character was probed by mutagenesis followed by a functional complementation analysis after construction of a genomic library of the wild-type strain using the cosmid vector pLAFR 1. A cosmid clone, pANF 17 was able to complement the antifungal toxin production in a

defective mutant. The chromosomal origin of the DNA fragment in the cosmid clone was confirmed by a Southern blot. Sub-clones of the three *EcoRI* fragments from pANF 17, however, failed in the complementation test. The complementing cosmid was found to be stably maintained and expressed in the defective mutant under laboratory plate assay and *in vivo* conditions as evidenced by the extraction and detection of the toxin and biological control experiment.

RHIZOSPHERE-competent bacteria and fungi are the most preferred candidates as biological control agents. Soil pseudomonads are important among them as they colonize the underground growing plant organs efficiently and

survive in a variety of diverse conditions<sup>1</sup>. The biological activities by which these pseudomonads bring about disease control include rhizosphere colonization, antibiosis, iron chelation by siderophore production, production of volatile compounds, induction of systemic resistance and competition for nutrients<sup>2</sup>.

\*For correspondence.