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Developments in capture-$\gamma$ libraries for nonproliferation applications

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Abstract. The neutron-capture reaction is fundamental for identifying and analyzing the $\gamma$-ray spectrum from an unknown assembly because it provides unambiguous information on the neutron-absorbing isotopes. Nondestructive-assay (NDA) 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 $\gamma$-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-$\gamma$ project is to add new $\gamma$-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 will enhance 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.

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where strong primary $\gamma$ from the neutron-source location. flux is calculated across a spherical surface at a distance 10 cm to 1 MeV, with 99% of the neutrons below 450 keV. The simulation assumes a natural lead sphere of radius 30 cm with a centrally-located source covering a neutron-energy range from $\gamma$-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 $\gamma$ rays.

concerning this particular issue [6]. The capture-$\gamma$ 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 $\gamma$-ray spectroscopic line data. Here, we briefly outline the methodology underpinning this process:

- Partial $\gamma$-ray production cross sections ($\sigma_\gamma$) 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 $\sigma_\gamma$ data are validated by comparison with theoretical predictions using the statistical model for $\gamma$ decay (DICEBOX [8]) to calculate a system of partial widths for a series of $\gamma$ cascades:

$$\langle \Gamma_{\nu}^{XL} \rangle = \frac{f^{(X)}(E_\gamma) \cdot E_{\nu}^{2L+1}}{\rho(E_\gamma, J_\nu, \pi_\nu)}$$

Here, $\rho(E_\gamma, J_\nu, \pi_\nu)$ is the level density at an initial excitation energy $E_\gamma$ characterized with a spin-parity $J^n_\nu$. $f^{(X)}$ is the photon strength function for a multipole of order $L$ where $X$ denotes electric ($E$) or magnetic ($M$) character of a transition, and $E_\nu$ is the $\gamma$-ray energy. However, for light low-Z nuclei, a nonstatistical approach may be adopted to a good approximation.

- The validated $\sigma_\gamma$ data are then processed into the correct format for incorporation into ENDF and correlations with other sections of the library are verified. The discrete $\gamma$ rays are stored in File 12 (MF12 MT102) of the relevant ENDF library and the calculated quasimonochromatic, stored in File 15 (MF15 MT102), is scaled to achieve agreement with the total thermal neutron-capture cross section $\sigma_0$ 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 $\gamma$-ray spectrum: (i) at high energy where $E_\gamma \gtrsim 3$ MeV; (ii) at low energy where $E_\nu \lesssim 100$ keV. For example, our work on tungsten [13–15] and rhenium [16], in particular, highlights both of these issues. Primary $\gamma$ rays were identified for the first time in the $^{186}$W(n,$\gamma$) measurement [14], while 50 new primaries were assigned to the $^{186}$Re decay scheme via the $^{186}$Re(n,$\gamma$) measurement [16]. Because the high-energy regime of the capture-$\gamma$ 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-$\rho$ materials, also demonstrated significant $\gamma$-ray attenuation that is at odds with the existing partial $\gamma$-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-$\gamma$ data for other high-density materials that may be used to supply the ENDF library.

Throughout the course of the capture-$\gamma$ project, new and improved neutron-capture $\gamma$-ray line data were
The new ENDF library are listed in Table 2, together with project that have not yet been incorporated into a γ-enhanced isotopic-identification fingerprints. Inferences in NDA applications through provision of shown in Fig. 3. The superior quality of the new data neutron reveals a marked improvement over the previous model analyses, an integral component to the validation of the measured capture-γ cross-section data for the 27Al(γ,γ)-ray production reaction with thermal neutrons. These MCNP histograms) in the ENDF/B-VII Rev: 532 library. These MCNP source decay-scheme information for ENDF. (RIPL) [29], a derived database that also provides the reformatted into the Reference Input Parameter Library (RIPL) [29], a derived database that also provides the source decay-scheme information for ENDF.

### 4. Conclusion

New and improved neutron-capture γ-ray line data were used to upgrade nine ENDF libraries: 4.7Li, 11B, 23Fe, 23Na, 27Al, 28Si, and 35,37Cl. 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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