

Development of a Database for Prompt Gamma-ray Neutron Activation Analysis

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The increasing importance of Prompt Gamma Activation Analysis (PGAA) and its broad range of applicability have been emphasized at many meetings related to this topic. Inaccuracy and incompleteness of the data available for use in PGAA are a significant hindrance in the qualitative and quantitative analysis of complicated capture-gamma-ray spectra. A Coordinated Research Program on the subject organized by the International Atomic Energy Agency has as its goal to create a new evaluated database that includes a combination of nuclear physics data from the Evaluated Nuclear Structure Data File, physical theory, and recent systematic measurements. Laboratories from China, Hungary, India, Korea, Vietnam and the United States of America are participating in this project. Main results from these laboratories are presented in this paper.

KEYWORDS: *prompt gamma, neutron activation analysis, PGAA, evaluated database, capture-gamma spectra, neutron capture, radiative capture*

I. Introduction

The increasing importance of Prompt Gamma Activation Analysis (PGAA) and its broad range of applicability have been emphasized at many meetings related to this topic. Inaccuracy and incompleteness of the data available for use in PGAA are a significant hindrance in the qualitative and quantitative analysis of complicated capture-gamma-ray spectra. The International Nuclear Data Committee of the International Atomic Energy Agency (IAEA), at its 1997 meeting in Vienna,¹⁾ strongly recommended that the Nuclear Data Section support the update and new measurements of data needed in PGAA. An Advisory Group Meeting on the Co-ordination of the Nuclear Structure and Decay Data Evaluators Network held at Budapest, 14–18 October 1996²⁾ stated that “there is a need for a complete and consistent library of cold and thermal neutron capture gamma-ray and corresponding cross section data” and recommended the organization of a Coordinated Research Program (CRP) on the subject.

The goal of this CRP^{3,4)} is to replace the most-used compilation, consisting of twenty-year-old data⁵⁾ from a single laboratory, with a fundamentally new compilation: an evaluated database that includes a combination of nuclear physics data from the Evaluated Nuclear Structure Data File (ENSDF), physical theory, and recent systematic measurements.

II. Participating Experimental Facilities

Laboratories from Hungary, India, Korea, Vietnam and the United States of America are providing useful experimental new data.

The PGAA facility at the Budapest Research Reactor (BRR) uses guided neutron beams. The BRR is a 10 MW research reactor. Recently upgrades to cold neutron source and super mirror guides were achieved while most measurement has been done on the thermal neutron guide. The beam size is $2 \times 2 \text{ cm}^2$. Thermal equivalent neutron flux for the thermal guide is about $2 \times 10^6 \text{ n/cm}^2\text{s}$ with nearly Maxwellian distribution.⁶⁾ After installation of the cold source the neutron flux has increased up to 50 times. The detection system is a Compton-suppressed type using a HPGe-BGO.⁷⁾ The sample to detector distance is 23.5 cm. There is a 2 cm diameter gamma-ray collimator in front of the HPGe detector.

In India, the PGAA facility⁸⁾ is installed at 100 MW Dhruva research reactor. The thermal guided beam is transported to about 30 m away from the reactor core. The dimension of the beam is $2.5 \times 10 \text{ cm}^2$. The total neutron flux and Cd-ratio are $1.4 \times 10^7 \text{ n/cm}^2\text{s}$, 3.04×10^3 , respectively, which are measured by indium foil. The detection system is a single 22% HPGe detector and 8k MCA. The detector distance is about 40 cm from the sample. A lead collimator of 3 cm diameter and 30 cm long is used in front of the detector.

Since May 2001, the SNU-KAERI PGAA facility is being operated at HANARO, a 30 MW research reactor in Korea. The facility uses polychromatic thermal neutrons diffracted vertically by a set of pyrolytic graphite crystals.⁹⁾ To obtain

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maximum flux, the mosaic spread of crystal was optimized¹⁰⁾ and the focusing technique for the diffracted beam was applied. The beam size is $2 \times 2 \text{ cm}^2$. The neutron flux and Cd-ratio for gold are $7.9 \times 10^7 \text{ n/cm}^2\text{s}$ and 266, respectively, at 24 MW reactor power. The detector is a single 43% HPGe located at 25.5 cm position with a tapered lead collimator of 3.6 cm diameter in the front and 15 cm long.

The PGAA facility¹¹⁾ of Vietnam Atomic Energy Commission is operated at the 500 kW Dalat Nuclear Research Reactor. The filtered beam by 98 cm Si, 10 cm Ti and 50 g/cm²S is used for the facility. The diameter of the beam is 4 cm. The neutron flux and Cd-ratio are $2.1 \times 10^7 \text{ n/cm}^2\text{s}$ and 210, respectively, determined by gold foil activation. The detection system is a single mode HPGe detector of 90 cc. Typical beam stability is 1.2% for one reactor operation cycle (100 hours).

Two PGAA spectrometers are operated at the 20-MW reactor of the NIST Center for Neutron Research in the USA. A thermal-neutron instrument on a collimated, sapphire-filtered vertical beam gives a flux of $3.0 \times 10^8 \text{ cm}^{-2}\text{s}^{-1}$. A second system¹²⁾ on a cold-neutron guided beam has a thermal-equivalent flux of 8.3×10^8 , soon to be increased with a new cold source. The collimated gamma spectrometers on both systems are equipped with BGO Compton suppressors. The reactor operates continuously for a 38-day fuel cycle every seven weeks.

III. Neutron Beam Characterization

As part of the CRP, each experimental participant will characterize his own neutron beam and detector system, and then use it to analyze an unknown sample. A set of five materials have been prepared and distributed to aid in this effort:

- Titanium foil, 99.65%, 0.25 mm thick
- Gold foil, 0.025 mm thick by 5 mm diameter
- Borophosphosilicate glass, on silicon $\sim 5 \times 10^{16} \text{ atoms }^{10}\text{B/cm}^2$
- ¹⁰B-aluminum alloy sheet, 1.3 mm thick, 4.5 wt % ¹⁰B
- An "unknown" mixture of a complex aluminosilicate and graphite

Thermal equivalent neutron flux ($\text{cm}^{-2}\text{s}^{-1}$) can be measured by the conventional activation method¹²⁾ using the gold foil. Titanium foil is used to measure the comparative sensitivity of the PGAA system: the product of the neutron flux and the detector efficiency (counts/s-mg Ti at 1382 keV). The effective velocity or wavelength of the beam is measured by comparing the capture rate from the thick and thin boron samples irradiated in the same neutron beam. Because the capture cross section of ¹⁰B is proportional to the reciprocal of the neutron velocity (the "1/v" law), the capture rate in the thin sample depends on the average velocity. By contrast, the ¹⁰B-Al sample is sufficiently thick that every neutron is stopped, regardless of its velocity. From the ratio of counting rates the effective neutron velocity can be directly derived.¹³⁾

IV. New Experimental Contributions

In Hungary, at the Institute of Isotope and Surface Chemistry Chemical Research Centre, Budapest, the neutron cap-

ture gamma-ray spectra have been measured for 14 elements (Ca, Co, As, Mo, Ru, Sn, I, Ho, Tb, Lu, Hf, Ta, W, Re). The aim was to complete the new, high-quality spectrum library for all naturally occurring elements, except the noble gases. Internal standardisation measurements using stoichiometric (Cl-, H- or N-) compounds for 15 elements (Ga, Ge, Se, Ru, Rh, In, Ce, Tb, Ho, Tm, Lu, Hf, Ta, Re, Os) have also been performed. Hence normalising factors with respect to the H(n, γ) reaction cross-section could be deduced and partial gamma-ray production cross sections and k₀-factor values determined. By comparing the new experimental data with ENSDF, a new catalogue of prompt gamma-ray data for PGAA is being created.^{3, 4, 6, 7, 14)}

In India at the Bhabha Atomic Research Centre activities are centred on the experimental determination of prompt k₀-factors for H, B, Co, Cu, Ca, Ti, Cr, Cd, Ba, Hg and Gd with respect to 1951 keV gamma line of ³⁶Cl using mostly chloride salts and in some cases using mixture of ammonium chloride and other suitable stoichiometric compounds.^{3, 4)} Abundances of capture gamma rays from ⁶⁰Co were also determined.⁸⁾ Compilation of prompt k₀-factors for different elements using the latest literature data is in progress.

In Korea, prompt k₀-factors for light elements (A < 45) are calculated by using absolute gamma intensity data from LBNL(Lawrence Berkeley National Laboratory, USA)-IKI(Institute for Isotope, Hungary)^{3, 4)} and Lone table,⁵⁾ respectively. The chlorine is adopted as comparator. For the measurement, a prompt gamma neutron activation analysis system (SNU-KAERI PGNA system) using diffracted polychromatic thermal neutrons is being set up in HANARO, a multipurpose research reactor in the Korea Atomic Energy Research Institute. Thermal neutron flux of $7.9 \times 10^7 \text{ n/cm}^2\text{s}$ and the Cd-ratio of 364 are achieved at sample position.^{3, 4, 10)} The facility is expected to have more advantages in the measurement of prompt k₀-factors for non-1/v absorbers than those using continuous spectrum neutron beam. Commissioning runs are planned in 2001.

In Vietnam at the Atomic Energy Commission PGAA prompt k₀-factors are measured for elements C, Na, K, Cr, Mn, Fe, Co, Ni, Cu, Cd, Ba, Ti, Sm, Gd, Hg, and Pb with respect to 1951 keV gamma line of ³⁶Cl.^{3, 4)} Measuring well-determined concentrations in reference materials have validated the results.

In the USA at NIST the intense thermal and cold neutron beams are used to measure relative capture rates (k₀) for critical elements to cross check the results of the measurements mentioned above. Standard materials samples and blind samples of complex material are distributed to participants for data validation purposes.^{3, 4, 12)}

V. New Evaluations and Compilations

The China Institute of Atomic Energy released a new evaluation of energy levels and decay schemes properties of thermal-neutron capture for nuclides with mass number A=1–35. The calculations of prompt gamma-ray emission probabilities (absolute intensities) and normalization factors have been included in the evaluation.^{15, 16)}

The Lawrence Berkeley National Laboratory (LBNL), USA is compiling capture gamma data, in collaboration with China, from the Evaluated Nuclear Structure Data File (ENSDF) updated to the current literature from Nuclear Science Reference (NSR) file, and recent measurements from the Budapest Reactor to develop a comprehensive database of gamma-ray energies, cross-section yields, and k_0 -factors. The more intense Budapest gamma rays for all elements have now been assigned to their associated isotopes on the basis of comparison with ENSDF. For the elements with atomic numbers $Z=1-20$, the ENSDF and Budapest datasets have been combined to create adopted PGAA gamma-ray datasets. These adopted datasets are typically sufficiently complete to determine the total thermal neutron cross section from the level scheme intensity balance.

VI. Characteristics of the database

The ENSDF file is organized by isotope and the capture gamma ray intensities are typically normalized per 100 neutron captures. The published capture data for each isotope are supplemented with related results from reaction and decay to give more complete gamma-ray information. The ENSDF data have been updated using the recent compilations of $A=1-35$ ^{15,16)} and the literature.

Elemental neutron capture gamma ray data have been measured at the Budapest Reactor guided neutron PGAA facility. Data for about 79 elements were calibrated for energy using ³⁵Cl internal standards. Gamma-ray production cross sections and k_0 -factors were determined with respect to the hydrogen 2223.24835(9) keV gamma ray, $\sigma=0.3326\pm 0.0007$ b assuming 99.9885(70)% isotopic abundance, also using internal standards. The data spanned an energy range of 40 keV to 11 MeV with energies typically accurate to ± 0.08 keV and cross-sections accurate to $< 5\%$ for strong transitions.^{3,4,6,7,14)}

The computer code NGMATCH was developed to match the Budapest gamma rays to those assigned to isotopes in ENSDF and create an ENSDF format dataset from the Budapest data. This match was based solely on the energy comparison. The ENSDF gamma-ray intensities were renormalized to elemental cross section yields using the thermal neutron capture cross sections compiled by Mughabghab et al.¹⁷⁾ and IUPAC standard isotopic abundances.¹⁸⁾ Energy or cross section discrepancies between the ENSDF and Budapest gamma ray matches were highlighted in the NGMATCH output for further analysis, and the Budapest dataset was manually edited accordingly.

The ENSDF and Budapest datasets were both prepared in standard ENSDF format and all transitions were assigned to a level scheme describing the deexcitation of the neutron capture state to the ground state. Gamma rays that were not placed in the ENSDF level scheme were ignored because their elemental assignment was not considered certain. Each dataset was tested for the internal consistency of the energies and intensities (cross section yields). The level energies were calculated by a weighted least-squares fit of the gamma-ray energies to the level scheme with the computer code GAMUT.

The input gamma-ray energies were then compared to the level energy differences and values that lie outside of experimental error were noted in the GAMUT output. The transition intensity balance through the level scheme is determined with the computer code BALANCE. Ideally, the intensity deexciting the capture state equals that feeding the ground state, and all intermediate states have equal feeding and deexcitation intensities. Deficiencies in the intensity balance may result from incomplete data, incorrect assignments, and unrecognized interferences. The GAMUT and BALANCE results were used to reanalyze the ENSDF and Budapest datasets to obtain the best possible information from both sources.

Finally, the ENSDF and Budapest datasets were combined into an Adopted PGAA dataset containing the recommended values derived from all sources. This is accomplished using GAMUT in multi-dataset mode. Gamma ray energies from both datasets were simultaneously fit to the level scheme and statistical outliers were noted. Discrepant data can then be edited or removed from the calculation, and, in some cases, the uncertainties of the entire dataset was increased accordingly. The least squares fitted values for the energies were adopted. For intensities (cross section yields) we normalized the ENSDF values to the Budapest scale with GAMUT as follows. A normalization factor N was determined for the ENSDF data such that the weighted sum $\Sigma(N \cdot \sigma_{ENSDF} - \sigma_{Budapest})^2$ for all transitions is minimized. The renormalized ENSDF intensities were then averaged with the Budapest values to obtain the adopted cross section yields. The intensity balance in the Adopted PGAA dataset was then checked with the computer code BALANCE. All of the statistical methods described here are discussed in detail in the introduction to the Table of Radioactive Isotopes.¹⁹⁾

1. Present Status

ENSDF and Budapest datasets have been generated for all elements from $Z=1-82$, 90, and 92. For most of the noble gases, Technetium, and Promethium, only ENSDF data were available. The ENSDF database contains about 35,000 gammas of which 14,000 were observed at Budapest. This difference is not surprising because ENSDF includes data from separated isotope (n,γ) experiments, gammas known from other experiments, and many weak transitions that could not be resolved at Budapest. The ENSDF and Budapest data have been combined into Adopted PGAA datasets for $Z=1-20$. These datasets and the associated information about the energy fits and intensity balances will be made available to members of the CRP on the Internet but will not be generally released until the end of the CRP. Summary tables were generated for each element comparing the Adopted PGAA data, Budapest data, and the data compiled by Lone et al.⁵⁾

The level schemes for nearly all of the elements from $Z=1-20$ were very well determined and could be used to derive thermal neutron cross sections for comparison with the Mughabghab et al.¹⁷⁾ values. Neutron separation energies were also determined and were compared⁴⁾ with those of Audi et al.²⁰⁾ Significant cross section discrepancies have been ob-

served for ${}^6\text{Li}$, ${}^{12}\text{C}$, ${}^{33}\text{S}$, ${}^{34}\text{S}$, ${}^{40}\text{K}$, and ${}^{44}\text{Ca}$ that will require further scrutiny. The neutron separation energies agree with those of Audi et al, but are generally more precise.

2. Dissemination

Two databases have been developed for the PGAA CRP. The gamma-ray database contains A, element name, E(keV), dE(keV), σ_γ , $d\sigma_\gamma$, I_γ (relative), k_0 , dk_0 , $t_{1/2}$ (level), $dt_{1/2}$, and a flag indicating whether the gamma is prompt or delayed. The isotope/element database contains A, Z, element name, atomic weight, σ_0 (element), isotopic abundance, σ_γ (isotope), Westcott g-factor, and continuum component. The g-factors and continuum components are not available at this time. These databases will be made available as tab-delineated database files and text tables. The data will also be available on the Internet through the WWW Table of PGAA Gamma Rays that can be searched by gamma-ray energy and/or element, and by a similar Java-based version for standalone use. We are indebted to Jyri Ranki and Samuli Ruuskanen from EVITech, Finland, for their help in developing these applications.

VII. Conclusion

In addition to its direct utility in PGAA, more accurate values of neutron capture cross sections and gamma-ray yields in the database resulting from this program will improve the accuracy and traceability of radiation shielding calculations. The PGAA database will be, for the first time, comparable in quality with that for radioactive decay. Publication of the data in the peer-reviewed scientific literature is planned, and also on a CD-ROM and on the Web sites of national and international nuclear data centers.

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