Recent Work Leading Towards a New Evaluation of the Neutron Standards


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(Received 29 May 2014; revised received 18 September 2014; accepted 22 September 2014)

A new version of the ENDF/B library has been planned. The first step in producing this new library is evaluating the neutron standards. An evaluation is now underway with support from a Data Development Project of the IAEA. In addition to the neutron cross section standards, new evaluations are being done for prompt fission neutron spectra and a number of reference data. Efforts have been made to handle uncertainties in a proper way in these evaluations.

I. INTRODUCTION

Since most measurements of neutron cross sections are made relative to a standard, it is important to maintain an active measurement and evaluation program to improve those standards. In the next sections the changes that led to improved evaluations with more defendable uncertainties will be discussed. All uncertainties in this paper represent coverage factors corresponding to one standard deviation.

II. PREVIOUS CROSS SECTION STANDARDS EVALUATIONS

For ENDF/B-I there really were no specific evaluations identified as community standards. The first use of standards was in ENDF/B-II. The ENDF/B-III efforts led to laboratories/individuals taking on the responsibility for specific standards evaluations for which they had expertise and interest. Uncertainties did not play a large part in this version.

With ENDF/B-IV more objective evaluation techniques for the standards came about largely focused on the light-element standards with the use of R–matrix analyses. For the heavy-element standards, older evaluation methods were used - basically drawing a curve on a graph of acceptable data. Such evaluations are difficult to document and it is not clear how to determine meaningful uncertainties and covariance information.

First efforts to use an objective evaluation method for the heavy element standards occurred with ENDF/B-V [1] when Poenitz did an evaluation of the $^{235}\text{U}(n,f)$ cross section.

For the ENDF/B-VI evaluation [2] of the standards, considerable effort was devoted to improved evaluation procedures. In previous evaluations a hierarchical approach was followed. This approach does not include absolute and ratio data on the same basis as they were measured. For example, a ratio of the $^{10}\text{B}(n,\alpha)$ to the $^{6}\text{Li}(n,t)$ cross sections would be used in the $^{10}\text{B}(n,\alpha)$ cross section evaluation but not in the $^{6}\text{Li}(n,t)$ cross section evaluation. The difficulties with that procedure led to a combining approach. The combining procedure was achieved by using a simultaneous evaluation using generalized least-squares with separate R–matrix analyses.
Least-squares methods allowed the combining of input data consistent with the experimental uncertainties. A database was established that could handle the full information content available. Thus data were evaluated simultaneously to assure proper use of the available information. Ratio measurements were properly handled so there would be an impact on each of the cross sections in the ratio. Correlations among the experimental data both within an experiment and with other experiments were taken into account in the simultaneous evaluation. The R–matrix fits for the evaluation of the light element standards allowed a large class of data in addition to angle integrated neutron cross sections to be used in these analyses. An important condition was that there cannot be any correlations between the database used for the simultaneous evaluation and the database used for the R–matrix evaluations. This procedure led to a consistent evaluation in which correlations and ratio measurements were properly taken into account. To satisfy the correlation condition, the boron and lithium experimental data were separated into two uncorrelated groups, one for use in the R–matrix analyses and the other for use in the simultaneous analysis. All the standards except the H(n,n), \( ^3\)He(n,p) and C(n,n) cross sections were evaluated using the simultaneous evaluation and R–matrix analyses. Separate R–matrix evaluations were performed for the H(n,n), C(n,n) and \( ^3\)He(n,p) cross sections. Total cross section and scattering measurements for \( ^6\)Li and \( ^10\)B were contained in the database since they put constraints on the reaction data. Measurements of \( ^{235}\)U and \( ^{239}\)Pu fission cross sections in a \( ^{252}\) Cf spontaneous fission neutron spectrum in addition to \( ^{238}\)U(n,\( \gamma\)) and \( ^{239}\)Pu(n,f) data were included since they improved the quality of the standards. Another subset which was used as input to the simultaneous evaluation was an evaluation of the thermal data for \( ^{233}\)U, \( ^{235}\)U, \( ^{239}\)Pu and \( ^{241}\)Pu by Axton with the associated variance-covariance data.

The R–matrix analyses for the light–element standards were done by Hale using the code EDA [3]. The simultaneous evaluation was done with the program GMA [4] written by Poenitz. A separate code written by Peelle was used to combine the simultaneous evaluation and R–matrix analyses and produce the final cross sections and uncertainties. All experiments which are correlated and all ratio measurements (except those to the hydrogen standard) were put into the simultaneous evaluation data subset. In the R–matrix analyses, the experimental data were weighted based on the quoted random uncertainties and it was assumed that no correlations other than the overall normalization were present among the data from a particular experiment.

It was found that very unusual mean values and reduced uncertainties can be obtained with discrepant correlated data. This was the first observation of the Peelle Pertinent Puzzle (PPP) effect [5]. A method was established to minimize problems associated with discrepancies by down weighting discrepant data. It had the effect of reducing \( \chi^2 \) per degree of freedom to essentially 1. In some cases very small uncertainties in the combined output of the evaluation were found even with this down weighting and increasing of the R–matrix uncertainties by a factor of the square root of \( \chi^2 \) per degree of freedom.

The \( ^{252}\) Cf spontaneous fission neutron spectrum is used as a standard for fluence determination. An independent generalized least-squares evaluation of that spectrum by Mannhart [6] was used as the standard for ENDF/B-VI. The evaluation includes the spectrum and its covariances.

### III. AN INTERNATIONAL EVALUATION OF THE CROSS SECTION STANDARDS (ENDF/B-VII)

This is the first standards evaluation [7] that was done internationally so that full use of world wide capabilities could be available for the evaluation. The evaluation was a cooperative effort of the CSEWG, the WPEC and the IAEA. The work involved updating the previous work by including new measurements and improving the evaluation process. Before the evaluation process was started a number of tasks were initiated.

**A. Handling of Discrepant Data**

To reduce the effect of discrepant data, deviations of experimental neutron measurements from the output of the evaluation were compared with the uncertainties on the data. The outliers were defined as those with a difference from the evaluated value above two standard deviations for a single point or above one standard deviation for a few sequential points. The uncertainty of outliers was increased by adding an additional component to the covariance matrix of the uncertainty of each outlying data set. The length of correlation for this additional medium energy range correlation component was estimated from an analysis of the energy dependence of the discrepancy. This resulted in a much better \( \chi^2 \) per degree of freedom and larger uncertainty in the evaluated results. The change in the evaluated cross section was small.

**B. Code Comparisons**

There was an extensive effort comparing evaluation codes, both R–matrix and model-independent, to ensure that the results obtained were not code dependent. The average uncertainty of the evaluated values due to different procedures used is about 0.2% - 0.3% and this difference was added to the uncertainty assigned in the final evaluation.

The code intercomparison led to an investigation on how to minimize the PPP effect. Several different methods were explored.

The average difference between results obtained from the various codes and options used to minimize PPP is
about 0.3% (see Fig. 1). The results labeled "GMA" were obtained with GMA using different computational steps. The Box-Cox [8], GLUCS03 [9] and SOK [10] results are also shown. The Chiba-Smith method [11] for minimizing PPP effects was incorporated as an added uncertainty into the final estimate of the uncertainties of the evaluated fission cross sections at all energy points (excluding the thermal energy). This exclusion of an additional uncertainty for the thermal energy points is appropriate because the thermal cross sections are rather strongly decoupled from all other experimental data in the GMA database. This additional uncertainty was also not applied to the capture cross sections since doing so would have introduced only a very small change in the capture cross section covariance matrices owing to the fact that their evaluated uncertainties were already quite large in comparison to 0.3%.

C. Evaluation Procedure

An evaluation procedure similar to that used for the ENDF/B-VI standards evaluation was used to obtain the standards. The simultaneous evaluation was done using the generalized least-squares code GMAP. The combination code used in the previous evaluation was not used. All the standards except the H(n,n), 3He(n,p) and C(n,n) cross sections were evaluated using the GMAP code. This code provides the combining procedure by using input from two separate R–matrix analyses, RAC [12] and EDA, and a thermal constants evaluation in addition to the direct data sets normally used. The R–matrix input and thermal constants data were treated as the additions of other data sets to the GMAP code. For this evaluation, the only lithium and boron data for direct use in the GMAP code were the ratio measurements and correlated data. For the 6Li(n,t), 10B(n,α) and 10B(n,α,γ) R–matrix work, the evaluated cross sections obtained from the RAC and EDA analyses were not identical. In Fig. 2, differences that increase with increasing neutron energy are apparent. The differences between the two fits are a result of uncertainty in evaluation method. Thus for the lithium and boron standards, the cross sections from the RAC and EDA analyses were averaged (un-weighted) and used as the R–matrix input to GMAP. The covariance matrix used with these central values was that from the RAC code. At each energy point, half the difference between the RAC and EDA fits was treated as a method uncertainty and was added as an additional uncorrelated component of the total uncertainty for the RAC covariance matrix used in the R–matrix evaluation. This then takes into account the differences obtained between the RAC and EDA analyses. Such differences limit the highest energies that these standards can be used.

D. Covariance Data

The data from this international standards evaluation were adopted for use in the ENDF/B-VII evaluation, but, only cross-energy correlations between data points for a given standard reaction were used. Due to the evaluation procedure used for the standards evaluation, cross-material and cross-reaction correlations were also obtained and they are available. Clearly the preferred option should be to use the most complete covariances that are available. Cross-material and cross-reaction correlations can be very large in some cases. If those correlations are not taken into account in practical calculations of nuclear systems involving those cross sections, the covariance weighting will lead to incorrect re-
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sults and incorrect uncertainties. For example, in Fig. 3, correlations between the $^{238}\text{U}(n,f)$ and $^{235}\text{U}(n,f)$ reactions are shown. The correlations are quite large in the region near the diagonal. Such large correlations are present since many $^{238}\text{U}(n,f)$ cross section measurements were made relative to the $^{235}\text{U}(n,f)$ cross section for the data used in this evaluation. Using complete covariance information is more complicated to implement, but it is necessary for correct propagation of uncertainties in simulations.

![238\text{U}(n,f) - 235\text{U}(n,f) Correlation Matrix](image)

**FIG. 3.** (Color online) Cross material correlation matrix for the $^{238}\text{U}(n,f)$ and $^{235}\text{U}(n,f)$ reactions.

E. Concern About Small Uncertainties

Again, as for the ENDF/B-VI standards evaluation, some concern was expressed by the nuclear community over the small uncertainties that were obtained even though they were generally somewhat larger than those obtained in the ENDF/B-VI evaluation. It must be noted that a very large database was used for these evaluations. This would suggest that small uncertainties would be obtained unless important correlations and unknown systematic uncertainties were not taken into account. Also, for the light element standards the R–Matrix evaluations produce small uncertainties over an extended energy range due to strong model correlations leading to large off-diagonal covariances. Concerns about uncertainties in the R–Matrix model may need further study. A study was made of the process used for obtaining the evaluations. Factors that might lead to low uncertainties include the following:

1. **Underestimation of Correlations**

Use of common samples and detectors for different measurements can lead to 100% correlations for these components of the uncertainties in the results of different measurements. These correlations were carefully analyzed by Poenitz [4] when data were entered (and updated) in the GMA database. Sets of experimental data obtained by the same group or at the same facility are usually combined in data blocks that account for the correlations between data sets.

2. **Unrecognized Systematic Uncertainties**

The discrepancies among measurements indicate there must be unknown systematic uncertainties in some of the measurements in the standards database. Outlying data relative to expected values can be identified as possessing unrecognized systematic uncertainties. In the present evaluation an additional component of uncertainty is assigned to these discrepant experimental data (as was described previously) and this will increase the uncertainties of the evaluated data. It is expected that this process will compensate for the presence of these discrepant data. However, introducing an additional component of uncertainty to these data only increased the overall uncertainty of the evaluated data by a small amount. The small uncertainty paradox with evaluated data takes place when the absolute covariance of the experimental data between two energy points is larger than the smallest variance in those points. Introducing the restriction, $\text{abs}(v_{ik}) \leq \text{min}(v_{ii}, v_{ik})$, of the elements of the covariance matrix allows this paradox to be avoided.

3. **Uncertainties for Correlated Data Cannot be Characterized only by Percentage Uncertainties or Variances**

Different model and non-model fits can be used in the evaluation of given experimental data, and even if the cross sections evaluated in these different fits are close, the covariance matrices for the evaluated uncertainties can be very different. Thus it is essential to consider the covariances, not just the variances, in applications of cross sections to practical systems. The use of models in the evaluation of the standards leads to a redistribution of the uncertainties between variances and off-diagonal covariances of the uncertainty matrix with a reduction of the variances. As a result, the standard deviations (the square roots of the variances) are reduced but the uncer-
tainty of integral quantities dependent on the evaluated data, if covariances are used in their calculation in a wide energy region, should be conserved in general.

This led to the conclusion that based on the assumptions used in the evaluations, the uncertainties appear to be reasonable.

4. The $^{252}$Cf Spontaneous Fission Neutron Spectrum

The $^{252}$Cf spontaneous fission neutron spectrum results from ENDF/B-VI were carried over since very few new experiments have been made.

5. Smoothing

The lithium and boron cross sections obtained from this evaluation process are smooth since they were dominated by the R-matrix data. However, in some cases the results obtained for the heavy-element standards showed fluctuations which seemed unreasonable based on expectations from the theory of average cross sections. It was determined that a simple three-point smoothing procedure would be satisfactory for most cases. However, the uncertainties were not smoothed.

The standards and their energy ranges are shown in Table I.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Standards Energy Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^1$H(n,n)</td>
<td>1 keV to 20 MeV</td>
</tr>
<tr>
<td>$^3$He(n,p)</td>
<td>0.0253 eV to 50 keV</td>
</tr>
<tr>
<td>$^6$Li(n,t)</td>
<td>0.0253 eV to 1 MeV</td>
</tr>
<tr>
<td>$^{10}$B(n,α)</td>
<td>0.0253 eV to 1 MeV</td>
</tr>
<tr>
<td>$^{10}$B(n,γ)</td>
<td>0.0253 eV to 1 MeV</td>
</tr>
<tr>
<td>C(n,n)</td>
<td>10 eV to 1.8 MeV</td>
</tr>
<tr>
<td>Au(n,γ)</td>
<td>0.0253 eV, 0.2 MeV to 2.5 MeV</td>
</tr>
<tr>
<td>$^{235}$U(n,f)</td>
<td>0.0253 eV, 0.15 MeV to 200 MeV</td>
</tr>
<tr>
<td>$^{238}$U(n,f)</td>
<td>2 MeV to 200 MeV</td>
</tr>
</tbody>
</table>

IV. FUTURE WORK — A NEW INTERNATIONAL EVALUATION OF THE STANDARDS

Updated standards evaluations for national cross section libraries (e.g. ENDF/B, JENDL, CENDL and JEFF) are needed as new versions of those libraries are anticipated. The research effort has been broadened to include some activities not normally considered related to the standards. Some of the topics are briefly discussed in the sections below. Additional information can be found in reports [13–15].

A. The Neutron Cross Section Standards

Measurements have been made related to all the cross sections since the completion of the last standards evaluation. See [16] for information on those measurements. A new evaluation is now underway.

1. Evaluation Method

The evaluation process is expected to be similar to that used for the previous standards evaluation. GMAP will be used for the simultaneous evaluation. At this time it is not clear how many R–matrix evaluation codes will be involved. The EDA R–matrix code of Hale is expected to be used. There is also an R–matrix code AMUR [17] by Kunieda et al. that may be used in the evaluation effort if it is updated to include polarization data. The R–matrix code FDRR [18] by Tao may be used if several actions are satisfied including use of systematic and statistical uncertainties, and production of the covariance matrix of evaluated cross sections as output.

2. Smoothing

Smoothing will be done with either the simple three-point smoothing, referred to previously, or smoothing using the shapes predicted by physical model calculations. For the previous standards evaluation there was one case where a model was used to provide smoothing. In that case a rather large $^{235}$U(n,f) cross section fluctuation, assumed statistical, occurred in the 50-60 MeV energy region. A patch using the shape of the Maslov [19] evaluated curve was used. A large uncertainty in the long energy range correlation component (normalization) and a small uncertainty in the medium energy range correlation component (shape) were used. For the new evaluation it is anticipated that models will be used. Smoothing should reduce the uncertainties obtained from a basic experimental data evaluation. It is expected that all changes in the matrix introduced by smoothing will occur for elements near the diagonal of the matrix and their sum will be changed little. In Fig. 4, the preliminary results of such smoothing by Pronyaev [20] for the Au(n,γ) cross section are shown.

Such smoothing keeps the kinks in the cross section shape predicted by the model near the thresholds of inelastic scattering levels and causes little change in the uncertainties in the regions where the uncertainties are smooth.
performed recently by Danon et al. [21], but final results have not yet been released. A new code is being developed at LANL to better address the many sources of PFNS uncertainties and correlations, and plans exist to apply it to a re-evaluation of the $^{252}$Cf (sf) PFNS standard.

There are two relatively new measurements of the PFNS of $^{235}$U(n,f) by Kornilov et al. [22] and Vorobyev et al. [23]. For both of these experiments, data were obtained with both $^{235}$U and $^{252}$Cf deposits during the measurements. The results from both experiments show a softer spectrum below 1 MeV compared with evaluations. At higher energies, above about 9 MeV, the Kornilov et al. data have a larger yield, whereas the Vorobyev et al. data tend to agree with ENDF/B-VII.0 values. Older data have also been obtained by Starostov, et al. [24].

A non-model evaluation of PFNS requires the availability of accurate data sets at very low (below 500 keV) and very high (above 8 MeV) outgoing neutron energies. Those two energy regions present distinct and very real challenges: at low-energy, multiple scattering corrections become the dominant and large uncertainty, while at high energies, statistics become scarce.

Model calculations help on both the low and high energy sides of the spectrum, but also introduce long-range correlations that impact the shape of the PFNS as well as the associated evaluated uncertainties. It was shown in work at LANL and the IAEA that strong model uncertainty correlations in combination with the normalization condition on the PFNS lead to surprisingly low uncertainties near the average outgoing energy of the PFNS and result, due to the strong model correlations, in unrealistically small uncertainties also in the wings of the PFNS. Work is ongoing at LANL and the IAEA to address this problem for PFNS evaluations.

An approach for a combined non-model evaluation of prompt fission neutron spectra using the GMA code has been proposed. In this approach, a generalized least-squares fit was made using experimental data for the PFNS of $^{235}$U(n$_{th}$,f), and the non-model and non-smoothed evaluation of the PFNS of $^{252}$Cf(sf) done by Mannhart. Most data for $^{235}$U(n$_{th}$,f) PFNS are obtained relative to $^{252}$Cf(sf) PFNS as a standard. Because of this, the evaluation of the $^{235}$U(n$_{th}$,f) PFNS is coupled in the combined fit with evaluated data for $^{252}$Cf(sf) PFNS. The changes in the $^{252}$Cf(sf) PFNS standard in the combined fit are small, because of its relatively small uncertainty. However an improved evaluation for $^{235}$U(n$_{th}$,f) PFNS is expected. In Fig. 5 preliminary results of Pronyaev are given. There is still discussion on the effect of smoothing on the covariances. There is also a separate Bayesian evaluation of the $^{235}$U(n$_{th}$,f) PFNS by Mannhart. These two projects are ongoing.

C. Prompt Fission Neutron Spectra Calculations

A Unified Monte Carlo approach is being developed to perform model evaluations based on estimation of the
model uncertainties used as a prior combined with a Bayesian least-squares fit of the experimental data. An attempt to utilize this approach for the uncertainty quantification of the PFNS was presented by Rising et al. [25]. Traditionally, evaluations of PFNS rely on some version of the Los Alamos Madland-Nix model [26]. Recently, Monte Carlo calculations following the evaporation of excited fission fragments by successive evaporations of neutrons and γ rays were performed by Talou et al. [27], Vogt et al. [28], Litaize et al. [29] and Schmidt [30]. Within this new approach, physical quantities beyond the average PFNS and neutron multiplicity can be studied. When provided with accurate primary fission fragment yields, this approach has been shown to compute the average neutron multiplicity and spectrum on a par with existing evaluations, while also predicting exclusive and average neutron multiplicity and spectrum. Those event-by-event models are more realistic than the Los Alamos model, but still face important challenges. They make several important assumptions regarding the mechanisms of emission, e.g., neutron emission from fully accelerated fragments only, and are sensitive to initial input parameters, e.g., fission fragment yields and nuclear structure, for which experimental data are lacking. Quantifying realistic uncertainties associated with this type of calculations also remains to be addressed.

In a parallel work, uncertainties associated with current evaluated PFNS that use Los Alamos model calculations have been quantified for 239Pu(nth,f) PFNS by Talou et al. [32], and are being extended to other PFNS of interest, such as 235U(nth,f) and 252Cf(sf).

The evaluation of the spectrum for 235U(nth,f) PFNS and the calculational efforts in this project will contribute to the Coordinated Research Project on evaluation of PFNS for a wide energy range of incident neutrons and for a number of nuclides.

D. Reference γ-Ray Production Cross Sections

Measurements of neutron-induced γ-ray production cross sections are most easily performed using a reference cross section in which a discrete γ-ray is detected. The need exists for accurate reference cross sections for such measurements. The preliminary conclusion is that the best candidates are natLiF and natTi. For natLiF, the 7Li(n,n') reaction with a gamma-ray of 478 keV has a yield that is isotropic; has little structure in the cross section for energies, 1 to 4 MeV; and the cross section is reasonably large. For the natTi(n,n') reaction, large yields are present for the 984 keV gamma from 48Ti(n,n'). This cross section may only be used above about 3 MeV due to the structure in the cross section. New gamma-production cross section data for Li and Ti with GEANIE at LANSCE by Nelson et al., and at JRC-IRMM by Plomp et al., are in progress or planned. For current experiments, it is estimated that absolute cross sections can be obtained with uncertainties in the 2 to 3 % range. Correlations exist due to the use of HPGe detectors and fission flux monitors in current experiments. Theoretical and GMA evaluation analyses performed on the basis of available experimental data have shown that the 984 keV γ-ray production cross section from the 48Ti(n,n') reaction can be estimated with an uncertainty of about 5 % from threshold up to 20 MeV [33]. In Fig. 6, the results of that evaluation are shown. The presence of structure limits this cross section, as a reference, to energies above 3 MeV. The limited experimental database is a challenge for this effort.
E. Measurements and Evaluations of the Low Energy $^{197}$Au(n,$\gamma$) Cross Section

![Diagram](image.png)

FIG. 7. (Color online) Comparison of several Au(n,$\gamma$) cross section measurements with the ENDF/B-VII standards evaluation. The plot includes the energy region for a Maxwellian of 25 keV.

The Maxwellian averaged cross section for $^{197}$Au(n,$\gamma$) for a temperature of 25 keV (MACS) is used in neutron capture cross-section measurements as a reference for reactions important for astrophysics and reactor applications. This reference cross section was obtained from an evaluation based on the results of measurements by Ratynski and Käppeler [35] of the $^{197}$Au(n,$\gamma$) cross section averaged over a Maxwellian-like experimentally simulated spectrum with temperature near 25 keV and measurements by Macklin [36]. That evaluation is approximately 5% to 7% below the results of the standards evaluation. That discrepancy has led to several measurements; that calculation is approximately 2 standard deviations from the Ratynski and Käppeler value. New JRC-IRMM measurements [41] of the $^{197}$Au(n,$\gamma$) cross section at the GELINA facility with a 1.5% total uncertainty are within 2% of the standards evaluation. Earlier measurements by Borella et al. [42] at GELINA are also in good agreement with the standards evaluation. Fig. 7 shows a comparison of measurements with the standards evaluation. The standards evaluation below the energy range for which it is considered to be a standard is shown as dots.

Work was done to check the simulated Maxwellian spectrum used in the Ratynski and Käppeler measurements. Measurements of that spectrum at PTB [43] are slightly softer, but have an effect of only 0.5% on the averaged Au cross section. A comparison [44] with thick target yields calculated using the PINO code and evaluated microscopic differential cross sections gives good agreement with the experiment. Independently, measurements of the neutron spectrum at JRC-IRMM by Feinberg et al. [40] showed good agreement with the findings of Ratynski and Käppeler and of the PTB result. Thus the discrepancy apparently is not due to the use of a poor spectral representation.

V. CONCLUSIONS

An IAEA Data Development Project, Maintenance of the Neutron Cross Section Standards continues to provide a mechanism for allowing new experimental data and improvements in evaluation procedure to be used in new evaluations of the neutron standards and associated covariances.

Acknowledgements: The support of the IAEA Nuclear Data Section and the United States Department of Energy in carrying out this work is appreciated.


[41] C. Massimi et al., submitted to EPJA.

