SAND2010-2475C

International Conference on Nuclear Data for Science and Technology 2010

USE OF MODEL-BASED COVARIANCE ESTIMATES IN DOSIMETRY APPLICATIONS

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Received
Accepted for Publication

Improved nuclear physics modelling codes and computational methods have made significant advances in recent years and have enabled the cross section modelling community to automate the parameter variation and generate model-based cross section evaluations that include covariance matrices. Current dosimetry community standards do not permit the use of purely model-based cross sections for many applications, e.g. for spectrum adjustment in support of pressure vessel surveillance activities. This paper examines the implications of using model-based cross section libraries for typical dosimetry applications.

KEYWORDS: ND2010, Nuclear Data, Dosimetry, Covariance, Uncertainties, ASTM, Spectrum Adjustment

1. INTRODUCTION

With the availability of model-based cross sections and covariance matrices produced by nuclear data codes such as EMPIRE [1] and TALYS [2], the community is considering the role these covariance matrices should play in nuclear reactor dosimetry applications. The purpose of this paper is to examine the implications to nuclear reactor dosimetry applications of some suggested approaches to the use of model-generated cross section data. A major concern is sufficiency and accuracy of the uncertainty estimate when used for dosimetry applications.

2. BACKGROUND

Many current standards, e.g. ASTM E1018 [3], require that cross sections used for dosimetry applications have a covariance matrix consistent with the methods used to derive the cross section and one that has given due consideration to the available experiment data. This guidance was motivated, in part, by the decision in the late 80s by one dosimetry library to keep its evaluations but to apply the IRDF85 covariance matrices to their evaluations. The dosimetry community soundly rejected this approach, and that dosimetry library soon replaced the covariance matrices with high quality consistent covariance matrices that reflected the evaluations they recommended. The need for a covariance estimate to match the derivation of the basic cross section data is fairly intuitive. Who would have permitted an uncertainty estimate derived for a time measurement using the resonance frequency of a quartz crystal to be applied to a time measurement obtained from a sundial? Despite this seemingly self-evident principle, as recently as 2008 there were serious suggestions to apply purely model-based covariance matrices to existing ENDF/B-VII evaluations when they were issued in future revisions.

This paper assumes a consistent pairing of a covariance matrix with the cross section, and explores the adequacy of model-based covariance estimates for dosimetry applications. While it is straightforward to do correlated sampling on the input parameters of a model, it is not always clear how to capture underlying correlations intrinsic to the theory that is being applied. This underlying correlation can have a significant effect on uncertainties derived by the parameter variation and can affect the interpretation of results derived from use of the covariance matrix. A simple example of this is the unrealistically low uncertainty for a ^{235}U thermal fission spectrum that results

[1] This work was supported in part by the U.S. Department of Energy under contract DE-AC04-94AL85000. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy.

of one uses an *a priori* covariance matrix based on a simple two-parameter Watt fission representation [4].

3. DOSIMETRY FOR 14-MEV DT SOURCES

3.1 Details of the Application

The nuclear metrology community captures guidance for its applications in various community standards. For dosimetry at neutron generators utilizing the ^{3}H(d,n)^{4}He reaction, this guidance is encapsulated in the ASTM E 496 Standard Test Method for Measuring Neutron Fluence and Average Energy from ^{3}H(d,n)^{4}He Neutron Generators by Radioactivation Techniques [3]. This test method recommends the use of some dosimetry reactions that have a flat cross section in the vicinity of 13.5 – 15.5 MeV as good fluence monitors, e.g. the ^{93}Nb(n,2n)^{92m}Nb reaction. For the determination of the average neutron energy, the recommendation is to use the ratio of the activities for two reactions, one that is decreasing in this energy range and one that is increasing. An example of such a reaction ratio is the ratio of ^{54}Fe(n,p) to the ^{58}Ni(n,2n) reactions. This reaction has a large slope in this energy region and a small uncertainty.

3.2 Comparison of Model-based and Experimental Results

Table 1 lists the various reactions[2] that are used in Test Method E496. It also compares the ratio of the cross sections and the separate uncertainties found for the TENDL-2008 model-based evaluation [2] with the recommended IRDF-2002 [5] cross section evaluation. The last column in Table 1 shows the number of standard deviations by which the cross section ratio (IRDF-2002/TENDL-2008) differs from unity. Figure 1 shows a comparison of the ^{58}Ni(n,2n)^{57}Ni experimental data in the 14-MeV energy range from the EXFOR database [6] with the IRDF-2002 and the TENDL-2008 cross section evaluations.

3.3 Observations on Comparison

An inspection of Table 1 shows that the uncertainty for the IRDF-2002 dosimetry quality cross-section evaluations have a standard deviation that is, on average, a factor of 8X smaller than the model-based TENDL-2008 evaluations. This trend is expected since the IRDF-2002 cross section evaluations have been able to use the measured cross sections as a constraint on the evaluation and associated uncertainty, but the magnitude of the difference is a concern for dosimetry applications. What is even more troubling about the results depicted in Table 1 is that, when one forms the ratio of the IRDF and TENDL cross sections at 14-MeV and uses RMS sum of the respective uncertainties to represent the uncertainty for the ratio, the last column in Table 1 shows that 3 of the 13 entries, or 23% of the reactions, show a cross section ratio that deviated by more than two standard deviations from unity. This indicates that the TENDL cross sections tend to under-estimate the cross section uncertainty in the 14-MeV region.

Table 1 Reactions used in ASTM Test Method E 496

Reaction	Cross section ratio	TENDL Unc. (%)	IRDF2002 Unc. (%)	# Std. Dev.
^{27}Al(n,α)	0.87	15.0	3.74	0.84
^{27}Al(n,p)	0.62	14.9	7.31	2.28
^{63}Cu(n,α)	0.83	23.4	1.61	0.73
^{63}Cu(n,2n)	0.99	10.8	1.30	0.13
^{65}Cu(n,2n)	0.89	6.93	1.51	1.62
^{54}Fe(n,p)	1.06	4.46	3.37	1.03
^{56}Fe(n,p)	0.98	18.5	1.07	0.10
^{24}Mg(n,p)	0.96	10.1	0.70	0.41
^{58}Ni(n,2n)	1.40	17.0	2.38	2.31
^{58}Ni(n,p)	0.98	5.26	2.04	0.39
^{32}S(n,p)	0.67	12.8	1.33	2.58
^{64}Zn(n,p)	0.90	16.6	1.81	0.60
^{90}Zr(n,2n)	1.08	10.6	0.59	0.77

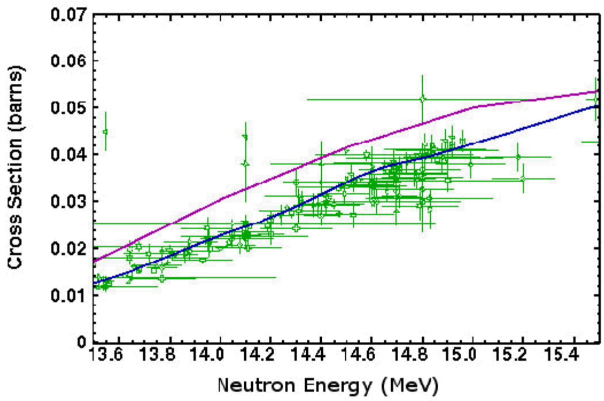

Fig. 1. Comparison of Cross Section Evaluations with Experimental Data for the ^{58}Ni(n,2n)^{57}Ni Reaction

In the Figure 1 comparison for the ^{58}Ni(n,2n)^{57}Ni reaction, the IRDF-2002 cross section is, as expected, seen to be a very consistent with the experimental data. The TENDL cross section has a systematic offset from the IRDF and clearly is a poorer match to the data. The TENDL cross

[2] Note that the ^{93}Nb(n,2n)^{92m}Nb reaction does not appear in the table since the available model-based cross sections only record the total cross section, not the cross section to the meta-stable state.

section for this reaction quotes a 17% uncertainty, but it is seen to deviate from the IRDF and experimental values by ~39%, or ~2.3 standard deviations.

4. DOSIMETRY FOR STANDARD FISSION FIELDS: ^{252}Cf SPONTANEOUS FISSION

4.1 Details of the Application

When recommendations are made for the cross sections to be used by the dosimetry community, the decisions are typically influenced by the consistency of the cross section with integral measurements in the ^{252}Cf spontaneous standard neutron field [5]. Recommended evaluations, typically from IRDF-2002, and TENDL evaluations were compared for 29 reactions of dosimetry interest in the ^{252}Cf standard neutron field.

4.2 Comparison of Model-based Results with Experimental Results

Figure 2 shows the calculated-to-experimental (C/E) ratios for the recommended evaluations (red) and the TENDL (black) evaluations.

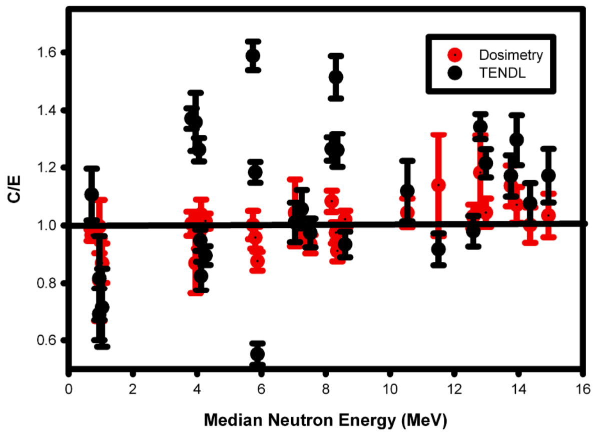

Fig. 2. Comparison of Foil Activities in ^{252}Cf Field

4.3 Observations on Comparison

Figure 2, as expected, shows good C/E comparisons for the recommended dosimetry cross sections in this standard neutron field. The maximum C/E ratio is 1.18 while the minimum is 0.811. The average number of standard deviations by which this ratio deviated from unity is 1.05, showing excellent agreement with what is expected from the formal definition of the cross section uncertainty. 17 of the 29 reactions, or 59%, showed a C/E that deviated by less than one standard deviation from unity. This is consistent with the expectation of 66% of the ratios being within one standard deviation for a normal distribution.

The C/E data for the TENDL cross sections, shown in black in Figure 2, show considerably more spread and show many points where the ratio is more than one standard deviation from unity. The maximum C/E ratio is 1.59 while the minimum is 0.55. The average number of standard deviations by which this ratio deviated from unity is 3.8, considerably exceeding the statistical expectations for a normal distribution. One reaction had a C/E ratio that deviated from unity by over 12 standard deviations. Only 4 of the 29 reactions, or 14%, showed a C/E that deviated by less than one standard deviation from unity – this is inconsistent with the expectation of 66% of the ratios being within one standard deviation for a normal distribution.

5. SPECTRUM ADJUSTMENT

5.1 Details of the Application

One of the most important applications of dosimetry cross sections is a neutron spectral adjustment. Typically a prior spectrum is obtained from a high fidelity calculation and a least squares formalism is used to determine an adjusted spectrum along with an energy-dependent covariance matrix [7]. The covariance matrix can be used to obtain uncertainties in various integral metrics. This application is addressed in ASTM standards E844 and E944 [3].

5.2 Comparison of Model-based Results with Experimental Results

We have selected foil activity data used in a neutron spectrum for a fast burst reactor (FBR) and for a pool-type reactor. These two spectrum variations bound the expected variation by the dosimetry community and provide insight into what might be seen in a typical spectrum adjustment application. In this example, we have taken existing sets of experimental data and *a priori* calculated spectra and then pruned down the activity set to only include those reactions that are in the TENDL-2008 library, e.g. we have eliminated fission reactions and reactions to a meta-stable state (non-ground state) of a residual isotope. A least squares spectrum adjustment was than made using the LSL code [8]. Two identical spectrum adjustments were made, varying only the cross section library. Tables 3 and 4 show a comparison of some spectral metrics for the two types of reactor environments along with the associated uncertainty.

Table 2 Spectrum Adjustments for a Fast Burst Reactor

Metric	IRDF-2002	TENDL	Ratio
dof	8		---
Chi-sq per dof	1.313	1.889	---
Fluence > 3 MeV	6.897E13 ± 5.6%	8.894E13 ± 5.7%	0.775 ± 8.0%
Fluence > 1 MeV	2.409E14 ± 9.6%	2.461E14 ± 10.1%	0.979 ± 13.9%
Fluence > 0.1 MeV	4.834E14 ± 10.8%	4.794E14 ± 11.1%	1.008 ± 15.5%
Fluence > 10 keV	4.994E14 ± 10.7%	4.949E14 ± 11.0%	1.009 ± 15.3%
Fluence < 1 eV	4.511E11 ± 20.0%	4.399E11 ± 20.1%	1.025 ± 28.4%
Fluence, total	5.00E14 ± 10.7%	4.960E14 ± 11.0%	1.009 ± 15.3%

Table 3 Spectrum Adjustments for a Pool-type Reactor

Metric	IRDF-2002	TENDL	Ratio
dof	31		---
Chi-sq per dof	1.89	7.535	---
Fluence > 3 MeV	1.646E13 ± 3.0%	1.663E13 ± 3.1%	0.990 ± 4.3%
Fluence > 1 MeV	7.785E13 ± 4.8%	7.772E13 ± 5.1%	1.002 ± 7.0%
Fluence > 0.1 MeV	1.790E14 ± 5.0%	1.777E14 ± 5.2%	1.007 ± 7.2%
Fluence > 10 keV	2.258E14 ± 4.7%	2.232E14 ± 4.8%	1.012 ± 6.7%
Fluence < 1 eV	4.490E13 ± 11.7%	4.507E13 ± 11.8%	0.996 ± 16.6%
Fluence, total	3.749E14 ± 3.19%	3.726E14 ± 3.2%	1.006 ± 4.5%

5.3 Observations on Comparison

The spectrum adjustment for the FBR, after the removal of the fission reactions and the reactions to a meta-stable state in the residual isotope, only used 8 reactions. The chi-squared (χ^2) per degree-of-freedom (dof) was very good, 1.313, for the IRDF cross section library, but was a poor 8.97 for the TENDL cross library. This indicates a conflict within the set of TENDL cross sections that is not adequately captured in the specified covariance matrices. The fluence > 3-MeV shows a very significant difference between the adjustments using the two different libraries – a difference that exceeds the stated uncertainties by nearly four standard deviations. This large difference is probably related to the small number of reactions used in the adjustment and the fact that only one or two reactions drive the fluence in this >3-MeV energy region. All the other fluence metrics show good agreement.

The spectrum adjustment for the pool-type reactor environment used 31 reactions. This adjustment has an acceptable χ^2 per dof of 1.89 for the IRDF library, but a large χ^2 per dof of 7.5 for the TENDL library. In this case, with the larger set of reactions, there was good agreement between the two adjustments for all of the fluence metrics.

6. CONCLUSION

This paper has examined the implications of using model-based cross sections and covariance matrices for several different types of dosimetry applications. The analysis shows, as expected, that the model-based cross sections have a significantly higher uncertainty than cross section evaluations that took the experimental data into account. But, this analysis also showed the much more troublesome issue that the model-based covariance matrices exhibited internal inconsistencies as exhibited by the χ^2 for the spectrum adjustments and understated the uncertainties relative to the best available validation data. The higher cross section uncertainty would be acceptable for many applications where higher quality data did not exist, e.g. for spectrum adjustment in neutron fields with a significant neutron fluence above 20 MeV. The understatement of the model-based uncertainty can probably be attributed to the fact that the uncertainty estimates were generated with model variation and missed intrinsic uncertainties in the model-based physics. This understatement of uncertainties and the resulting inconsistency between the cross sections from different reactions implies that exclusively model-based cross sections should not yet be used for most dosimetry applications. Some groups are looking into ways to add a consideration of available experimental data into model-based calculations. The combination of the more rigorous parameter variation in the calculations and the consideration of the experimental data have the promise of offering improved dosimetry cross section evaluation.

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