Status Of The International Neutron Cross Section Standards File

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Abstract. A report is given of the progress achieved in an IAEA Co-ordinated Research Project (CRP) to improve the cross sections standards. The objectives of the CRP, started in 2002, were initially the understanding of the origin of the strong uncertainty reduction in R-matrix model fits and the improvement of the evaluation methodology. These aims were extended in 2003 to the preparation of new evaluations for the standard ^{6}Li(n,t), ^{10}B(n,α), ^{10}B(n,α_1), ^{197}Au(n,γ), ^{235}U(n,f) and ^{238}U(n,f) reactions. The methodology, codes and experimental database developed by Poenitz and Hale for the ENDF/B-VI standards evaluation were taken as the basis for the new evaluation. The major results achieved by the CRP participants include the testing and intercomparison of a number of codes that can be used in the standards evaluation, updating the database of experimental results, analysis of the reasons leading to the strong uncertainty reduction in model fits, and a study of the bias in evaluated data caused by the Peelles's Pertinent Puzzle (PPP) effect, that has been widely discussed in the nuclear data community since the ENDF/B-VI standards evaluation was completed. Preliminary results of new standards evaluation are shown. The use of the new ^{235}U(n,f) cross section leads to better consistency in calculations of some important integral experiments.

INTRODUCTION

The previous evaluation of the neutron cross section standards was completed in 1987 and disseminated as the NEANDC/INDC [1] and ENDF/B-VI [2] standards. A joint list of reactions used in both sets is shown in Table 1. The ^{3}He(n,p) reaction was included only in the ENDF/B-VI standards and the ^{238}U(n,f) reaction only in the NEANDC/INDC standards. The evaluation was based on R-matrix model fits of experimental data for reactions leading to formation of the compound nuclei ^{7}Li and ^{11}B [3], and on a non-model generalized least-squares fit for the heavy nuclide standards [4]. Reaction ratios were a substantial part of the experimental database, resulting in uncertainties that can be presented only by common covariance matrices with blocks of cross-reaction correlations. A combination of these R-matrix and non-model fits produced all the standards except for H(n,n), ^{3}He(n,p) and C(n,n) which were obtained from independent R-matrix analyses. Unfortunately, the evaluated

uncertainties from this approach were so low that many cross section users considered them to be unrealistic. The percentage uncertainties for the evaluated data were considered to be approximately a factor of two lower in the non-model heavy nuclide evaluations, and from 4 to 10 times too low for the R-matrix model light nuclide evaluations. Consequently, these uncertainties were scaled up by factors of between 2 and 10 to the "expected values" and simple diagonal covariance matrices were assigned for the uncertainties [1].

TABLE 1. Neutron Cross Section Standards .

Reaction	Neutron Energy Range
H(n,n)	1 keV to 20 MeV
^{3}He(n, p)	0.0253 eV to 50 keV
^{6}Li(n, t)	0.0253 eV to 1 MeV
^{10}B(n, α)	0.0253 eV to 250 keV
^{10}B(n, $\alpha_1\gamma$)	0.0253 eV to 250 keV
C(n, n)	0.0253 eV to 1.8 MeV
Au(n, γ)	0.0253 eV, and 0.2 to 2.5 MeV
^{235}U(n, f)	0.0253 eV, and 0.15 to 20 MeV
^{238}U(n, f)	threshold to 20 MeV

A difficulty may appear when fitting strongly correlated discrepant data: there is a systematic bias of the evaluated data relative to the expected "true" values (Peelle's Pertinent Puzzle (PPP)). PPP has been discussed and studied in the nuclear data community since this standards evaluation was completed (see e.g. [5]). Another point of concern is that the standards evaluations were based solely on two codes: EDA multi-channel R-matrix [3] and GMA generalized least-squares codes [4].

An IAEA Co-ordinated Research Project (CRP) entitled "Improvement of the Standard Cross Sections for Light Elements" was initiated in 2002 to explore the strong uncertainty reduction observed in R-matrix model fits, to improve the methodology for determination of the covariance matrices of the uncertainties, and to prepare newly recommended cross-section standards for the light nuclides. However, the scope of this CRP was subsequently extended in 2003 to cross-section standards for heavy nuclides at the request of the nuclear data community, and the timetable was reduced so that the first release of a test version of the recommended data would occur by the end of 2004. The methodology, codes and experimental database developed by Poenitz and Hale for the ENDF/B-VI standards were adopted as a suitable starting point. CRP participants determined appropriate solutions to overcome the difficulties experienced in the 1980s evaluation. Most of the work and the advances within the CRP are reported here and documented in summary reports of the two Research Co-ordination Meetings (RCM) held to date [6, 7]. A third RCM will be held 18 – 22 October 2004 in Vienna.

CODES INTERCOMPARISON

Two other R-matrix codes (SAMMY [8] and RAC [9]), two other non-model least-square codes (GLUCS [10], SOK [11]) and a code based on an analytical approximation model (PADE2 [12]) were used for inter-comparisons and evaluations. Using these codes is especially important for testing the implementation of the error propagation law within the various codes. The codes referred to above and those used in the ENDF/B-VI standards evaluation were tested and intercompared in the fit of 5 pseudo-experimental data sets (TEST1) prepared from real experimental data for the ^{6}Li(n,t) reaction [6]. The covariance matrices of uncertainty for these data sets included only two components: normalization (long energy range correlation (LERC)) and statistical (short energy range correlation (SERC)) components. A third component presenting medium energy range correlations (MERC) was not included, because the EDA code cannot use this component in the treatment of the uncertainty of the experimental data.

By analyzing the results of these comparisons, it was found that the version of the GMA code available to the CRP participants contained an error. Due to this error, only the last data set of each type was used in the fit and the evaluated values were then dependent on the order of the experimental data in the input to the code. This error did not effect the covariance matrix of the uncertainty of the evaluated data. After correcting the error, the fits obtained with GMA and GLUCS were consistent within 0.2%. These small differences can be explained by the numerical precision of the solution of the different matrix equations in the Gauss-Markov-Aitken approach of GMA and in the Bayesian approach of GLUCS.

GMA had shown numerical stability in another test when the same experimental data set was repeatedly introduced. Convergence to the expected values was observed. The results of fits (evaluated values and covariance matrices) using the TEST1 data and the codes RAC and PADE2 were different from those obtained with the GMA and GLUCS codes. As shown below, these differences are explained by the use of models for the RAC and PADE2 codes. The inter-comparison of cross sections calculated by EDA, RAC and SAMMY from the same set of R-matrix parameters showed only very small differences. EDA uses a relativistic approach, whereas RAC and

SAMMY use a non-relativistic approximation, that leads to slightly different shapes for resonances.

AMBIGUITY IN R-MATRIX MODEL FITS

The R-matrix model fits of experimental data have shown that the parameters obtained with these codes and the cross sections reconstructed from these parameters are not unique, and sometimes differences between the fits are above the evaluated uncertainties. This phenomenological model determines parameters from the fitting of experimental data; how the data and their uncertainties are introduced in the fitting procedure can substantially influence the evaluated parameters and their uncertainties.

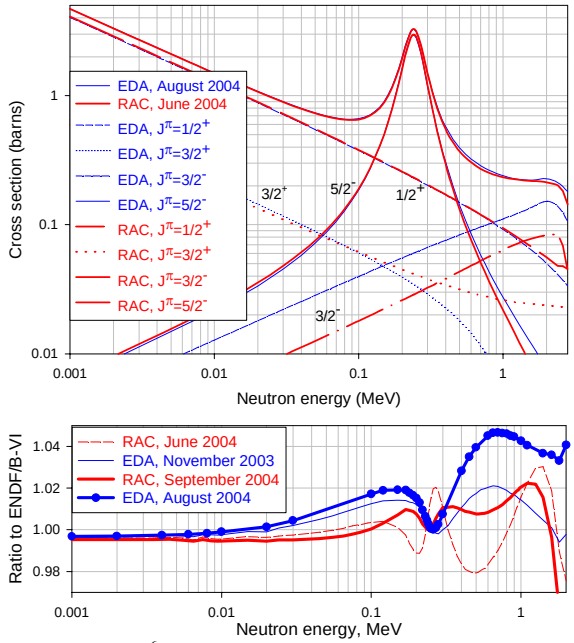

FIGURE 1. ^{6}Li(n,t) reaction R-matrix fits obtained with EDA and RAC codes with contribution of different channels (top) and their ratio to ENDF/B-VI evaluation (bottom).

Some results of fits by the EDA and RAC codes for the ^{6}Li(n,t) reaction are shown in the upper part of Fig. 1, and their ratio to the ENDF/B-VI evaluation in the lower part of Fig. 1. The uncertainty of the RAC fit is about 0.6 % over the energy range from 0.1 to 1 MeV, and the difference with EDA is well above this value. The reasons that the new EDA values are so different from the old EDA evaluation for the ENDF/B-VI standards are mainly due to an accounting change for competition in the 3-body break-up (n+d+α) channel above 2 MeV and inclusion of new data. Addition of Macklin's 1979 data (EXFOR sub-entry number 10895002) covering a wide energy range from 0.07 to 3 MeV has a major influence on the fit (compare EDA November 2003 and August 2004 fits at the lower part of Fig. 1). The RAC fit (June 2004) differs strongly from the EDA fit (November 2003) in the vicinity of the 0.24 MeV resonance. The origin of this discrepancy can be seen from a J^{π} partial channel expansion, arising from a shift of about -1.5 keV in the position of the $5/2^{-}$ resonance in EDA compared with that of RAC and also a relative decrease of 0.8%. This discrepancy was substantially reduced when uncertainties of the experimental charged-particle angular distributions in the RAC fit (September 2004) were considered as statistical with free normalization as in the EDA fit. The shift in position of the resonance has practically disappeared, and the difference in the shape of the resonance can be explained by a slightly different J^{π} channel expansion.

Other reasons leading to RAC-EDA fitting differences are as follows:

- RAC weighs the squared deviations of the chi-square functional by the inverse of the full covariance matrix of the uncertainties, while the EDA expression is the sum of terms weighted separately by the statistical and normalization components of the uncertainties. No medium energy range correlations in the covariance matrices of the experimental data can be accounted for by EDA. The covariance matrices of the uncertainties of the evaluated data obtained with EDA differ substantially from those obtained with RAC;

- the present EDA fit for the ^{7}Li system allows more normalization freedom than permitted by RAC using separate normalizations on angular distributions at different incident energies for some data sets. RAC treats them as absolute cross sections (if they really are) with all correlation components taken into account;

- for discrepant data, the option to increase the uncertainties of outlying data is often used in RAC. EDA does not usually adjust the uncertainties of fitted data, but scales its output covariances by the chi-square per-degree-of-freedom value of the fit. This procedure is equivalent to increasing the uncertainty of all the experimental data in the fit, not just the outliers;

- experimental data up to 25 MeV in neutron incident energy were used in the RAC fit but only up to 4 MeV in the EDA fit.

The R-matrix fits presented here are not final; improvements in the convergence of the EDA-RAC

results can be expected prior to the completion of this work.

UNCERTAINTIES OF EVALUATED DATA IN MODEL AND NON-MODEL FITS

There are several reasons why strong uncertainty reduction was observed in EDA R-matrix fits of standard reactions. Analysis of components of uncertainties describing the correlations between results of different measurements and even correlations within the same measurement was not as detailed as for the GMA database. Because of the way the uncertainties of experimental data are introduced in the EDA data fitting procedure, some important components of the data uncertainties are missed, leading to a reduction of the level of correlations in the covariance matrices of the experimental data.

Inclusion of a large amount of data for charged-particle-induced reaction channels also contributes to a large uncertainty reduction. Many charged-particle data, especially differential elastic scattering cross sections, are claimed to have very small uncertainties. Systematic errors probably have not been fully estimated.

Often those who analyzed uncertainty reduction paid attention only to the percent uncertainties or variances. But there is a substantial difference in covariance matrices obtained in the model and non-model least-squares fits. Fits with a model function reduce the variances substantially, but increase covariances near the diagonal. As a rule, this redistribution approximately conserves the sum of all elements of the covariance matrix. Because uncertainties cannot be characterized only by variances, it is difficult to discuss uncertainty reduction in a model fit and non-model fit comparing only variances. A row of the covariance matrix of evaluated data obtained with the non-model GMA is compared with RAC and PADE2 model fits in Fig. 2. The result obtained with GMA is the same as that obtained with GLUCS and SOK. The TEST1 data sets for the ^{6}Li(n,t) reaction were used in all fits. It is clearly seen that the model fits reduce the variance by about a factor of 2 compared with the non-model. But the sum of the covariances along this row of the matrix is 0.01116 barn^2 for GMA, 0.01119 barn^2 for RAC and 0.01101 barn^2 for the PADE2 fits. The reduction of variance in model fits is compensated by an increase in neighboring covariances.

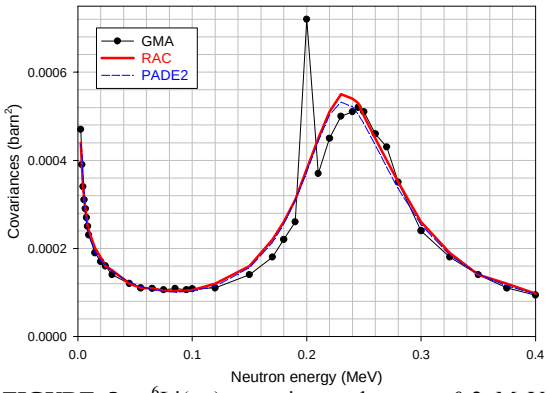

FIGURE 2. ^{6}Li(n,t) covariances between 0.2 MeV and other energy points evaluated with model (RAC, PADE2) and non-model (GMA) fits of TEST1 data.

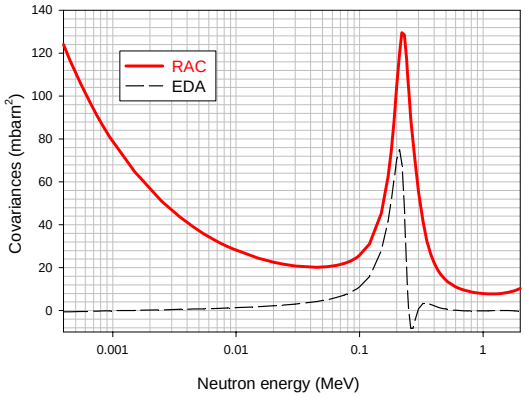

FIGURE 3. ^{6}Li(n,t) covariances between 0.2 MeV and other energy points evaluated in model fits of all experimental data.

The use of a chi-square function with a coherent contribution of statistical and normalization components leads also to a strong reduction of covariances in EDA compared with that in a RAC fit. As mentioned, RAC implements a full error propagation law, where the covariance matrix of the uncertainties of experimental data should be inverted in the chi-square expression. Because the inverse of the sum of the normalization and statistical components (RAC) is not equal to the sum of the inverse of the statistical and normalization components (EDA), their chi-square expressions cannot be reduced to show they are equivalent to each other. Fig. 3 shows a comparison of covariances for rows with variances at 0.2 MeV of the matrix of uncertainties evaluated with RAC and EDA for the ^{6}Li(n,t) reaction. The covariances obtained with RAC contain a rather large LERC component propagated from the covariance matrices of the experimental data. For EDA, it seems that only the intrinsic model correlation

properties are present in the covariance matrix of uncertainty of the evaluated data.

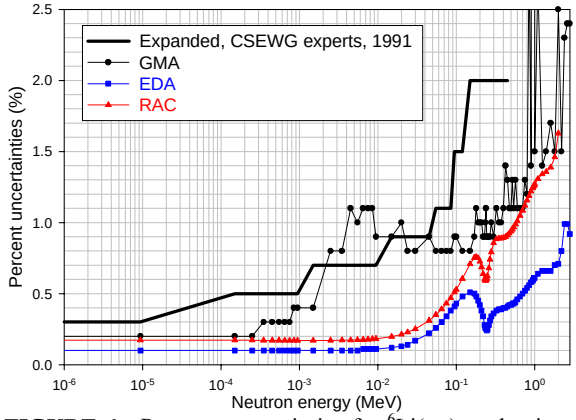

FIGURE 4. Percent uncertainties for ^{6}Li(n,t) evaluation as estimated by the CSEWG, and obtained in model (EDA, RAC) and non-model (GMA) fits.

The percent uncertainties obtained in different fits are shown in Fig. 4 compared with the estimated (expanded) uncertainties provided by the ENDF/B-VI standards evaluators of the Cross Section Evaluation Working Group (CSEWG). GMA is a non-model fit with neutron-induced reactions, ratios and constraints included in its database. RAC and EDA are model fits (with uncertainties reduced to obtain chi-square per degree of freedom of about 1) of neutron and charged-particle induced reactions forming the ^{7}Li system. The uncertainty of all fits in the low energy region is dominated by the 0.2% uncertainty of the thermal pre-evaluated value. In R-matrix model fits, this low uncertainty propagates in the *1/v* energy region. GMA has no *1/v* constraint on the shape above 1.5 keV and, as a result of this, the GMA uncertainty in this energy region is increased up to the level of the CSEWG expanded uncertainties. The CSEWG estimation above 0.1 MeV is based on considering the data only for the ^{6}Li(n,t) reaction. Including total cross sections, elastic cross sections and ratios to cross sections of other reactions in the GMA fit and charged-particle cross sections in the R-matrix model fit reduces the evaluated uncertainty in this region.

PEELLE'S PERTINENT PUZZLE IN EVALUATION OF STANDARDS

Statistical analyses of the derived cross sections pose problems that lead to PPP (attributed to a loss of information after the original measured values (number of counts) have been processed). PPP occurs in the fitting of the five data sets for the ^{6}Li(n,t) reaction selected for the codes inter-comparison studies (TEST1). The energy dependence of the cross section is the same as shown in Fig. 1. For convenience, all fits are shown in Fig. 5 as ratios to a GMA fit using the Chiba-Smith approach [13], which is considered nearly free of the PPP problem (GMAP). The approach is based on assigning absolute uncertainties to the experimental data as their percent uncertainties relative to a posterior evaluation. Because the final evaluation is not known in advance, the iteration of fits starting from some appropriate prior values is needed. The Box-Cox transformation, which in this case is very close to a logarithm transformation, is another approach to exclude PPP. As we can see, the Box-Cox (and the logarithm transformation which is not shown) result is very close to GMAP, while the GMA result with standard least squares fitting procedures is biased at about 10% and is below the bulk of the experimental data.

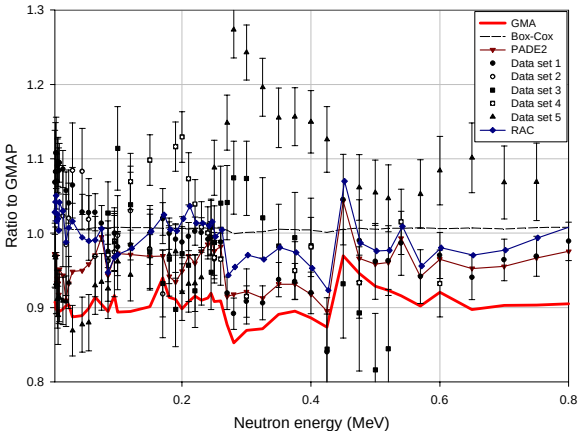

FIGURE 5. PPP manifestation in the fit of data for the TEST1 case and results obtained using different options for minimizing its effect.

The chi-square per degree of freedom for the GMA fit was about 14, but increasing the uncertainties of all the experimental data by the square root of chi-square per degree of freedom will not change the results. To restore consistency between the data, the outlying experimental data should be determined, the reasons for discrepancies should be analysed and either the data should be corrected or additional components of uncertainties should be added. RAC (R-matrix) and PADE2 (analytical expansion) fits also show some bias caused by PPP, although the effect in these pole-type expansions (transformations) is much less than in the non-model fits.

Comparing GMA results obtained with the Chiba-Smith option "on" or "off" in fits of all data included in the standards database shows that the PPP bias is within 30-50% of the uncertainty of the evaluated data.

For the ENDF/B-VI evaluation of the standards, the uncertainties of outlying experimental data were modified so that chi-square per degree of freedom for the whole standards database was close to 1.

UPDATING OF GMA DATABASE

The GMA database for the standards included 415 experimental data sets, pre-evaluated values and thermal constants [14]. Some non-standard reactions were in the database because of their high accuracy and available ratios to the standards. The data for the GMA portion of the ENDF/B-VI standards evaluation used the following reactions and ratios between them for the fit: ^{6}Li(n,t), ^{6}Li(n,n), ^{6}Li(n,tot), ^{10}B(n,α_0), ^{10}B(n,α_1), ^{10}B(n,α), ^{10}B(n,n), ^{10}B(n,tot), ^{197}Au(n,γ), ^{238}U(n,γ), ^{235}U(n,f), ^{238}U(n,f) and ^{239}Pu(n,f). Data with maximum incident neutron energies of 20 MeV were included for the fission cross sections, but the maximum incident energy was much less (1 or 2.8 MeV) for the other cross sections. Pre-evaluated values for the ^{6}Li(n,t), ^{197}Au(n,γ) and ^{238}U(n,γ) thermal cross sections, as well as 26 thermal constants for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu as evaluated by Axton [15], were part of the fit. The present evaluation effort has led to the addition of more than 30 high precision measurements.

For the present evaluation, the energy range for the fission reaction cross sections was extended up to 200 MeV which allows the low and high-energy standards to be evaluated in one combined fit. Unfortunately, some discontinuities do arise because of the small number of absolute cross-section measurements at high energies and differences in the fission cross sections above 20 MeV as measured in different laboratories, resulting in rather large uncertainties for the recommended high-energy fission standards. The results of nuclear model calculations were used as shape cross section "patches" to smooth cross sections in these areas.

Chi-square per degree of freedom for the fit of all data included in the GMA database was about 4. To reduce it, the data which are outliers relative to the true evaluation should be identified, and the data or their uncertainties should be corrected. Because we do not know the true evaluation at the beginning of the fit, we can use a good prior for identifying the outliers and iterative fitting converging to the true evaluation. Here the ENDF/B-VI evaluation was taken as a good prior and 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 component was evaluated from an analysis of the energy dependence of the discrepancy. After the first fit (iteration) the posterior evaluation was taken as the true evaluation and these additional components were either decreased or increased. Two iterations were enough for convergence. The outliers were classified if the difference from the true value was above two standard deviations for a single point or above one standard deviation for a few sequential points. After following this process, the chi-square per degree of freedom for the fit was about 0.8.

PRELIMINARY RESULTS AND TRENDS IN THE NEW EVALUATION

The procedure for combining the results of an R-matrix model fit for light nuclides and a non-model fit for heavy nuclides can be implemented in many different ways. Here the simplest procedure was used, where non-redundant cross sections and covariance matrices of their uncertainties reconstructed from the R-matrix parameters and their covariances were introduced as pseudo-experimental data sets in a combined GMA fit with the full GMA database. To avoid double counting, data sets used in the R-matrix fit were eliminated from the GMA database. The covariance matrix of the uncertainties of these pseudo-experimental data sets is not semi-positive definite because the number of parameters is less than the dimension of this matrix. But this does not cause problems with the numerical inversion of this matrix in the GMA solution and the evaluated matrices are "good" semi-positive definite matrices.

The trend in the new standards evaluation is a general increase in practically all cross sections and especially fission cross sections above 14.5 MeV. These changes are a result of the removal of an error in the original GMA code and including new experimental data (0.5% to 4% increase), and the elimination of the PPP problem (0.1% to 0.5% increase). Preliminary results are shown in Fig. 6 for the ^{235}U(n,f) cross section compared with the 1987 standards evaluation. The error bars are the uncertainties of the new standards. Some previously observed discrepancies are now resolved, e.g., results from high-precision ^{238}U(n,f) cross section measurements (1.2%) at about 14 MeV, undertaken at several laboratories [16], are in good agreement with the new evaluation.

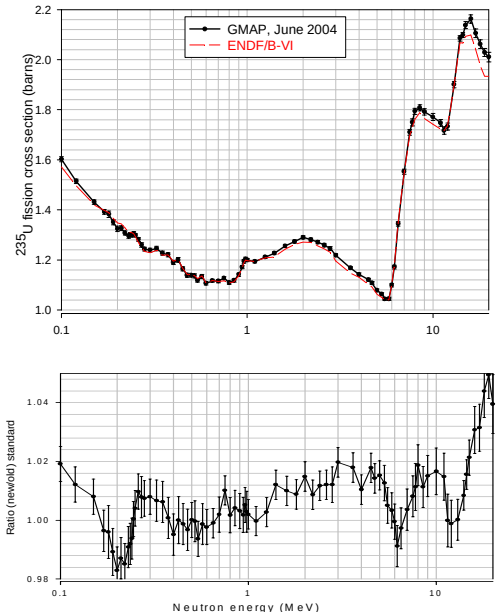

FIGURE 6. Comparison of old and preliminary new standards for the ^{235}U(n, f) cross section.

An important discrepancy in the ratio of the ^{10}B(n,α_0) to ^{10}B(n,α_1) cross sections appeared with new results by Weston [17], as shown in Fig. 7. Recently work has been undertaken by Hambsch on this ratio using a Frisch-gridded ionization chamber [18]. His latest analysis of these data was based on fits of the angular distributions of alpha particles in the ranges of angles which are free from the effects of "particle leaking" (which results from cases where both reaction products go in the forward direction). Their values were determined with an assigned systematic uncertainty of 2 - 4%. As can be seen in Fig. 7, the branching ratios determined from such an analysis are rather close to the ENDF/B-VI evaluation, the RAC (2004) evaluation done without using these data, and the GMA fit which includes these data.

Preliminary calculations using the new evaluation of the ^{235}U fission cross section show that the k-effective value for GODIVA (a ^{235}U fast neutron benchmark) can be predicted to within 0.1%. Another important benchmark value for ^{235}U fission is the K1 integral parameter for the ^{235}U thermal constants. Hardy has recommended the value of 722.7±3.9 barn (as taken from [15]), which gives the best characterization of k-effective for a system with a thermal neutron spectrum. Axton's thermal constants [15] which are used as pre-evaluated values in the GMA fit give K1 = 718.57 ± 2.22 barns. The GMA fit, using a highly accurate scattering cross section calculated from coherent scattering data [19] added to the database, gives 719.67 barns which is within the limits of the uncertainty of Hardy's value. Better agreement with Hardy's value can probably only be obtained with a re-evaluation of nu-bar for ^{235}U(n,f). Data on nu-bar for ^{235}U(n,f) at the time when Axton's evaluation was prepared were underestimated because the energy dependence of nu-bar was not well known at that time. Linear extrapolation of nu-bar used for reduction of data measured from 70 to 120 keV underestimates the thermal value. The most accurate measurement of nu-bar at an energy of 0.0253 eV was carried out by Gwin et al. [20]. Axton used Gwin's data (based on 3 sets of measurements) with a rather modest uncertainty assignment. If Gwin's data is used with the lowest uncertainties which can be assigned to them (0.12%), we obtain K1 = 721.35 barns in a GMA fit. More detailed tests and benchmarks need to be performed as part of a critical assessment of the new evaluations.

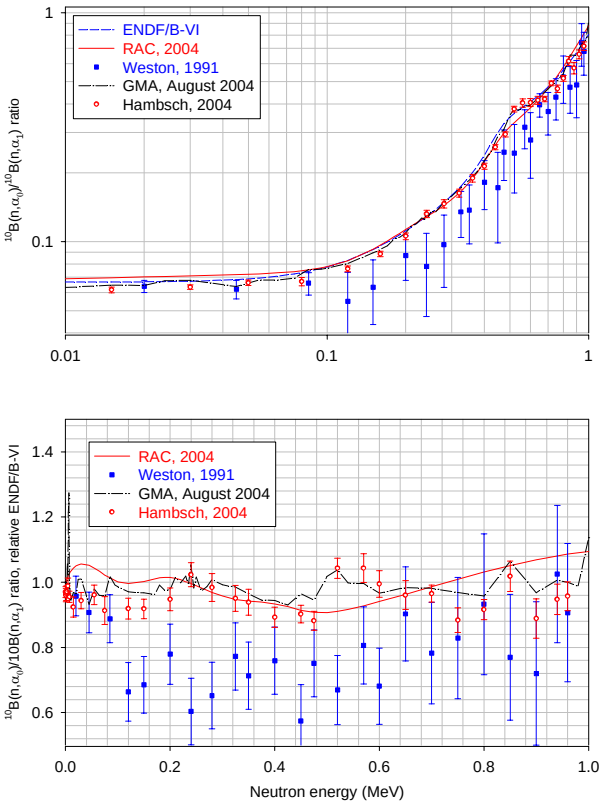

FIGURE 7. Comparison of measurements and calculated results from various evaluations for the ^{10}B(n,α) branching ratio.

EVALUATED DATA PRESENTATION

The data will need some post-processing, before release of the newly evaluation standards. This exercise will include a smoothing of some unphysical variations of the cross sections, and presentation of the

cross sections and the uncertainties in formats that are convenient for different users. A simple and well-controlled smoothing algorithm has been prepared and applied for the regions where the variations in the cross sections do not present the real physical behavior and are artifacts of the numerical procedures. Evaluated cross sections and covariance matrices will be presented in ENDF format, as well as simple tables of cross sections and their percent uncertainties. But it should be noted that for any use of the standards based on application of the error propagation law (calculation of integral quantities, use in further evaluations, etc.), the full covariance matrix of the correlated cross sections and constants should be used.

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