The Evaluated Gamma-ray Activation File (EGAF)

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Abstract. The Evaluated Gamma-ray Activation File (EGAF), a new database of prompt and delayed neutron capture γ ray cross sections, has been prepared as part of an International Atomic Energy Agency (IAEA) Coordinated Research Project to develop a "Database of Prompt Gamma-rays from Slow Neutron Capture for Elemental Analysis". Recent elemental γ-ray cross-section measurements performed with the guided neutron beam at the Budapest Reactor have been combined with data from the literature to produce the EGAF database. EGAF contains thermal cross sections for ≈35,000 prompt and delayed γ-rays from 262 isotopes. New precise total thermal radiative cross sections have been derived for many isotopes from the primary and secondary gamma-ray cross sections and additional level scheme data. An IAEA TECDOC describing the EGAF evaluation and tabulating the most prominent γ-rays will be published in 2004. The TECDOC will include a CD-ROM containing the EGAF database in both ENSDF and tabular formats with an interactive viewer for searching and displaying the data. The Isotopes Project, Lawrence Berkeley National Laboratory continues to maintain and update the EGAF file. These data are available on the Internet from both the IAEA and Isotopes Project websites.

1. INTRODUCTION

Low-energy neutron capture produces prompt γ-rays unique to each isotope that can be exploited in Prompt Gamma-ray Activation Analysis (PGAA) for nondestructive elemental analysis. Until now, PGAA has been severely limited by the lack of a reliable neutron capture γ-ray database. These data are also an important component of the Evaluated Neutron Data File (ENDF) used in neutron transport calculations.

Groshev *et al*[1] published the first compilation of prompt capture γ-ray energies and intensities, and Greenwood *et al*[2] the first spectrum catalog during the era of NaI detectors. With the advent of Ge detectors in the 1960s, Rasmussen[3] and Orphan[4] measured capture γ-ray spectra for all elements. These data were compiled by Lone *et al*[5] who published a database of over 10,000 γ-rays in 1981. This database has been 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)[6]. These data were used primarily to extract nuclear structure information and were not evaluated for applied use. Reedy and Frankel[7] 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 γ-ray intensities are normalized to units of per 100 neutron captures.

In the 1990's new capture γ-ray measurements were performed for all stable elements by Molnar *et al* at the Budapest Reactor. These measurements utilized a guided neutron beam with the target station far from the reactor where both primary and secondary γ-rays could be measured under low background conditions. They measured absolute γ-ray cross sections rather than relative intensities. An International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) was organized to evaluate these data. The IAEA CRP produced a database of ≈35,000 neutron capture γ-rays [8] which has been named the Evaluated Gamma-ray Activation File (EGAF)[9]. In this paper we will discuss the Budapest measurements, evaluation of the EGAF database, and recent developments in EGAF analysis.

2. BUDAPEST MEASUREMENTS

The Budapest Research Reactor is a light-water moderated and cooled reactor operating at 10 MW thermal power. A curved neutron guide transports the thermal neutron beam to the target position where the neutron flux is 2×10^6 cm^{-2}s^{-1}. The measurements discussed in this paper were performed with the thermal beam. In 2001 a liquid-hydrogen cooled source was commissioned increasing the neutron flux to 5×10^7 cm^{-2}s^{-1}. A

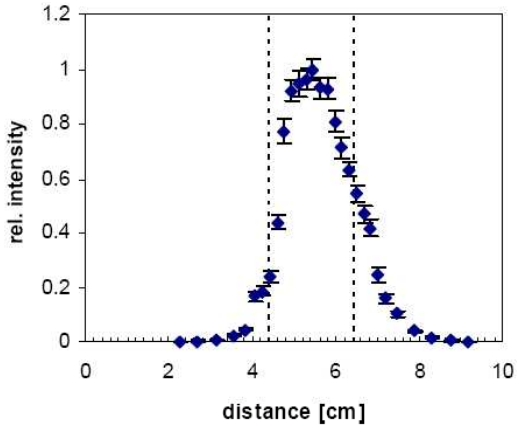

FIGURE 1. Neutron flux profile at the sample position.

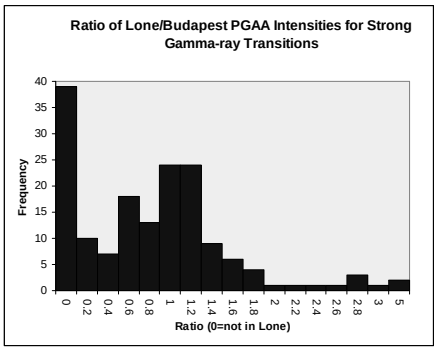

FIGURE 2. Comparison of Budapest and Lone *et al* data.

pneumatic beam shutter at the end of the neutron guide allows the neutrons to enter a 3-m long evacuated aluminum tube that extends across the experimental area to a beam stop at the rear wall of the guide hall. The neutron flux profile at the sample position is shown in Fig. 1.

2.1. Detectors

A 25% efficient high-purity germanium (HPGe) detector with a BGO-scintillator Compton suppression system was used in these measurements. The target to detector distance was 25 cm. Compton suppression improved the background by factors of ≈ 5 (1332 keV) to ≈ 40 (7000 keV). Energy and efficiency calibrations were determined over the range of 50 keV to 10 MeV using several multi γ-ray sources and (n,γ) reactions. Data were analyzed using the fitting code HYPERMET-PC [10].

2.2. Cross Section Standardization

Partial γ-ray cross sections were measured with internal standards that included H, N, Cl, Au, Ti, and S. Whenever possible, measurements were made with high purity compounds of stable stoichiometry containing a standard element, e.g. NaCl. If no stoichiometric compounds were available, homogeneous mixtures, typically water solutions, were used. The advantage of this method of standardization is that the measurement requires no knowledge of the beam flux and is independent of target geometry, impurities, or neutron scattering consideration.

In many cases more than one measurement was made with different compounds or mixtures to check the stoichiometry. Measurements were also made with pure target materials or oxides to record the complete elemental spectrum without interference from the internal standard. Low-energy γ-rays were corrected for attenuation when necessary.

2.3. Results

Measurements were performed on 79 elements from Z=1-83,90,92 with the exception of He, Ne, Ar, Kr, Tc and Pm. The accurate new energy and intensity data were sufficient to identify $\approx$13,000 γ-rays. Figure 2 shows a comparison of the intensities of the most intense capture γ-ray transitions measured at Budapest with those reported by Lone. About 25% of these transitions were not reported by Lone, and 60% of them differed by $>$20%.

3. CAPTURE γ-RAY DATABASE EVALUATION

The first IAEA Research Coordination Meeting for the Development of a Database for Prompt γ-ray Neutron Activation Analysis convened November, 1999 in Vienna[11]. Representatives from seven countries gathered to plan the compilation of the Budapest capture γ-ray data into a database suitable for nuclear applications. The Isotopes Project, Lawrence Berkeley National Laboratory was asked to take the lead role in this evaluation effort, and other members reviewed the evaluation and performed benchmark tests of the data.

3.1. Isotopic Assignment

The Budapest measurements were performed on natural elemental targets. ENSDF format isotopic capture γ-ray datasets were prepared from the literature compiled in the ENSDF [6] and Reedy [7], and were updated from

the Nuclear Science References [NSR][12] file. The Budapest γ-rays were then assigned to their respective level schemes by a comparison of their energies and relative intensities to the literature dataset and then entered into a second, ENSDF format Budapest dataset. Additional γ-rays were placed into the Budapest dataset by comparison with expected transitions from the *Table of Isotopes*[13].

TABLE 1. First iteration of a least squares fit of γ-ray energies to the level scheme for ^{24}Mg(n, γ). Numbers in parentheses are the number of standard deviations discrepancy in the number to the right, compared to the adopted value. The uncertainties in each dataset were increased and additional iterations were performed until χ^2/f=1.

ENSDF	Budapest	Adopted	Level-1	Level-2
389.69 5	(1)389.64 3	389.685 18	3	2
(2)585.06 3	(2)584.936 24	584.994 16	2	1
611.8 10		611.80 9	7	6
(1)836.95 10	836.75 8	836.82 6	6	4
849.9 3	849.93 16	850.01 3	7	5
(2)863.09 5	(2)862.88 4	862.962 23	8	7
(3)974.84 5	(1)974.61 3	974.669 18	3	1
989.7 4		989.98 9	4	3
1379.7 3	1379.69 19	1379.65 9	4	2
1448.7 10		1448.61 9	7	4
1474.8 10		1474.74 9	8	6
1588.65 9	(1) 1588.40 9	1588.58 3	5	3
-	-	-	-	-
-	-	-	-	-
3691.07 16	3690.98 18	3691.03 3	8	2
3916.86 4	(1)3916.65 16	3916.85 3	11	7
4141.4 3	4141.38 24	4141.31 14	10	3
	4357.9 6	4357.8 5	9	1
4528.47 20	4528.66 22	4528.55 9	11	6
4766.86 23	4766.68 25	4766.71 4	11	5
6355.02 10	6354.9 3	6354.96 3	11	3
(1)6744.9 3		6744.54 3	11	2
(1)7330.6 9		7329.37 3	11	1

Level	Energy		Level	Energy
1.	0.0		7.	3413.341 23
2.	585.001 16		8.	4276.32 3
3.	974.689 18		9.	4358.2 5
4.	1964.69 9		10.	5116.36 14
5.	2563.32 3		11.	7330.52 3
6.	2801.53 9			

ENSDF χ^2/f=1.561, f=25; Budapest χ^2/f=1.907, f=17.
χ^2/f=1.429, (fit of 61 γ-ray transitions to 10 levels

3.2. Adopted γ-ray Energies

Gamma-ray energies were determined by a weighted least-squares fit of both the isotopic and experimental γ-ray energies to the level energies. Since the adopted γ-ray energies are the level energy differences after correction for recoil, weak transitions could be determined to good precision. A chi-squared analysis was performed by comparing the input to the adopted data, and the uncertainties of individual outliers with χ^2/f>4 and/or all data in datasets with > 1 were increased and the fit repeated until χ^2/f = 1. Badly discrepant outliers were discarded, particularly when more accurate data were available. A typical fit of γ-ray energies is shown in Table 1 for ^{24}Mg(n,γ).

3.3. Adopted γ-ray Cross Sections

The Budapest experimental γ-ray intensities were reported as elemental cross sections, whereas the corresponding literature values were typically compiled in units of per 100 neutron captures of the isotope. These data were averaged by one of two methods:

(1) If a well-defined γ-ray cross section existed in the literature, the γ-ray intensities in the literature dataset were renormalized to that value, converted to an elemental cross section by means of the isotopic abundance, and averaged with the experimental values.

(2) If no precise normalization factor existed for most cross sections, the intensities in the literature dataset were renormalized by a factor chosen to minimize the weighted average difference between the literature and experimental intensity data. The renormalized intensities were then averaged with the experimental data to obtain the adopted cross sections.

A similar chi-squared analysis to that described for the energies was performed to handle outliers and discrepant data. The skew in the chi-squared distribution as a function of energy was used to probe systematic differences in the underlying efficiency curves, and discrepant data were adjusted or removed as necessary.

3.4. Intensity Balances

The level scheme γ-ray intensity balances were used to determine the quality and completeness of the evaluated data. The total γ-ray cross section feeding the ground state was compared with the corresponding values from Mughabghab *et al* [14-16], and the ratio of the total primary γ-ray cross section to the cross section feeding the ground state indicated the completeness of the dataset. Intensity balances through intermediary levels

TABLE 2. Intensity balance for ^{24}Mg(n, γ).

ENSDFE(Level)	σ(in)	σ(out)	$\delta\sigma$
0	0.0536(14)	0.0	0
585.01(3)	0.0406(11)	0.0398(14)	0.0008(18)
974.68(3)	0.0157(4)	0.0158(4)	0.0001(6)
1964.69(10)	0.00022(2)	0.00026(3)	0.00004(4)
2563.35(4)	0.00202(10)	0.00179(7)	0.00023(12)
2801.54(9)	0.00047(4)	0.00061(5)	0.00013(6)
3413.35(3)	0.0411(14)	0.0416(11)	0.0005(18)
4276.33(4)	0.0105(4)	0.0107(3)	0.0002(5)
4358.2(5)	0.00009(2)	0.0	0.00009(2)
5116.37(15)	0.00038(4)	0.00027(3)	0.00011(5)
7330.53(4)	0.0	0.0539(14)	0.0539(14)

σ(Mughabghab)=0.0536$\pm$0.015 b,σ(this work)=0.0538$\pm$0.014 b

indicate missing or anomalous intensities, and such problems were corrected whenever possible. An example of an intensity balance analysis with no important discrepancies is shown in Table 2. Level schemes are complete for the more abundant isotopes of the light nuclei, but significant inconsistencies in the intensity balance may arise for heavier nuclei and remain unresolved in the continuum.

82-Lead-207

Isotopic Abundance(%) 22.1(1)

Isotopic Capture Cross Section (barns): 0.63(3)

Number of Gammas: 23

Westcott g-factor: 1

#	E_γ(keV)	σ_γ^Z(b)	Type	Half-life	k_0
1	1025(3)	2.2E-05(5)	p	Stable	3.2E-07(7)
2	1155.06(16)	1.10E-05(19)	p	Stable	1.6E-07(3)
3	1332.82(19)	5E-06(3)	p	Stable	7E-08(4)
4	1383(4)	3.3E-05(7)	p	Stable	4.8E-07(10)
5	1437.40(17)	8E-06(3)	p	Stable	1.2E-07(4)
6	1523.36(19)	1.1E-05(3)	p	Stable	1.6E-07(4)
7	1615.16(16)	2.2E-05(3)	p	Stable	3.2E-07(4)
8	1640.17(8)	3.6E-05(4)	p	Stable	5.3E-07(6)
9	1983.13(15)	3.8E-05(8)	p	Stable	5.6E-07(12)
10	2086.8(21)	3.8E-05(8)	p	Stable	5.6E-07(12)
11	2322.62(8)	3.0E-05(4)	p	Stable	4.4E-07(6)
12	2430.72(8)	3.0E-05(4)	p	Stable	4.4E-07(6)
13	2614.53(3)	0.00040(3)	p	Stable	5.9E-06(4)
14	2770.21(15)	1.6E-05(3)	p	Stable	2.3E-07(4)
15	3113.17(8)	3.6E-05(6)	p	Stable	5.3E-07(9)
16	3282.39(7)	8.5E-05(9)	p	Stable	1.24E-06(13)
17	3729(3)	2.2E-05(11)	p	Stable	3.2E-07(16)
18	4085.46(7)	1.04E-04(17)	p	Stable	1.52E-06(25)
19	4753.31(7)	2.33E-04(17)	p	Stable	3.41E-06(25)
20	4937.12(8)	2.5E-05(25)	p	Stable	4E-07(4)
21	5384.71(15)	8E-06(3)	p	Stable	1.2E-07(4)
22	5844.46(20)	1.4E-05(4)	p	Stable	2.0E-07(6)
23	7367.78(7)	0.137(3)	p	Stable	0.00200(4)

p - Prompt, d - Delayed

FIGURE 3. Tabular listing of ^{207}Pb data displayed by the PGAA-IAEA Viewer.

4. IAEA TECDOC AND THE EGAF DATABASE

The database of $\approx$35,000 neutron capture γ-rays has been published as an IAEA TECDOC with accompanying CD-ROM [8]. The TECDOC includes discussions of neutron capture terminology, Westcott g-factors, characteristics of PGAA facilities, results of CRP benchmark experiments, and total radiative neutron capture cross sections and neutron separation energies. Transitions with cross sections greater than 1% of the most intense transition for each element are tabulated, and transitions $>$10% are listed in an energy-ordered table. The original Budapest Reactor cross section data and an extensive bibliography of measurements is given in the appendices. The complete data library is given on the CD-ROM in text, EXCEL, and Adobe Acrobat PDF formats. The TECDOC report and all of the data is available from the IAEA at http://www-nds.iaea.org/pgaa/.

4.1. PGAA-IAEA Database Viewer

A neutron capture γ-ray database viewer is available at http://www-nds.iaea.org/pgaa/pgaa7/index.html and on the CD-ROM. A periodic table interface provides access to the data by element and isotope. The viewer can display both tables of γ-ray cross sections and spectrum plots. Selection of ^{207}Pb data produces the γ-ray table shown in Fig. 3. The capture γ-ray database can also be searched by energy, atomic number, mass, and cross section with the PGAA-IAEA viewer.

4.2. EGAF Database

The archival capture γ-ray database has been named the Evaluated Gamma-ray Activation File (EGAF). This file is maintained in ENSDF format to preserve the nuclear structure aspects of the database. The EGAF file consists of an adopted dataset for each isotope, with supporting datasets prepared from the Lone *et al*[5] and Reedy [7] databases. The adopted γ-ray intensities are given as elemental cross sections. A normalization is provided to convert this intensity to isotopic cross section assuming normal abundance [17]. The EGAF database is maintained by the Isotopes Project, Lawrence Berkeley National Laboratory. It is disseminated by the IAEA on the TECDOC CD-ROM and on the Internet at http://www-nds.iaea.org/pgaa/egaf.html.

TABLE 3. History of the ^{12}C cross section measurements.

Measurement Method	σ_0(mb)	Reference
Capture	3.50±0.16	Prestwich (1981)
Capture	3.53±0.07	Jurney (1963)
Reactivity	3.57±0.03	Nichols (1960)
Pile Oscillator	3.65±0.15	Nichols (1960)
Pulsed Neutrons	3.72±0.15	Sagot (1963)
Pulsed Neutrons	3.83±0.06	Starr (1962)
Pile Oscillator	3.85±0.15	Koechlin (1957)
Capture	3.87±0.05	Molnar (2003)
Capture	4.01±0.15	Yonezawa (2003)
Average of Higher Values	3.84±0.03	
Current Adopted Value	3.53±0.07	Mughabghab (1981)

5. APPLICATIONS OF THE EGAF DATABASE

The capture γ-ray database has been used for Prompt Gamma-ray Activation Analysis (PGAA) at the Budapest Reactor for many years. Notable examples include the analysis of deep sea vent [18] and reagent materials [19]. Total radiative cross sections were determined for most isotopes [8,20]. This work has contributed to a new evaluation of the ^{238}U total radiative cross section [22].

5.1. Total Radiative Cross Sections

Many neutron cross sections were determined from the EGAF database with comparable precision to those reported by Mughabghab *et al* [14-16], and some disagreements were notable. For example, the most accurate ^{12}C measurements, including our current measurement, are summarized in Table 3. Our new, recommended cross section is 3.84±0.03 mb and differs significantly from the previously accepted value [14-16] of 3.53±0.07 mb.

5.2. Quasi-continuum Calculations

The EGAF database is often incomplete because continuum γ-rays can comprise up to 90% of the spectrum. We have been applying the γ-ray cascade code DICEBOX by Becvar [22] to calculate this continuum. These calculations are constrained by the EGAF γ-ray cross sections de-exciting low-lying levels in the capture nucleus. Several independent calculations were performed with DICEBOX to estimate theoretical variation. Table 3 shows preliminary results for ^{105}Pd(n,γ).

TABLE 4. Comparison of experimental and theoretical level feeding for ^{105}Pd(n, γ).

Level	Feeding per 100 captures		χ^2	J^π	
(keV)	Experiment	Theory		ENSDF	Fit
0.0	99.95	100.00		0+	0+
511.8	85.37±0.85	84.34±1.01	0.8	2+	2+
1128.0	20.32±0.23	22.27±1.40	1.4	2+	2+
1133.8	2.69±0.15	2.50±0.41	0.4	0+	0+
1229.2	16.59±0.19	16.77±1.24	0.1	4+	4+
1557.7	10.21±0.16	11.86±1.03	1.6	(3)+	3+
1562.2	8.91±0.20	7.78±0.76	1.4	2+	2+
1706.4	0.93±0.05	0.63±0.16	1.8	0+	0+
1904.3				2-,3-	**No level**
1909.4	2.97±0.14	3.85±0.36	2.3	2+	2+
1932.4	2.81±0.08	3.67±0.42	2.0	4+	4+
2001.6	0.44±0.03	0.41±0.08	0.4	0+	0+
2077.1	0.56±0.04	0.44±0.15	0.8	6+	6+
2077.4	3.00±0.07	2.78±0.31	0.7	4+	4+
2084.4	4.93±0.53	5.69±0.91	0.7	3-	3-
2242.4	2.38±0.12	2.32±0.28	0.2	2+	2+
2278.5	0.27±0.04	0.29±0.07	0.3	0+	0+
2282.9	1.31±0.07	1.80±0.25	1.9	4+	4+
2306.0	2.59±0.07	2.52±0.49	0.1	4-	4-
2308.7	1.21±0.06	1.43±0.25	0.9	**2+**	**1+**
2351.0	1.45±0.08	1.60±0.25	0.6	4+	4+
2366.1	0.55±0.04	0.75±0.13	1.4	5+	5+
2397.4	1.25±0.05	0.94±0.16	1.9	(5)-	(5)-
2401.0	1.43±0.09	2.39±0.53	1.8	2-,3-	2-
2439.1	1.39±0.10	1.73±0.20	1.5	2+	2+
2472.1	0.12±0.01	0.23±0.05	1.9	**1+,2+**	**0+**
2484.8	1.21±0.09	1.25±0.45	0.1	(1-)	(1-)
2500.0	1.41±0.09	2.10±0.90	0.8	2-	2-

An excellent fit with χ^2/f=1.1 was obtained, but only after revising the ENSDF spin/parity values, indicated in bold type, and removing one level that was inconsistent with the calculations and not confirmed by other experiments. The total radiative cross section derived from this calculation is 21.2±0.5 b, consistent with the compiled value [14-16] of 21.0±1.5 b.

6. FUTURE PLANS FOR EGAF

A new series of elemental and isotopic capture γ-ray measurements are planned at the Budapest Reactor with the cold neutron beam. EGAF will be updated to include

these data as well as continuum data from the quasi-continuum calculations discussed above. The data will be benchmarked with experimental spectra and provided in ENDF format. Evaluation of the continuum data will provide new nuclear structure information available for ENSDF evaluation. EGAF will also be expanded to include epithermal and fast neutron capture and reaction γ-ray data from ENSDF and other literature sources. Additional measurements are planned with LNBL 2.5 MeV D+D (10^{10} n/s) Neutron Generator Facility [23]. In addition to reaction data, we will measure fission γ-ray data. The EGAF database provides a repository for evaluated neutron-induced γ-ray data that can support a variety of other databases and applications. It offers a data bridge between the traditionally reaction based and nuclear structure based communities.

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