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Developments in capture-γ libraries for nonproliferation applications

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Abstract. The neutron-capture reaction is fundamental for identifying and analyzing the γ-ray spectrum from an unknown assembly because it provides unambiguous information on the neutron-absorbing isotopes. Nondestructive-assay applications may exploit this phenomenon passively, for example, in the presence of spontaneous-fission neutrons, or actively where an external neutron source is used as a probe. There are known gaps in the Evaluated Nuclear Data File libraries corresponding to neutron-capture γ-ray data that otherwise limit transport-modeling applications. In this work, we describe how new thermal neutron-capture data are being used to improve information in the neutron-data libraries for isotopes relevant to nonproliferation applications. We address this problem by providing new experimentally-deduced partial and total neutron-capture reaction cross sections and then evaluate these data by comparison with statistical-model calculations.

1. Introduction

The principal aim of the capture-γ project is to add new γ-ray spectroscopic data (high-resolution HPGe-quality data) to the Evaluated Nuclear Data File (ENDF) [1] libraries for several high-priority isotopes [2] that will enhance transport-modeling applications. This project leverages heavily upon an existing atlas of data and targeted new capture-γ measurements at reactor facilities that were initiated as an International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) and led to the development of the Evaluated Gamma-ray Activation File (EGAF) [3].

High-resolution γ rays produced in neutron-capture reactions (and inelastic scattering) provide an unambiguous fingerprint of the isotopes within an unknown sample. Nondestructive-assay (NDA) applications may exploit this phenomenon using passive interrogation if spontaneous-fission neutrons are present, or active interrogation where an external neutron source is used to probe the sample. The brightest high-energy "primary" γ-ray transitions (i.e. those that originate at the neutron-capture state) from thermal-neutron capture ($E_\gamma \approx 4$–12 MeV) are often easily seen in spectra from unknown assemblies and clearly indicate the presence of Special Nuclear Materials (SNM: ^{233,235}U and Pu [4]), fission products and an array of materials frequently associated with SNM. Strong secondary γ rays also produce reliable fingerprints. However, NDA applications depend on accurate data and there are well-known gaps in the neutron-capture γ-ray line data in the ENDF libraries that limit these capabilities. In particular, the ENDF libraries are lacking high-energy capture-γ lines for the actinides, although there are widespread problems elsewhere also. For example, the neutron-capture γ-ray spectrum for natural lead contains several high-energy primaries as shown in Fig. 1. However, although the simulations of the setup described in Fig. 2 with the Monte Carlo neutron-transport (MCNP) code [5] using the ENDF/B-V library for natural lead appears to represent the real spectrum reasonably well, much of the information on primary transitions is missing when the *more recent* ENDF/B-VII library, in which lead is separated into individual isotopes, is used. Simulations of purely thermal neutron capture show similar discrepancies. Neutron-capture γ-ray simulations utilizing the ENDF/B-VII library for this important shielding material will, therefore, clearly be at odds with the real spectrum (Fig. 1). An ENDF/B GForge bug-tracker item has been submitted

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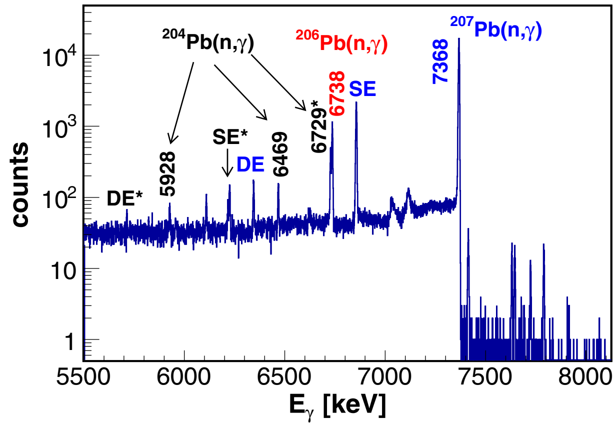

Figure 1. Spectrum of prompt γ rays with $E_\gamma \gtrsim 5500$ keV from a $^{\rm nat}$Pb(n,γ) measurement using thermal neutrons carried out at the Budapest Research Reactor [7]. The spectrum corresponds to an irradiation period of 50.9 h and reveals many prominent primary γ lines in naturally-occurring isotopes of lead. The γ-rays are color-coded according to the isotope of origin. Single-escape (SE) and double-escape (DE) peaks are also labelled and identified according to their parent γ rays.

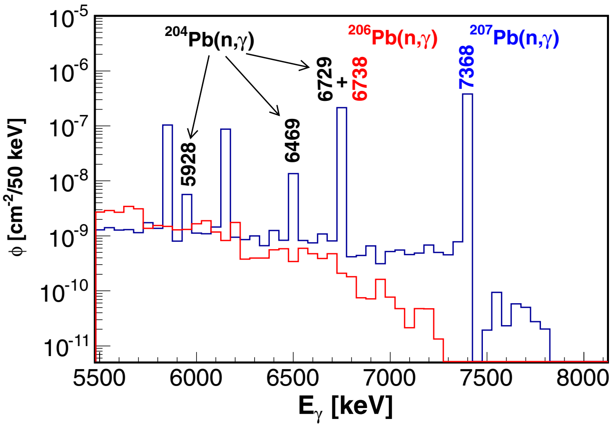

Figure 2. MCNP-simulated neutron-capture γ-ray spectra for the $^{\rm nat}$Pb(n,γ) reaction generated using ENDF-B/V (blue histograms) and ENDF-B/VII (red histograms), showing clear discrepancies. The spectra are expanded above $E_\gamma \gtrsim 5500$ keV where strong primary γ rays are expected to dominate. Each simulation assumes a natural lead sphere of radius 30 cm with a centrally-located source covering a neutron-energy range from 10 keV to 1 MeV, with 99% of the neutrons below 450 keV. The flux is calculated across a spherical surface at a distance 10 cm from the neutron-source location.

concerning this particular issue [6]. The capture-γ project represents an ongoing effort to counter known deficiencies for a wide variety of isotopes.

2. Technical approach

The aim of this project is to augment the ENDF libraries with new and improved γ-ray spectroscopic line data. Here, we briefly outline the methodology underpinning this process:

- Partial γ-ray production cross sections (σ_γ) for a particular isotope, selected according to a high-priority list [2], are measured at the 10-MW Budapest Research Reactor [7] or sourced directly from EGAF.
- For heavy nuclei, these σ_γ data are validated by comparison with theoretical predictions using the statistical model for γ decay (DICEBOX [8]) to calculate a system of partial widths for a series of γ cascades:

$$\langle \Gamma_{if}^{XL} \rangle = \frac{f^{(XL)}(E_\gamma) \cdot E_\gamma^{2L+1}}{\rho(E_i, J_i, \pi_i)}. \qquad (1)$$

  Here, $\rho(E_i, J_i, \pi_i)$ is the level density at an initial excitation energy E_i characterized with a spin-parity $J_i^{\pi_i}$, $f^{(XL)}$ is the photon strength function for a multipole of order L where X denotes electric (E) or magnetic (M) character of a transition, and E_γ is the γ-ray energy. However, for light low-Z nuclei, a nonstatistical approach may be adopted to a good approximation.
- The validated σ_γ data are then processed into the correct format for incorporation into ENDF and correlations with other sections of the library are verified. The discrete γ rays are stored in File 12 (MF12 MT102) of the relevant ENDF library and the calculated quasicontinuum, stored in File 15 (MF15 MT102), is scaled to achieve agreement with the total thermal neutron-capture cross section σ_0 in File 3 (MF3 MT102).
- The Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) validation and verification codes (e.g., PREPRO [9], NJOY [10], and FUDGE [11]) are then used to check the integrity of the new ENDF library.
- After generating successful transport-simulation output (e.g., the MCNP simulation presented in Fig. 3), the libraries are then sent to the National Nuclear Data Center (NNDC) at the Brookhaven National Laboratory (BNL) for further testing and ultimately disseminated in the next ENDF/B-VIII.0 [12] release.

3. Improving the ENDF libraries

Missing or problematic data are frequently encountered in two distinct regions of the neutron-capture γ-ray spectrum: (i) at high energy where $E_\gamma \gtrsim 3$ MeV; (ii) at low energy where $E_\gamma \lesssim 100$ keV. For example, our work on tungsten [13–15] and rhenium [16], in particular, highlights both of these issues. Primary γ rays were identified for the first time in the ^{180}W(n,γ) measurement [14], while 50 new primaries were assigned to the ^{186}Re decay scheme via the ^{185}Re(n,γ) measurement [16]. Because the high-energy regime of the capture-γ spectrum can be delineated and understood completely, e.g., see Refs. [14, 16], it provides enormous benefit as an auxiliary forensics tool. Our enriched-sample tungsten and rhenium measurements, both high-Z high-ρ materials, also demonstrated significant γ-ray attenuation that is at odds with the existing partial γ-ray production cross-section data for certain transitions [17]. We developed an analytical procedure [15], now tried [13] and tested [16], to correct for this effect. This problem also highlights potential concerns over the existing low-energy capture-γ data for other high-density materials that may be used to supply the ENDF library.

Throughout the course of the capture-γ project, new and improved neutron-capture γ-ray line data were

Table 1. ENDF/B-VIII.beta2 [12] libraries updated with new evaluated capture-γ lines. The final column shows the total radiative thermal neutron-capture cross section (σ_0) in File 3.

Element	A	Abundance [%]	(n,γ) lines	σ_0 [b]
Li	6	7.589	3	0.04
Li	7	92.411	3	0.05
B	11	80.18	13	0.01
F	19	100	166	0.0096
Na	23	100	244	0.53
Al	27	100	290	0.23
Si	28	92.2297	56	0.18
Cl	35	75.771	382	45.55
Cl	37	24.229	76	0.43

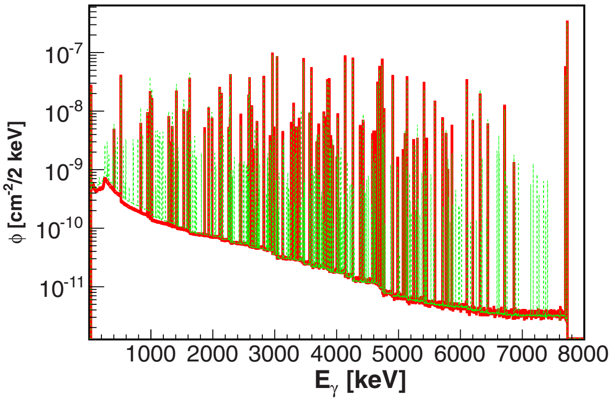

Figure 3. Gamma-ray spectra illustrating differences between the new ^{27}Al(n,γ) data (green histograms) and the previous data (red histograms) in the ENDF/B-VII Rev: 532 library. These MCNP spectra are calculated assuming a thermal-neutron beam incident upon a thin ^{27}Al target viewed by high-purity germanium (HPGe) detectors in a 4π arrangement.

incorporated into and used to update nine ENDF libraries. These libraries, listed in Table 1, have passed the rigorous testing requirements described in Sect. 2 including the successful generation of representative MCNP output, and are now available in the repository for ENDF/B-VIII.beta2 [12], and onward ENDF/B-VIII releases, via the NNDC at BNL. The high-resolution data in these libraries are needed for accurate simulations of interrogation systems. For illustrative purposes, the new partial γ-ray production cross-section data for the ^{27}Al(n,γ) reaction with thermal neutrons reveals a marked improvement over the previous data in the ENDF library (ENDF/B-VII Rev: 532), as shown in Fig. 3. The superior quality of the new data reinforces their utility in simulations allowing for accurate inferences in NDA applications through provision of enhanced isotopic-identification fingerprints.

Although the principal goal of this work is the development of a new ENDF library, the individual capture-γ measurements are also published as stand-alone projects in their own right. Many of the additional (n,γ) measurements undertaken as part of the capture-γ project that have not yet been incorporated into a new ENDF library are listed in Table 2, together with published measurements that have contributed towards an ENDF upgrade. Furthermore, the adopted statistical-model analyses, an integral component to the validation of the measured capture-γ data for the heavy isotopes ($A \geq 39$), provide improved decay-scheme information including nuclear structure inferences. For example, the statistical model has been used to deduce spin-parity (J^π) assignments for many of the excited states in ^{187}W [13] and ^{186}Re [16]. These decay-scheme improvements will later be accounted for in subsequent mass-chain and nuclide evaluations for the Evaluated Nuclear Structure Data File (ENSDF) [28]. Because of the inherent synergy between different libraries in the nuclear data pipeline, any improvements to the ENSDF database will ultimately be reformatted into the Reference Input Parameter Library (RIPL) [29], a derived database that also provides the source decay-scheme information for ENDF.

Table 2. List of published (n,γ) studies carried out at the Budapest Research Reactor as part of the capture-γ project. For all isotopes with $A \geq 39$ the measured partial γ-ray cross sections are validated against statistical-model analysis. The ^{6,7}Li(n,γ), ^{9}Be(n,γ), ^{10,11}B(n,γ) [19] and ^{23}Na(n,γ) [20] measurements have been used to upgrade ENDF [12].

Measurement	Reference
Methodology	Hurst [15]
^{2}H(n,γ), ^{16,17,18}O(n,γ)	Firestone [18]
^{6,7}Li, ^{9}Be, ^{10,11}B, ^{12,13}C, ^{14,15}N(n,γ)	Firestone [19]
^{23}Na(n,γ)	Firestone [20]
^{35}Cl(n,γ)	Molnár [21]
^{39,40,41}K(n,γ)	Firestone [22]
^{93}Nb(n,γ)	Turkoglu [23]
^{102,104,105,106,108,110}Pd(n,γ)	Krtička [24]
^{152,154}Eu(n,γ)	Basunia [25]
^{155,157}Gd(n,γ)	Choi [26]
^{180}W(n,γ)	Hurst [14]
^{182,183,184,186}W(n,γ)	Hurst [13]
^{185}Re(n,γ)	Matters [16]
^{242}Pu(n,γ)	Rossbach [27]

4. Conclusion

New and improved neutron-capture γ-ray line data were used to upgrade nine ENDF libraries: ^{6,7}Li, ^{11}B, ^{19}F, ^{23}Na, ^{27}Al, ^{28}Si, and ^{35,37}Cl. These libraries contain improved γ-ray spectroscopic line data necessary for simulations of interrogation systems. The nine libraries submitted to the NNDC at BNL satisfy all testing requirements and are available in the ENDF/B-VIII.beta2 release [12] (and beyond). This work has also led to several high-impact peer-reviewed publications and provided source material for graduate theses. In the future, ENDF libraries for all isotopes listed in Table 2, as well as for many other isotopes on the priority list [2], are planned to be upgraded. In turn, these improved capture-γ libraries will benefit nonproliferation applications based on screening technologies.

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