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Neutron Capture Gamma-Ray Libraries for Nuclear Applications*

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Abstract. The neutron capture reaction is useful in identifying and analyzing the gamma-ray spectrum from an unknown assembly as it gives unambiguous information on its composition. This can be done passively or actively where an external neutron source is used to probe an unknown assembly. There are known capture gamma-ray data gaps in the ENDF libraries used by transport codes for various nuclear applications. The Evaluated Gamma-ray Activation file (EGAF) is a new thermal neutron capture database of discrete line spectra and cross sections for over 260 isotopes that was developed as part of an IAEA Coordinated Research Project. EGAF is being used to improve the capture gamma production in ENDF libraries. For medium to heavy nuclei the quasi continuum contribution to the gamma cascades is not experimentally resolved. The continuum contains up to 90% of all the decay energy and is modeled here with the statistical nuclear structure code DICEBOX. This code also provides a consistency check of the level scheme nuclear structure evaluation. The calculated continuum is of sufficient accuracy to include in the ENDF libraries. This analysis also determines new total thermal capture cross sections and provides an improved RIPL database. For higher energy neutron capture there is less experimental data available making benchmarking of the modeling codes more difficult. We are investigating the capture spectra from higher energy neutrons experimentally using surrogate reactions and modeling this with Hauser-Feshbach codes. This can then be used to benchmark CASINO, a version of DICEBOX modified for neutron capture at higher energy. This can be used to simulate spectra from neutron capture at incident neutron energies up to 20 MeV to improve the gamma-ray spectrum in neutron data libraries used for transport modeling of unknown assemblies.

Keywords: Nuclear Data, ENDF, Neutron Capture Gammas

PACS: 21.10.-k, 21.10.Hw, 21.10.Ma, 21.10.Pc, 21.10.Tg, 21.60.-n, 21.60.Ka, 23.20.Lv, 24.10.Pa, 24.87.+y, 28.20.Fc, 28.20.Np

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INTRODUCTION

Prompt gamma-rays emitted following neutron capture provide unique elemental signatures that provide unambiguous information on the composition of unknown assemblies. For many years no reliable prompt gamma-ray database existed for this kind of analysis. Recently new, precise, prompt gamma-ray measurements have been made with the guided thermal neutron beam at the Budapest Reactor on all elemental targets with $Z=1-83$, 90, 92 with the exception of helium and promethium. These data have been evaluated and published as part of an IAEA Coordinated Research Project [1].

For light isotopes the neutron capture decay scheme is nearly complete, but for medium and heavy isotopes considerable unresolved continuum gamma-ray feeding exists. This feeding can be accounted for using the statistical model code DICEBOX [2]. These calculations can also be used to determine the total radiative thermal neutron capture cross sections σ_0 , capture state spin distributions, and improved nuclear structure information from the EGAF data.

Relatively little experimental data are available for prompt gamma-ray emission following higher energy neutron capture. We are investigating this using surrogate reactions [3] which produce the same compound nucleus at higher excitation energies. A Hauser Feshbach analysis can then be used to benchmark the CASINO version of DICEBOX [4]. We are evaluating the EGAF data using the DICEBOX and CASINO codes to develop a comprehensive ENDF capture gamma-ray library for use in transport modelling of unknown assemblies.

DATA

Experimental Measurements

Neutron capture gamma-ray cross sections were measured with the guided neutron beam at the 10-MW Budapest Reactor [5]. The target station is located ≈ 30 m from the reactor where both primary and secondary gamma-rays can be measured in low background conditions. Neutrons enter the evacuated target holder and continue to the beam stop at the rear wall of the guide hall. The thermal-equivalent neutron flux was $2 \times 10^6 \text{ n cm}^{-2} \text{ s}^{-1}$.

Prompt gamma-rays from the target were measured with an n-type high-purity, 25% efficient (HPGe) detector with closed-end coaxial geometry located 23.5 cm from the target. The detector is Compton-suppressed by a BGO-scintillator guard detector annulus surrounded by 10-cm thick lead

shielding. Counting efficiency was calibrated from 50 keV to 10 MeV with radioactive sources and (n,γ) reaction gamma rays to a precision of better than 1% from 500 keV to 6 MeV and better than 3% at all other energies [7]. The gamma-ray spectra were analyzed using the Hypermet PC program [7,8].

Elemental radiative thermal neutron gamma-ray cross sections were calibrated using natural elemental targets of stoichiometric compounds or homogenous mixtures containing elements with well known cross sections e.g. H, N, Cl, S, Na, Ti, or Au [8]. The guided neutron beam used in these measurements is a purely thermal so no correction was necessary for epithermal contributions to the measured gamma-ray cross sections. The gamma-rays were assigned to the isotopic level scheme on the basis of energy and intensity by comparison with data from the ENSDF file [9]. Elemental gamma-ray cross sections were measured and isotopic cross sections determined assuming the normal abundances [10].

EGAF Thermal Database

Until recently there was no reliable neutron capture gamma-ray database although these data are also an important component of the Evaluated Neutron Data File (ENDF) used in neutron transport calculations. Groshev *et al* [11] published the first compilation of prompt capture γ -ray energies and intensities, and Greenwood *et al* [12] the first spectrum catalog during the era of NaI detectors. With the advent of Ge detectors in the 1960s, Rasmussen [13] and Orphan [14] measured capture gamma-ray spectra for all elements. These data were compiled by Lone *et al* [15] who published a database of over 10,000 gamma-rays in 1981. This database was used for many years despite the inadequacies inherent to those early measurements. Prompt neutron capture γ -ray data are also compiled from the literature in the Evaluated Nuclear Structure Data File (ENSDF) [9]. These data were used primarily to extract nuclear structure information and were not evaluated for applied use. Reedy and Frankel [16] carefully re-evaluated the literature for light elements from hydrogen to zinc and provided this information in ENSDF format. The Lone *et al*, ENSDF, and Reedy gamma-ray intensities are normalized to units of per 100 neutron captures. In the 1990's new capture gamma-ray measurements were performed for all stable elements by Molnar *et al* at the Budapest Reactor. An International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) was organized to evaluate these data. The IAEA CRP produced a database of $\approx 35,000$ neutron capture γ -rays [17] which was named the Evaluated Gamma-ray

STATISTICAL MODEL CALCULATIONS

Theoretical feedings of low-lying levels by thermal neutron radiative capture were calculated using the computer code DICEBOX [2]. The algorithm of this Monte Carlo code is based on the generalization of the extreme statistical model embodying Bohr's idea of a compound nucleus [18]. Below a certain critical energy, E_{crit} , the level scheme, i.e. energies, spins and parities of all levels as well as all de-exciting transitions, is taken from experiment. Above E_{crit} , a set of levels is generated as a random discretization of an *a priori* known level density formula $\rho(E, J^\pi)$. Decay properties of an initial level i above E_{crit} are completely characterized by a full set of partial radiation widths to all final levels f below the level i . A partial radiative width, Γ_{if} , which characterizes the probability of gamma-ray decay with an energy $E_\gamma = E_i - E_f$ is assumed to be a random choice from the Porter-Thomas distribution [19] with a mean value

$$\langle \Gamma_{if} \rangle = \frac{f^{(XL)}(E_\gamma, \xi) \times E_\gamma^3}{\rho(E_i, J_i^\pi)}$$

Here $\rho(E_i, J_i^\pi)$ is the level density near the initial level i and $f^{(XL)}(E_\gamma, \xi)$ is the photon strength function (PSF) for a transition of given type X and multipolarity L . The argument ξ of the PSF represents possible dependence on quantities other than gamma-ray energy. In the extreme statistical model it is assumed that the Γ_{if} are uncorrelated. Selection rules for different types of transitions are fully accounted for in the generation of Γ_{if} .

The random generation of a system of all Γ_{if} which fully describe the decay properties of the nuclear levels is called a *nuclear realization*. Due to fluctuations involved there exist an almost infinite number of nuclear realizations, that differ in decay properties even for a single choice of $f^{(XL)}$ and level density. Consequently all simulated quantities are subject to statistical fluctuations arising from different nuclear realizations. Determination of these fluctuations with the DICEBOX code allows us to estimate the uncertainty coming from the statistical nature of the decay process. Typically a calculation consists of 50 nuclear realizations, each with 50000 capture state decays, generated by the Monte Carlo method. DICEBOX stores the simulated capture state

deexcitation data which are used to calculate populations of low-lying levels below E_{crit} and intensities of all gamma rays per neutron capture. Simulated quantities can be renormalized to absolute cross sections by comparison with the experimental gamma-ray cross sections depopulating low-lying levels.

Energy dependence and absolute values of $f^{(XL)}(E_\gamma, \xi)$ are not sufficiently known for nuclei with $A < 50$. We therefore have used only simple models for these quantities in our simulations. For $E1$ transitions we adopted three different models: (i) The Lorentzian shape of Giant Dipole Electric Resonance (GDER) [20] in conjunction with the Brink hypothesis [21], also called the Standard Lorentzian (SLO) model, (ii) the KMF model [22], which modifies the shape of low-energy tail of GDER, and (iii) the single-particle (SP) model [23] where $f^{(E1)} = \text{const.}$ is independent of gamma-ray energy. Parameters of GDER can be found in the survey of Dietrich and Berman [24].

For $M1$ strength two different models were used: (i) the single-particle SP model where $f^{(M1)} = \text{const.}$, and (ii) a model where $f^{(M1)}$ is described as Lorentzian-shaped resonance Giant Dipole Magnetic resonance (GDMR) at about 12 MeV with a width about 3 MeV [25]. The strengths of $f^{(M1)}$ were adjusted to match the observed population of positive and negative parity levels at low excitation energies. For $E2$ strength a single-particle SP model was used.

The Back-Shifted Fermi Gas (BSFG) and Constant-Temperature (CT) models, in the parameterization from von Egidy, Schmidt, and Bekhami [26], were used for level density. All possible combinations of $f^{(XL)}$ and level density were tested in these simulations.

RESULTS

Low-Z Isotope Analysis

For low-Z isotopes the EGAF prompt gamma-ray data are usually complete. For example the $^{12}\text{C}(n, \gamma)^{13}\text{C}$ decay scheme is shown in Fig. 1. All six gamma-rays from neutron capture are observed and the cross section balance through the decay scheme is excellent with 3.86(6) mb deexciting the capture state and 3.85(6) mb populating the ground state. This value is significantly larger than 3.53(7) mb in ENDF, adopted by a measurement by Jurney et al [26], but consistent with other values given in Table 1. The Jurney et al measurement of the ^2H total radiative thermal neutron cross section, $\sigma_0 = 0.508(15)$ mb, was also measured in [26] using the lower ^{12}C cross section

as a standard. As shown in Table 1 renormalizing this cross section gives a new value, $\sigma_0=0.552(16)$ mb, also consistent with other values.

FIGURE 1. $^{12}\text{C}(n,\gamma)^{13}\text{C}$ thermal neutron capture decay scheme.

Table 1. Comparison previous measurements of ^2H and ^{12}C total radiative thermal neutron cross sections with EGAF results.

Author (Year)	$\sigma_0(\text{mb})$	Author (Year)	$\sigma_0(\text{mb})$
Prestwich(1981)	3.50 ± 0.16	Trail (1964)	0.36 ± 0.03
Jurney (1982)	3.53 ± 0.07	Alfimenkov(1980)	0.476 ± 0.020
Nichols (1960)	3.57 ± 0.03	Jurney (1982)	0.508 ± 0.015
Sagot (1963)	3.72 ± 0.15	Merritt (1968)	0.521 ± 0.009
Jurney (1963)	3.8 ± 0.4	Silk (1969)	0.523 ± 0.029
Starr (1962)	3.83 ± 0.06	Ishikawa(1973)	0.55 ± 0.01
Hennig (1967)	3.85 ± 0.15	Kaplan (1952)	0.57 ± 0.01
Matsue (2004)	3.81 ± 0.11	Jurney (1963)	0.60 ± 0.05
EGAF (2007)	3.90 ± 0.06	Sargent (1947)	0.92 ± 0.22
ENDF	3.53 ± 0.07	ENDF	0.508 ± 0.015

High-Z Isotope Analysis

For high-Z isotopes the neutron capture gamma-ray decay scheme is usually too complex to resolve all transitions. This is because as the level density becomes large level separations become smaller than the intrinsic resolution of HPGe detectors resulting in a high density, unresolved continuum of transitions. Often a large fraction of the neutron capture cross

section will still populate the ground state through secondary transitions. Observed primary transitions may be much weaker, especially if the neutron separation energy S_n is large, in which case most of the primary transitions will populate levels near S_n where the level density is very high. An example is $^{105}\text{Pd}(n,\gamma)^{106}\text{Pd}$ where the neutron capture decay cross section balance is shown in Table 2 [27]. More than 20 b are observed feeding the ground state of ^{106}Pd , nearly the 21.0(15) b cross section recommended by Mughabghab [28]. By comparison only 0.55 b are observed in primary transitions deexciting the 9561-keV capture state.

Table 2. Cross section balance for $^{105}\text{Pd}(n,\gamma)^{106}\text{Pd}$

E(level)	J ^π	$\Sigma\sigma_i(\text{in})$	$\Sigma\sigma_i(\text{out})$	$\Delta\Sigma\sigma$
0	0+	20.26		
511.844	2+	13.88	17.91	4.03
1128.04	2+	2.371	4.263	1.892
1133.79	0+	0.227	0.565	0.338
1229.2	4+	1.630	3.479	1.849
1557.67	3+	1.183	2.142	0.959
1562.16	2+	0.312	1.860	1.557
1706.44	0+	0.012	0.193	0.181
1909.39	2+	0.063	0.724	0.661
1932.37	4+	0.217	0.590	0.373
2001.56	0+	0.029	0.118	0.089
2077.1	6+	0.001	0.103	0.102
2077.37	(4)+	0.057	0.440	0.383
2084.39	-3	0.123	1.033	0.910
2242.4	2+	0.026	0.499	0.473
2278.47	0+	0	0.056	0.056
2282.89	4+	0.0007	0.275	0.274
2306.01	-3	0.053	0.542	0.489
2308.73	2+	0.000	0.283	0.283
2350.96	4+	0.018	0.304	0.286
2366.09	5+	0.003	0.116	0.114
2397.37	(5)-	0.055	0.263	0.209
2401	(2-,3-)	0.037	0.300	0.263
2430.11	2+	0.065	0.293	0.227
2472.09	0+	0.000	0.055	0.055
2484.76	(1-)	0.043	0.253	0.211
2500.01	-2	0.028	0.296	0.267
2578.64	(4-)	0.00004	0.221	0.221
...	...	...	...	...
...	...	...	...	...
9561.4	2+,3+		0.554	

Fortunately, we can calculate the missing continuum intensity with the statistical model code DICEBOX assuming only average properties for the continuum of transitions. Level density and photon strength parameters can be determined by comparisons with photomuclear data and experimental capture state widths. The quality of the analysis can be determined by comparing the observed transition cross section deexciting levels below E_{crit} with the calculated cross section populating these levels as shown in Fig.2. This fit can be optimized to determine the admixture of

spins in the neutron capture state.

Figure 2. Population/depopulation plot for $^{105}\text{Pd}(n,\gamma)^{106}\text{Pd}$.

This statistical analysis can also be used to improve the nuclear structure data at low E_{crit} . When a poor fit is observed to a level in the population/depopulation plot it is likely that there is a problem with the J^π of that level. An example was found in $^{184}\text{W}(n,\gamma)^{185}\text{W}$, Fig. 3, where the 332-keV level was found to be more consistent with $9/2$ than $(7/2)$ recommended in ENSDF [9]. In addition, nine new transitions were placed in the ^{185}W level scheme from the EGAF data.

Figure 3. ^{185}W level scheme determined using EGAF data with DICEBOX statistical model calculations. Two new levels and nine new transitions (red) were determined.

Higher Energy Neutrons

ENDF libraries require capture cross sections for neutron energies to 20 MeV as a minimum. As there is a paucity of experimental data for capture gamma spectra above thermal energies, we are investigating the use of surrogate reactions [3]. These can start gamma cascades above (and below) the neutron separation energy in the same compound nucleus as that in thermal capture. Figure 4 shows Hauser Feshbach calculated ground band transition probabilities vs. excitation energy from the $^{156}\text{Gd}(p,p'\gamma)^{156}\text{Gd}$ surrogate reaction to $^{155}\text{Gd}(n,\gamma)^{156}\text{Gd}$. The band transitions (which are measured) are shown for two schematic spin distributions (not measured) and will be fitted to the experimental data. If the experimental spectra are accurately predicted via this Hauser Feshbach nuclear reaction model, far better knowledge of the cascades from higher energy neutrons is obtained. Accordingly the CASINO version of DICEBOX can be benchmarked in this way also.

Figure 4. Calculated ground band transition probabilities for the $^{156}\text{Gd}(p,p'\gamma)^{156}\text{Gd}$ surrogate reaction. The 2 plots have different spin parity distributions. $S_n=8.54$ MeV

CONCLUSIONS

The use of EGAF data combined with statistical model calculations using DICEBOX provides a powerful method for improving the capture gamma-ray data in ENDF. A comparison of the original ENDF library data for $^{104}\text{Pd}(n,\gamma)^{105}\text{Pd}$ without new, EGAF based library is shown in Fig. 5. Although the broad resolution of the original ENDF library is similar to the new library, narrow line information in the new library makes this far more useful for the analysis of unknown assemblies. Table 3 provides a summary of the completed beta libraries to date.

Figure 5. Comparison of the original $^{104}\text{Pd}(n,\gamma)^{105}\text{Pd}$ ENDF library with the new EGAF-based library.

Table 3. EGAF capture gamma-ray library completion status.

Z	El	A	%NA	barns	N _γ
1	H	1	99.9844	0.33260	1
1	D	2	0.01557	0.00052	1
3	Li	6	7.589	0.04000	3
3	Li	7	92.411	0.05000	3
4	Be	9	100	0.01000	12
5	B	10	19.82	0.50000	9
5	B	11	80.18	0.01000	9
6	C	12	98.892	0.00353	6
7	N	14	99.6337	0.08000	60
8	O	16	99.7628	0.00019	4
9	F	19	100	0.00960	1622
11	Na	23	100	0.53000	233
12	Mg	24		0.06000	283
13	Al	27	100	0.23000	291
14	Si	28	92.2297	0.18000	54
15	P	31	100	0.17000	202
16	S	32		0.53000	470
17	Cl	35	75.771	45.55000	383
17	Cl	37	24.229	0.43000	77
26	Fe	56	91.75	2.59000	193
46	Pd	104	11.14	0.60000	13
74	W	182	26.4985	19.90000	126
74	W	183	14.3136	10.30000	212
74	W	184	30.6422	1.70000	64
74	W	186	28.4259	38.50000	152
82	Pb	207	22.0827	0.62500	25

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