Improved Photon Production Data for MCNP™

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Abstract

Computer simulations with MCNP are often used to obtain information from measurements of neutron-induced gamma-ray spectra. For such simulations to be useful, the complicated spectra produced by a wide variety of nuclides must be reproduced, requiring high-quality nuclear data. A previous assessment of the neutron-induced photon production data in the MCNP data libraries indicated a need for improvement. The photon production data were often based on outdated experiments and binned in such wide energy groups as to be of limited value for some applications.

This paper describes the work that is underway at Los Alamos National Laboratory to improve the photon production data for thermal neutron capture reactions. To date, high-quality photon production data for each stable isotope of chlorine, chromium, iron, copper, and nickel have been obtained. The improved spectra have been incorporated into ENDF-formatted evaluations and processed into corresponding MCNP data files. Similar improvements for aluminum, manganese, silicon, calcium, and vanadium are also planned. The methodology used to produce the spectra is discussed, and sample results for chlorine are presented.

1. INTRODUCTION

Gamma-ray spectroscopy is used in a wide variety of applications such as oil well logging , the identification of environmental contaminants, and the determination of structural integrity. In many applications, a neutron source is placed near the sample of interest and the spectrum of gamma-rays emitted by the sample is measured. The spectrum is then analyzed to determine the isotopic makeup of the sample. Nuclides are typically identified by the presence of their strongest characteristic gamma-rays. The sample's composition can then be used to infer whether or not oil is likely to be found, what harmful materials are present, or how sound a structure is.

In practice, analyzing the complicated spectrum of gamma-rays emitted by an arbitrary sample is very difficult. A single nuclide may emit hundreds of measurable gamma-rays when exposed to a neutron source. Samples contain numerous nuclides of differing concentrations, and the gamma-rays they emit sometimes overlap, complicating the analysis. The equipment that is used to make the measurements also affects the quality of the measured spectra and must be considered in the analysis.

In addition to these analytical difficulties, it is often difficult to make benchmark measurements for every possible combination of sample composition, sample condition, and experimental configuration, especially when measurements must be made in the field. The high cost and time-consuming nature of such experiments means that spectra for only a few benchmark scenarios can be measured and thoroughly analyzed. Because of these limitations, computer simulations are performed to help interpret experimental measurements. For example, in oil well logging, a borehole is drilled into a formation that may contain oil and a tool is inserted to interrogate the borehole. The tool typically consists of a neutron source, various detectors and electronics, and structural and shielding materials. The device is complicated, expensive, and may affect the measured gamma-ray spectra significantly. When design

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changes are considered, simulations are performed to estimate the impact of the changes on the tool's performance. These simulations are much faster and cheaper than building and testing a number of prototype tools. When benchmark measurements are made, the data are compared to computer-generated spectra to validate the computer simulation codes and corresponding nuclear data libraries. Once a simulation code and its data libraries have been validated, many simulations can then be performed corresponding to many different sample compositions, sample conditions (wet, dry, hot, cold) and tool geometries.

MCNP,[1] with its ability to represent complex three-dimensional geometries and elaborate physical modeling, is one of the most widely used codes for calculating neutron-induced gamma-ray spectra. Unfortunately, its ability to calculate these spectra accurately is dependent upon the nuclear data it uses. The data libraries used by MCNP are based primarily on ENDF,[2] the Evaluated Nuclear Data File. A previous assessment of the photon production data contained in these nuclear data libraries revealed several inadequacies.[3] The ENDF photon production data are often based on outdated experiments, and for many elements, isotopic evaluations are not provided, or the elemental photon production data are used for each isotope. In many of the ENDF evaluations, the photon production spectra are binned over very wide energy regions (as large as 250 keV). An example of such wide binning is given in Figure 1, which shows the thermal-neutron capture spectra for magnesium as simulated by MCNP using both ENDF/B-V and ENDF/B-VI data. The ENDF/B-V thermal-neutron capture spectrum for magnesium is based on data from a 1970 publication by Orphan et al.[4] As Figure 1 shows, the ENDF/B-VI spectrum is just a translation from ENDF/B-V and does not employ a finer bin structure or make use of more recent measurements. In fact, the entire evaluation for magnesium did not change from ENDF/B-V to ENDF/B-VI, but was simply translated into the new ENDF/B-VI format. In many new ENDF/B-VI evaluations (Fe, Ni, Cr, etc.), the photon production data from non-capture reactions were greatly improved. Unfortunately, the photon production data for neutron capture were often not upgraded.

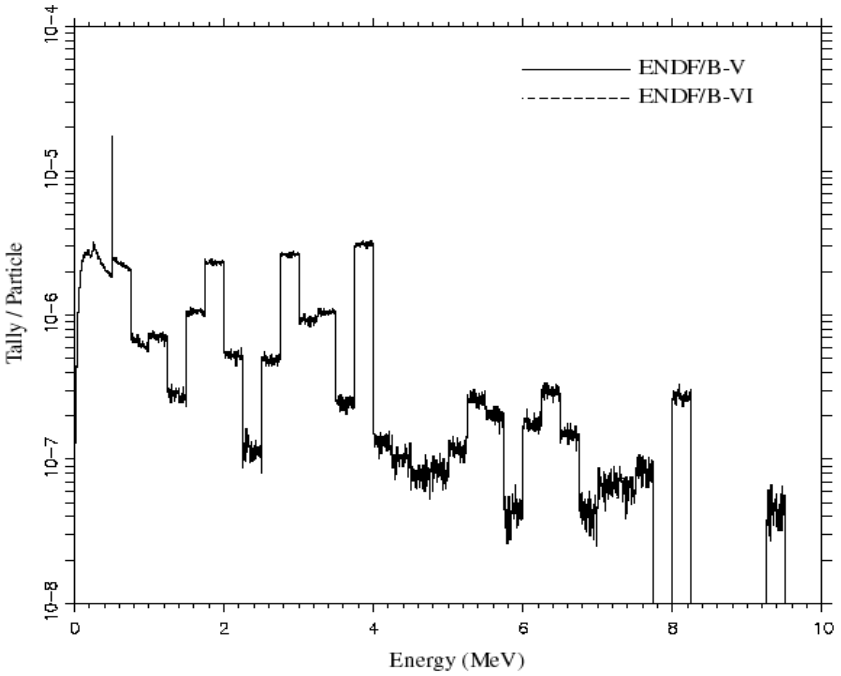

Fig. 1 ENDF/B-V and ENDF/B-VI thermal-neutron capture spectra for magnesium.

Because of these problems, work is underway at Los Alamos[5-8] to improve the photon production data for thermal-neutron capture reactions. This work is partly funded by a collaborative research and development agreement with several well-logging companies[9] and is driven by the need for high-quality thermal-neutron capture data for oil well logging calculations. This paper focuses only on work done to improve photon production data

from thermal-neutron capture. Gamma-rays produced by other neutron interactions, such as inelastic scattering, will not be discussed. Throughout this paper the phrase "photon production data" will refer only to the spectrum of gamma-rays produced by radiative capture of thermal-energy neutrons. The process of obtaining improved photon production data is discussed in Section 2. In Section 3 the status of this work is discussed and some sample results are presented. Finally, a summary is given in Section 4.

2. OBTAINING IMPROVED PHOTON PRODUCTION DATA

2.1 FINDING DATA SOURCES

The basic method used to obtain better photon production data for any particular isotope was quite simple; find the best possible sources of experimental data and evaluate them. Experimental papers were found through an exhaustive search process. First, extensive searches were performed using the Los Alamos National Laboratory library's web-based "SciSearch" program. All available years (1974 – 1997) were searched using many different search strategies. Second, the "Recent References" sections of all volumes of Nuclear Data Sheets from the present back to 1966 were combed for appropriate (n,γ) papers. Third, the Nuclear Science References section of the National Nuclear Data Center's (NNDC) online data service (see http://www.nndc.bnl.gov/ for a link to this service) was extensively searched. The NNDC online data service was also used to obtain photon production data listings from ENSDF,[10] the Evaluated Nuclear Structure Data File. The numerous references listed in the ENSDF retrievals were also obtained. Finally, each paper found was searched for additional references. Compilations of *elemental* data such as those by Orphan[4] and Lone[11] were also considered, but they were not very useful since we were interested in obtaining spectra for all stable isotopes of each element considered. For many elements, these two compilations were also found to be superseded by more recent experimental measurements.

2.2 DETERMINING THE BEST DATA SOURCE

Once the search process was completed, the papers were studied and carefully compared to determine the best source or sources of data. For our purposes, the ideal experiment is one that measures the complete spectrum of gamma-rays that are emitted after thermal-neutron capture in the nuclide of interest. It is very important to have as complete a spectrum as possible to ensure accurate gamma-ray intensities and heating numbers in MCNP for each incident neutron energy. All energy not carried away by secondary particle emission is deposited locally as heating.

In any experiment, the weakest gamma-rays will not be observed and gamma-rays from other nuclides may be incorrectly attributed to the nuclide of interest. Good equipment and experimental techniques can minimize these two problems. High-purity germanium (HPGe) detectors have over an order of magnitude better energy resolution than do sodium-iodide (NaI) detectors, allowing for the identification of many more weak gamma-rays. Compton suppression greatly reduces the background in gamma-ray measurements and also helps tremendously in measuring and identifying the source of low intensity gamma-rays. Longer experimental runs reduce statistical uncertainties and allow more accurate identification of weak gamma-rays. The use of multiple facilities (which have different background spectra) allows a more accurate background subtraction since only the gamma-rays of interest are usually dominant in both facility's spectra. Using methods such as these results in much improved spectral measurements. For each nuclide considered, the best sources of data were determined by considering the extent to which each of the above methods were used.

2.3 FORMING THE BEST SPECTRUM

Once the data sources were analyzed, all of the spectra were compared gamma-ray by gamma-ray. In general, if one source was clearly superior to the others (based on the analysis described in 2.2), its energies and intensities were adopted unless a majority of the other sources were in strong disagreement for a particular gamma-ray. For example, if there were 10 sources of data and nine agreed with each other but disagreed with the best data source, the average energy and/or intensity of the nine sources in agreement was adopted. Likewise, if all of the sources observed a gamma-ray not seen by the best source, the average was taken and the gamma-ray was included in the final spectrum. Gamma-rays observed only by the best source and not by other sources were included since the best source was generally able to measure more gamma-rays than the other sources.

When there was no clearly dominant data source, the process was more difficult. For each gamma-ray, if a majority of the sources were in agreement (in either energy and/or intensity) the average of the agreeing values was

adopted. If there was generally no agreement between sources but the gamma-ray in question was observed by most of the experimenters, the average of all measured values was adopted. If a gamma-ray was observed by less than half of the experimenters, it was not included in the adopted spectrum. Various numerical quantities were also defined to quantify the level of agreement between the experimental spectra. These quantities were useful in determining if one data set was in consistently poor agreement with the others, but in the end the final spectra were formed on a gamma-ray by gamma-ray basis.

Once an improved spectrum for a particular nuclide was produced, the yield was compared to the Q-value listed by Audi and Wapstra.[12] The spectrum yield (sum of each gamma-ray energy multiplied by its intensity) should equal the total energy available for neutron capture (the Q-value). Here the recoil energy of the target nuclide and the incident neutron energy (0.0253 eV) are neglected since they are so small. If the photon production spectrum is sufficiently complete (yield within about 10% of the Q-value), the intensities are normalized so that the yield equals the Q-value. This normalization ensures accurate heating numbers for MCNP. The normalization factor *NF* used to normalize the intensities is given by the following expression

$$NF = Q \Big/ \sum_i E_i \cdot I_i$$

where E_i is a gamma-ray energy, I_i is a gamma-ray intensity, and the sum i is over all gamma-rays in the thermal-neutron capture spectrum. Of the 17 spectra produced so far, the value of *NF* differed from 1.0 by more than 7% for only one nuclide (^{61}Ni). For ^{61}Ni, the experimental spectra were so incomplete that the unnormalized yield was only about 60% of the Q-value. In this case, normalizing the spectrum gives grossly inflated intensities and is a dangerous practice since the measured intensities may be correct even though the spectrum is incomplete. In cases like ^{61}Ni, which spectrum one uses (normalized or unnormalized) depends on what is more important for the application at hand – accurate gamma-ray intensities or correct heating. In this case, we decided to *not* normalize the ^{61}Ni spectrum to the Q-value because *NF* would have been very large (about 1.8). Since ^{61}Ni contributes only 0.61% to the spectrum of natural nickel, the incorrect heating from ^{61}Ni will have minimal impact on most calculations.

Each improved spectrum was incorporated into the corresponding ENDF evaluation and processed into MCNP data files using NJOY.[13] For a more detailed description of the methods used to generate the improved photon production spectra, see references 5 – 8.

3. STATUS AND SAMPLE RESULTS

To date, improved photon production spectra have been produced for all stable isotopes of chlorine, chromium, nickel, iron, and copper. A brief comparison of the improved spectra to their corresponding ENDF/B-VI spectra is given in Table 1. The number of gamma-rays and total yield of each spectrum is listed. The ratio of each spectrum's yield to the isotopic Q-value from Audi and Wapstra is also listed. Note that although most of the elements in Table 1 have isotopic ENDF/B-VI evaluations, the photon production spectra are often the same for each isotope, with only the total photon multiplicity being varied. Only an elemental comparison is given for chlorine because the ENDF/B-VI evaluation for chlorine is not isotopic. The elemental Q-value for chlorine was calculated by weighting each stable isotope's Q-value for thermal-neutron capture by its natural abundance and thermal-neutron capture cross-section, then summing over the stable isotopes. For an element with *n* stable isotopes, the elemental Q-value at thermal neutron energies is given by the expression

$$Q_{element} = \frac{\sum_{i=1}^{n} Q_i \cdot A_i \cdot \sigma_i}{\sum_{i=1}^{n} A_i \cdot \sigma_i}$$

where Q_i is the Q-value for radiative capture in isotope i, A_i is the abundance of isotope i, and σ_i is the thermal-neutron capture cross section for isotope i. For the ENDF files whose photon spectra are binned, the number of individual gamma-rays in the original spectra cannot be determined, but the yields can be obtained by integrating the

spectra. Except for ^{61}Ni, all of the new spectra were normalized to ensure their yields equaled the appropriate Q-value from Audi and Wapstra.

Each MCNP data file goes through a rigorous testing and validation process. As part of the testing process of the improved data files, a simple MCNP problem was set up, and comparisons between the standard ENDF and improved data were made. The simple problem consisted of a spherical void region of radius 1 cm with a thermal-energy (2.53×10^{-8} MeV) point neutron source at the center. The void region was surrounded by a 0.5 cm spherical shell of one of the materials in Table 1. If the natural density of the material was less than 5 g / cm^3, it was increased to 10 g / cm^3 so the run could be performed in a reasonable amount of time without significantly affecting the photon transport. An F4 tally (photon flux averaged over a volume) was then calculated over a thin void region surrounding the material. The photon flux was tallied from $E_\gamma = 0.0 - 10.0$ MeV and binned in 5 keV energy bins.

Table 1. Comparison of ENDF/B-VI and new photon production spectra

	ENDF/B-VI spectrum			New spectrum		
Isotope or element	Number of gamma-rays	Yield[a] (MeV)	Yield[a] / Audi[b]	Number of gamma-rays	Yield[a] (MeV)	Yield[a] / Audi[b]
Cl	31	6.243	0.73	482	8.573	1.00
^{50}Cr	Binned	9.249	1.00	71	9.262	1.00
^{52}Cr	Binned	7.940	1.00	11	7.939	1.00
^{53}Cr	Binned	9.674	1.00	89	9.719	1.00
^{54}Cr	Binned	6.232	1.00	85	6.246	1.00
^{58}Ni	Binned	9.000	1.00	243	9.000	1.00
^{60}Ni	Binned	7.814	1.00	142	7.820	1.00
^{61}Ni	Binned	10.601	1.00	77	5.912	0.56
^{62}Ni	Binned	6.839	1.00	92	6.838	1.00
^{64}Ni	Binned	6.098	1.00	33	6.098	1.00
^{54}Fe	Binned	9.298	1.00	42	9.298	1.00
^{56}Fe	Binned	7.800	1.02	252	7.646	1.00
^{57}Fe	Binned	10.044	1.00	99	10.044	1.00
^{58}Fe	Binned	6.581	1.00	139	6.581	1.00
^{63}Cu	Binned	7.916	1.00	322	7.916	1.00
^{65}Cu	Binned	7.068	1.00	424	7.066	1.00

[a]Sum of gamma-ray energies times their intensities.
[b]Q-value from Audi and Wapstra.[12]

The resulting photon flux for chlorine was calculated using ENDF60[14] data (based on release 2 of ENDF/B-VI and originally evaluated in 1967) and the new data with improved photon production. The flux calculated with the old data is shown in the top plot of Figure 2, and the flux from the new data is shown in the bottom plot of Figure 2. The new spectrum contains 482 gamma-rays compared to 31 in the old spectrum, which is a vast improvement. In addition to the new data having far more gamma-rays, there are several serious discrepancies between the two spectra. For example, the first gamma-ray in the ENDF spectrum is at 79 keV with an intensity of 20.17 photons per 100 neutron captures. No source of experimental data that we found for chlorine lists a gamma-ray near this energy and intensity. Since the intensity of this line is close to the intensity of the 786/788 keV doublet listed by many experimental sources, this is most likely a typographical error in the ENDF evaluation. Another example is the important 6108 keV gamma-ray, which has an intensity of 25.03 photons per 100 captures in the ENDF spectrum. In all of the experimental sources we found, this gamma-ray has an intensity of about 20 or 21 photons per 100 captures, a discrepancy of about 25%.

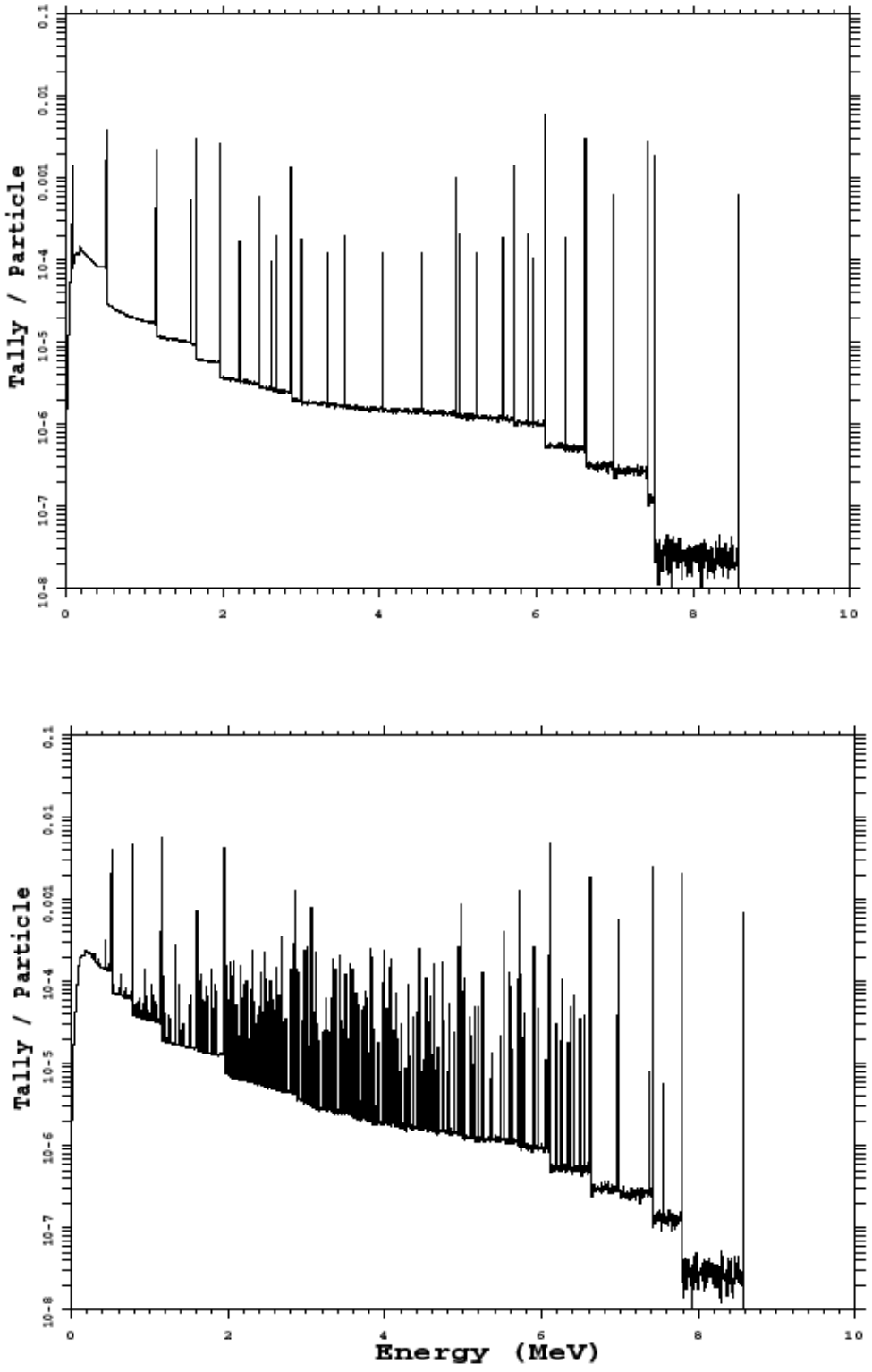

Fig. 2 Comparison of ENDF/B-VI and new photon production spectra for natural chlorine. The top plot is the ENDF/B-VI spectrum and the bottom plot is the new improved spectrum.

4. SUMMARY

MCNP is used in a wide variety of applications requiring high-quality photon production data. Recent work at Los Alamos has shown that the photon production data in the MCNP data libraries needed improvement before it could be successfully used in such applications. Work is underway to improve this aspect of the MCNP data libraries. Data files with improved thermal-neutron capture spectra have been produced for each stable isotope of chlorine, chromium, nickel, iron, and copper. The improved spectra are based on the best available experimental data. Except for ^{61}Ni, the yield of each spectrum was normalized to the appropriate Q-value listed by Audi and Wapstra. Similar improvements for additional elements such as aluminum, manganese, silicon, calcium, and vanadium are planned in the near future. A new MCNP data library with the improved photon data will be made available after our proprietary contracts with several well-logging companies expire, perhaps in late 1999.

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