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## The Development of a New Edition of the Gamma-ray Spectrum Catalogues Designed for Presentation in Electronic Format

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### ABSTRACT

New editions of the original Gamma-ray Spectrum Catalogues are being prepared for publication in electronic format. The objective of this program is to produce versions of the Catalogues in CD-ROM format and as an Internet resource. Additions to the original content of the Catalogues will include integrated decay scheme drawings, tables of related decay data, and updated text on the techniques of gamma-ray spectrometry. Related decay data from the Evaluated Nuclear Structure Data File (ENSDF) are then added, and all data converted to the Adobe Acrobat (PDF) format for CD-ROM production and availability on the Internet. At a later date the catalogues will be expanded to include spectra representing the response of large-volume Ge detectors, alpha-particle spectra, prompt neutron capture and inelastic scattering gamma-ray spectra, and gross fission product spectra characteristic of fuel cycle waste materials.

Characterization of radioactivity in materials is a requirement in many phases of radioactive waste management. Movement, shipping, treatment, all activities which involve handling of mixed waste or TRU categories of waste at all DOE sites will require that measurements and assessment documentation utilize basic nuclear data which are traceable to internationally accepted standard values. This program will involve the identification of data needs unique to the development and application of specialized detector systems for radioactive waste characterization.

### INTRODUCTION

# MASTER

A program is presently underway at the INEL to produce new editions of the original Gamma-ray Spectrum Catalogues<sup>1-3</sup> for publication in electronic format. The objective of this program is to produce versions of the Catalogues in CD-ROM format and as an Internet resource. Additions to the original content of the Catalogues will include integrated decay scheme drawings, tables of related decay data, and updated text on the techniques of gamma-ray spectrometry. All of the original spectral plots for over 300 radionuclides, gamma-ray energy and intensity data, and text have been converted to digital format. Related decay data from the Evaluated Nuclear Structure Data File (ENSDF) are then added, and all data converted to the Adobe Acrobat (PDF) format for CD-ROM production and availability on the Internet from the

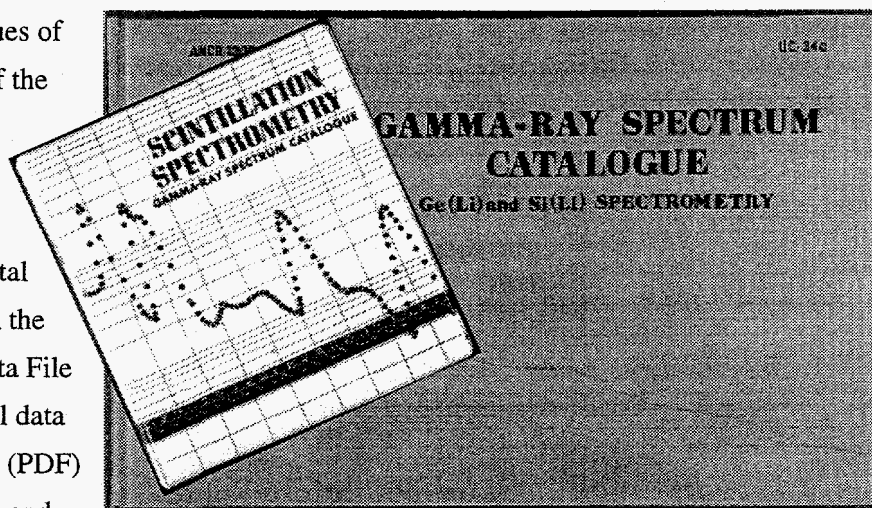


Fig. 1. The original published editions of the Gamma-ray Spectrum Catalogues which contain gamma-ray spectra for Ge semiconductor and NaI(Tl) scintillation detectors.

Gamma-ray Spectrometry Center web site at the INEL.. Major objectives of the program include: (a) production of a baseline CD-ROM version of the original Gamma-ray Spectrum Catalogues for NaI(Tl) Scintillation detectors and Ge semiconductor detectors (illustrated in Fig. 1) and, (b) development of enhanced content for follow-on expanded versions of the spectrum catalogues. Characterization of radioactivity in materials is a requirement in many phases of radioactive waste management. Movement, shipping, treatment, and activities which involve handling of mixed waste or TRU categories of waste at all DOE sites will require that measurement documentation utilize basic nuclear data which are traceable to accepted standard values. This program will target the identification of data needs unique to the development and application of specialized detector systems for radioactive waste characterization. To meet these needs, expanded versions of the Spectrum Catalogues are being developed which will include alpha-particle spectra, prompt neutron capture and inelastic scattering gamma-ray spectra, and spectra for relevant detector configurations.

### CONTENT FOR THE BASELINE CD-ROM EDITION OF THE SPECTRUM CATALOGUES

In addition to the original gamma-ray spectra, the first version of the new edition of the Catalogues will be enhanced to include: (1) decay schemes in an integrated format designed for monitor display and, (2), the addition of decay data from the Evaluated Nuclear Structure Data File (ENSDF)<sup>4</sup>. Initially, the original data set, enhanced with decay data and decay schemes, will be available in CD-ROM format with outline content on the INEL web site in 1997. Examples of the content of the first edition of the new Spectrum Catalogues in CD-ROM format are presented in Figs. 2 and 3. Fig. 2

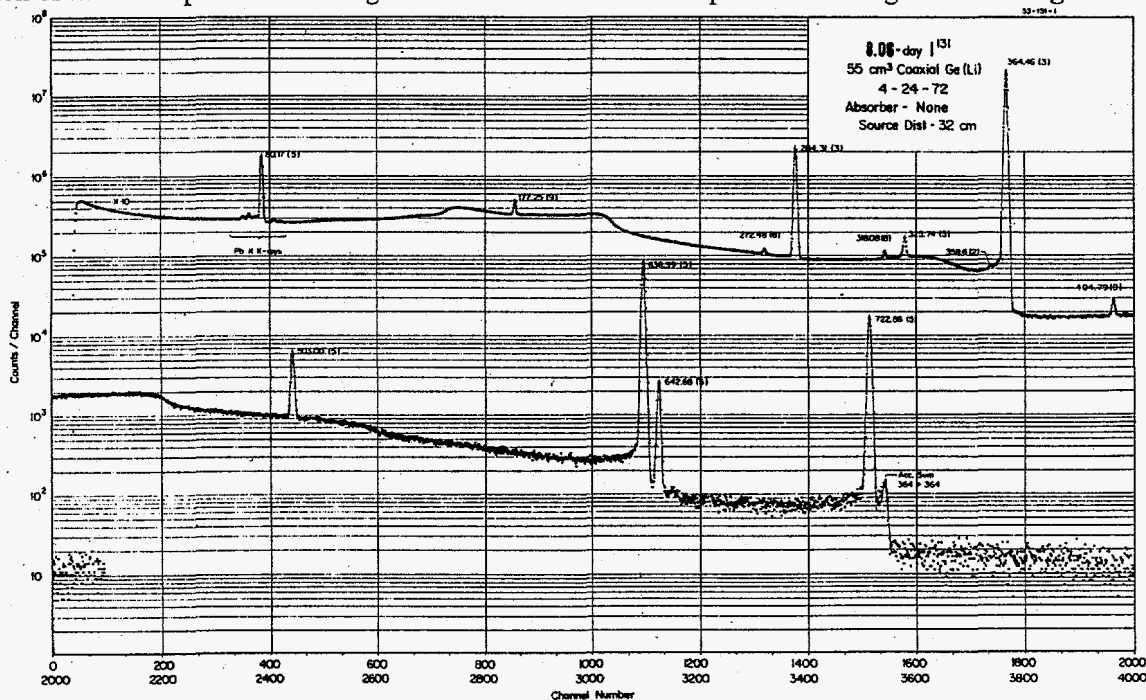


Fig. 2. Gamma-ray spectrum of 8.02-day <sup>131</sup>I. shown is a plot of a digital scan for version 1.0

shows the spectrum of  $^{131}\text{I}$  from the Ge Spectrum Catalogue as it appears in the new Edition. Fig. 3 illustrates decay schemes for 49 yr.  $^{44}\text{Ti}$  and 2.14 min.  $^{211}\text{Bi}$  and their associated decay products. These examples serve to illustrate the concept of the "integrated decay scheme" adopted for the new

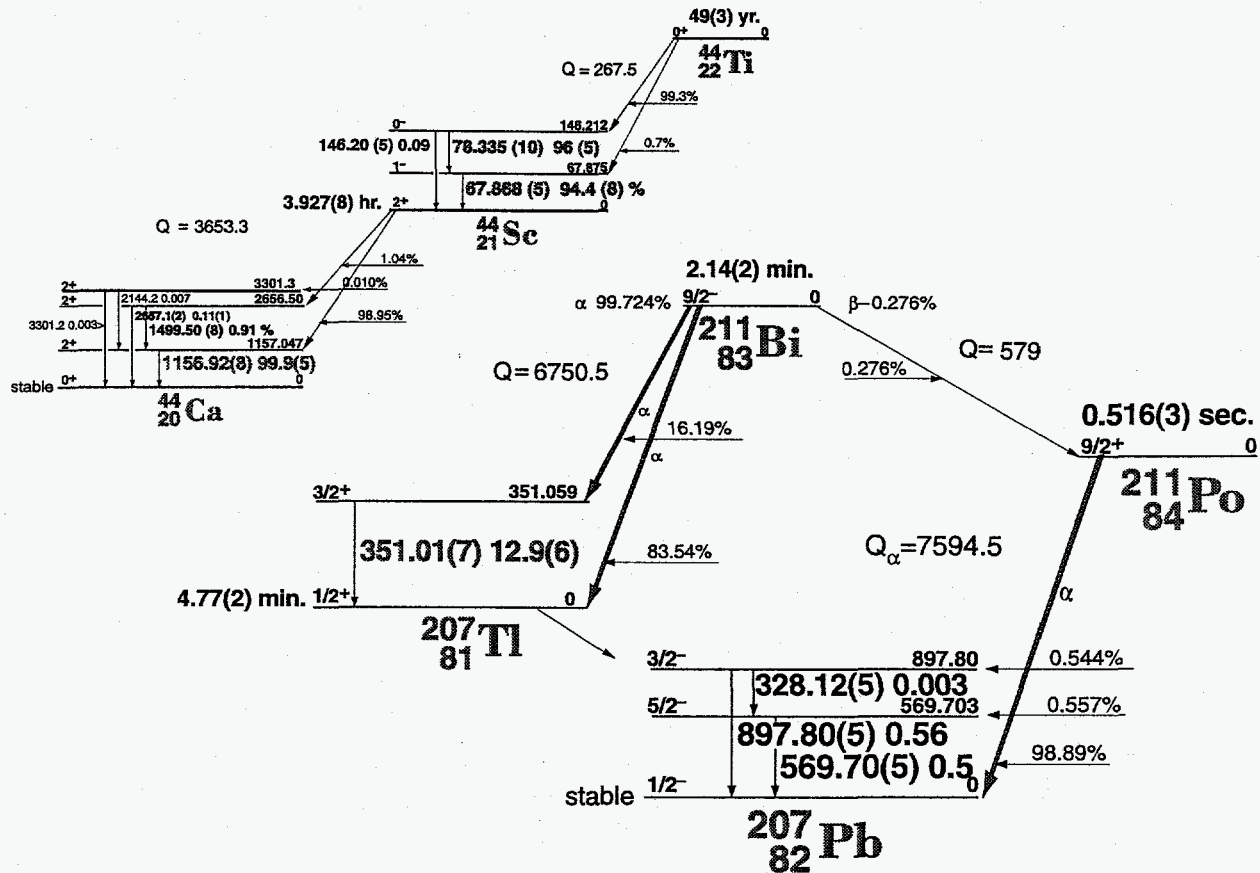


Fig. 3 Examples of decay schemes in the format adopted for the new Gamma-ray Spectrum Catalogues. These decay schemes include all radiations associated with the decay of a source of a given radionuclide, as would be observed in an experimental measurement.

Spectrum Catalogues. These decay schemes illustrate all modes of decay associated with a given nuclide, providing values for gamma-ray energy, absolute intensity, and half life with associated uncertainties. Color is used to highlight gamma-ray transitions which are significant in the interpretation of measured pulse-height spectra.

### ENSDF DATA IN CATALOGUES

The original Ge Spectrum Catalogues contained plots of experimentally measured gamma-ray spectra, gamma-ray energies, and relative intensities. The new editions have incorporated values for absolute gamma-ray intensities obtained from the Evaluated Nuclear Structure Data File<sup>4</sup>. Table I, shown below, is an example of the data included with each spectrum in the catalogue. The

format followed in these data tables is as follows:

- Header: Isotope, half -life, detector data, and method of source production
- Column 1 Comments on source of gamma-ray line
- Column 2 Gamma-ray energy (keV) - source of data indicated, e.g. [c] refers to the source as the original experimental values published in the Catalogue
- Column 3 Experimental uncertainty for gamma-ray energy in Column 1
- Column 4 Experimental value for Gamma-ray intensity (relative)
- Column 5 Gamma-ray Intensity (Absolute) - data from ENSDF
- Column 6 Sensitivity Index value for gamma-ray line in spectrum (S), where the quantity S is related to the magnitude of a given line above the underlying background. Thus a value of 1 for S indicates a gamma-ray peak exceeding the underlying "continuum" in the pulse amplitude distribution by more than one order of magnitude.

**Table I - Decay Data Tables in Spectrum Catalogues**

**GAMMA-RAY ENERGIES AND INTENSITIES**

Nuclide:  $^{144}\text{Ce}$  -  $^{144}\text{Pr}$  Half Life: 284.893(8) day - 17.28(5) min.  
 Detector 65 cm<sup>3</sup> coax. Ge(Li) Method of Production: U (n,f) chem

	$E_{\gamma}(\text{keV})[\text{C}]$	$\Delta E$	$I_{\gamma}(\text{rel})$	$I_{\gamma}(\%) [\text{E}]$	$\Delta I_{\gamma}[\text{E}]$	S
$^{144}\text{Ce}$	53.34	$\pm 0.02$	6.64	0.10	$\pm 0.01$	3
$^{144}\text{Ce}$	<b>80.12</b>	$\pm 0.025$	<b>108.3</b>	<b>1.36</b>	$\pm 0.06$	2
$^{144}\text{Ce}$	99.96	$\pm 0.035$	2.67	0.040	$\pm 0.003$	3
$^{144}\text{Ce}$	<b>133.53</b>	$\pm 0.03$	<b>80.0</b>	<b>11.0</b>	$\pm 0.3$	1
$^{144}\text{Pr}$	<b>696.492</b>	$\pm 0.019$	<b>100</b>	<b>1.30</b>	$\pm 0.06$	1
	814.13	$\pm 0.15$	0.18	0.0032	$\pm 0.(2)$	4
	864.28	$\pm 0.18$	0.21	0.0024	$\pm 0.(2)$	4
DE	<b>1163.73</b>	$\pm 0.05$	<b>3.56</b>		$\pm 0.25$	2
	1387.92	$\pm 0.18$	0.57	0.0087	$\pm 0.(7)$	3
	<b>1489.124</b>	$\pm 0.032$	<b>21.</b>	<b>0.28</b>	$\pm 0.02$	1
SE	<b>1674.59</b>	$\pm 0.080$	<b>3.53</b>		$\pm 0.25$	2
	<b>2185.608</b>	$\pm 0.046$	<b>57.0</b>	<b>0.69</b>	$\pm 0.03$	1

**DEVELOPING CONTENT FOR EXPANDED VERSIONS OF CATALOGUES**

The second phase of the Gamma-ray Spectrum Catalogue program will include several major additions which should enhance the Catalogues as laboratory reference material. These will include:

**1. New digital plots of original gamma-ray spectra**

2. Spectra representing response of large-volume Ge detectors
3. A new tutorial text volume on gamma-ray spectrometry using Ge detectors

**NEW DIGITAL PLOTS OF ORIGINAL SPECTRA** - Fortunately, due to a well established data archiving effort at the INEL, the original experimental data for all spectra in both the NaI{TI} and Ge Catalogues are available in digital format. Using these data, a plot format has been developed for screen display, annotated with gamma-ray energies, absolute intensities, and half-lives with uncertainties. Fig. 4 shows two Ge gamma-ray spectra for the radioisotope  $^{133}\text{Ba}$ , comparing a digital scan of the spectrum appearing in the original Ge Catalogue and a new

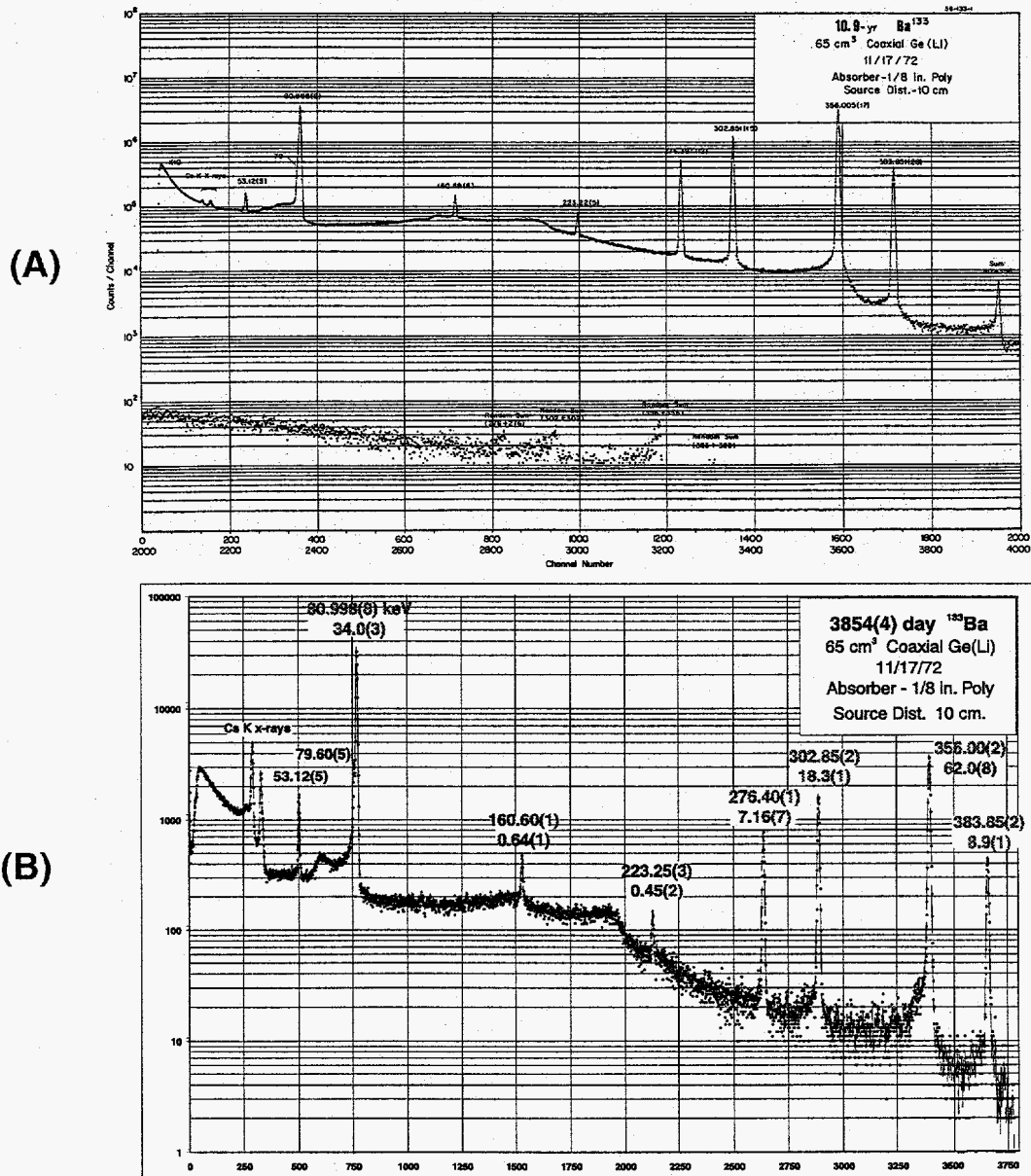
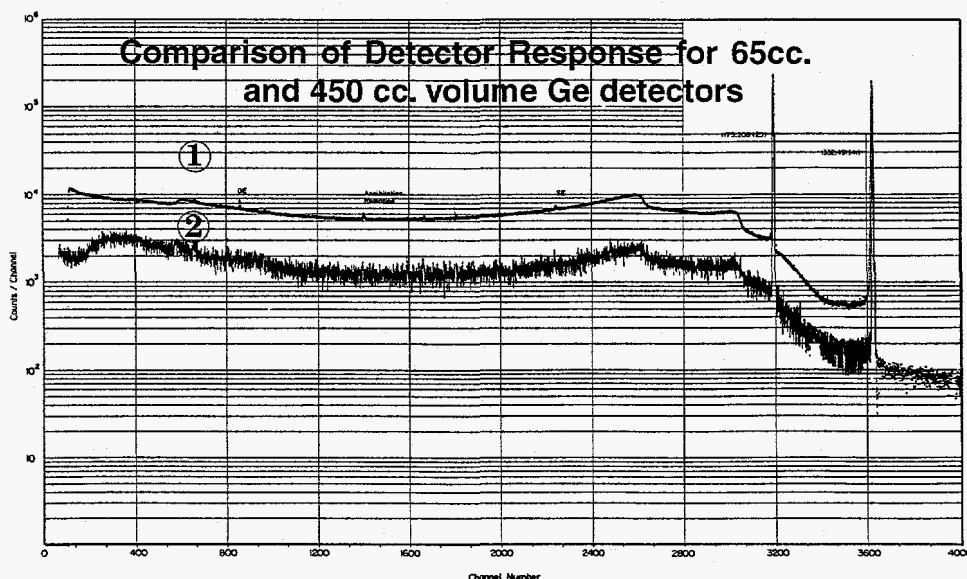


Fig.5. Examples of Ge gamma-ray spectrum of  $^{133}\text{Ba}$ . Shown are (A) a digital scan of the spectrum from the original edition and (B) a new digital plot (vector) incorporating decay data from ENSDF.

digital plot incorporating evaluated decay data. The plan is to produce new plots for all spectra in the NaI(Tl) and Ge Spectrum Catalogues to be included in the second CD-ROM release.

**SPECTRA FOR LARGE-VOLUME GE DETECTORS** - Reviewing the state-of-the-art in gamma-ray spectrometry at the present time as compared with technology in use at the time the original Gamma-ray Spectrum Catalogues were produced leads us to the following conclusions. The experimental techniques have remained relatively unchanged. Some improvement has been made in precision of data available (gamma-ray energies and intensities) and during the past 10 to 15 years significant advances have been made in detectors, electronics, and computer data analysis techniques for field implementation. This has largely resulted from the development of the PC computer as a standard lab platform.

In the detector arena, improvements in material purity has made it possible to increase the volume of intrinsic germanium detectors from a few cubic centimeters to over 500 hundred cc. The availability of large-volume detectors produces many practical advantages. The detector response to monoenergetic radiation is greatly improved as the detector volume is increased. This results from more multiple events occurring after the initial interaction of high-energy photons with the detector material. The net result of this is that a larger fraction of the events result in total-energy absorption and the detector response is simplified. For radioactive sources emitting very complex photon energy spectra the ability to analyze complex spectra is enhanced. This is important in the assay of mixed fission products and similar complex radiation sources. The development of large-volume Ge detectors over the past several years has greatly improved the performance of these devices for the analysis of complex gamma-ray spectra. **Fig. 6** compares the response of two Ge semiconductor detectors to a  $^{60}\text{Co}$  source. The upper curve, designated "1", is the spectrum obtained using a 65 cc. volume Ge(Li) coaxial detector. The lower curve, designated "2", is the response of a 450cc



**Fig.5 - Plot of the response of two Ge semiconductor detectors to a  $^{60}\text{Co}$  source. Detector 1 has a 55 cc. volume and detector 2 a volume of 450 cc. The large detector has a much reduced fraction of pulses appearing in the "Compton electron distribution" which greatly increases the sensitivity of the device to low intensity, low-energy gamma rays.**

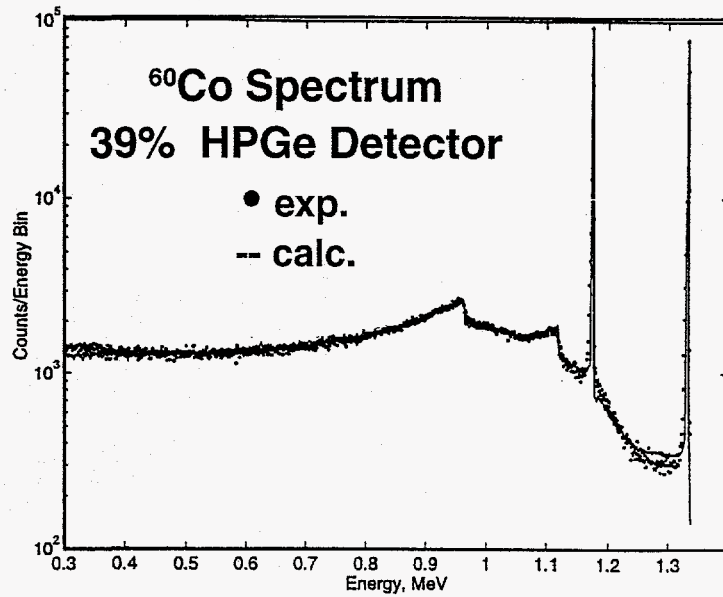
intrinsic HPGe p-type coaxial detector. The spectrum obtained with the large detector indicates a significant reduction in the fraction of pulses appearing in the Compton-electron distribution, due to increased probability for total-energy absorption in events characterized by multiple Compton scattering and/or photoelectric events. The reduction of the Compton distribution, relative to the full-energy peak, will result in an increased sensitivity to low-energy, low-intensity gamma rays in complex gamma-ray spectra. For this reason the trend has been to utilize large-volume detectors. This leads to a need to provide new gamma-ray spectra from this class of detector in the Spectrum Catalogue database.

### TECHNIQUES FOR THE CALCULATION OF SPECTRA

Techniques are presently under development to produce synthesized spectra representing the response of large-volume intrinsic Ge detectors (100% or greater relative efficiency) to isotopic sources. The method to be employed for the generation of spectra for radionuclides will utilize gamma-ray energies and intensities obtained from the evaluated nuclear decay database (ENSDF)<sup>4</sup> and detector response functions for monoenergetic gamma rays obtained using a combination of empirical modeling techniques, experimental spectra from monoenergetic gamma-ray sources, and monoenergetic detector response functions generated using Monte Carlo calculations. This will augment available experimental data.

The Monte Carlo calculational approach uses basic information on cross sections for the various processes occurring during the interaction of photons and secondary electrons with detector material and the geometry used for the measurement. All cascading events which occur following the primary interaction of each gamma ray with the detector material are followed, taking into account all mechanisms which deposit energy in the detector. Performing such calculations for a large number of primary events can produce a representation of the pulse-amplitude spectrum which would result from illuminating a detector with a monoenergetic source of radiation. For this approach to generating monoenergetic detector response functions we will apply the CYLTRAN Monte Carlo photon transport code<sup>5</sup> and the MCNP code<sup>6</sup>. Work is currently in progress to develop a method for calculating spectra in collaboration with Prof. Robin Gardner at North Carolina State U. and Russian scientists at the V. G. Khlopin Radium Institute in St. Petersburg, Russia. A summary description of the application of Monte Carlo techniques for the calculation of response functions is presented by Yin, Gardner, and Verghese<sup>7</sup>. To illustrate the capabilities of the Monte Carlo modeling approach to the calculation of detector response **Figure 6** presents a plot comparing an experimental spectrum of <sup>60</sup>Co obtained using a 39% HPGe coaxial HPGe detector with a spectrum generated using Monte Carlo calculated response functions for the two gamma rays emitted by this nuclide. Examination of the plot indicates excellent agreement between the two spectra.

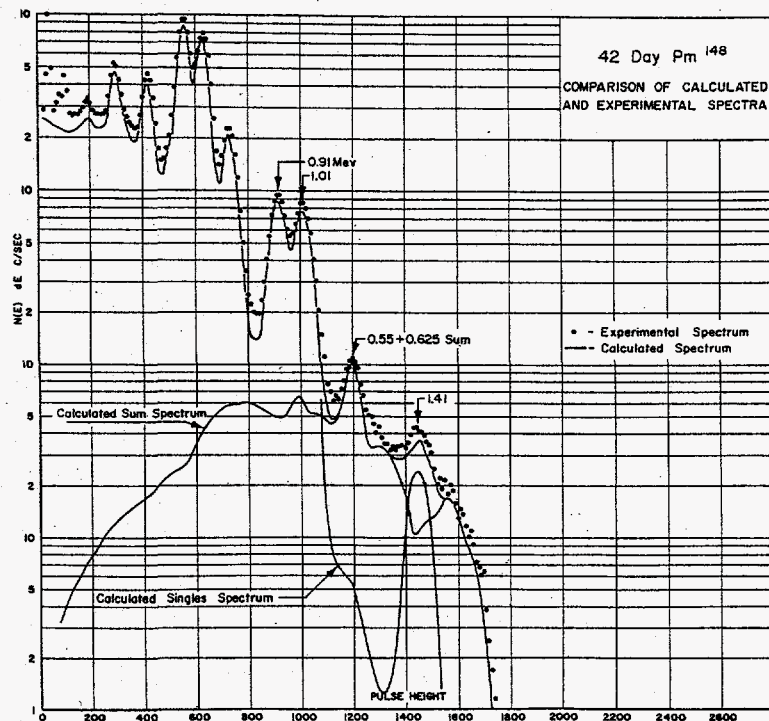
Fig.6- Comparison of experimental  $^{60}\text{Co}$  spectrum obtained with a 39% HPGe detector with a spectrum generated from Monte Carlo calculated spectral shapes for monoenergetic gamma rays emitted in the decay of this nuclide.



### GENERATION OF SPECTRA USING DETECTOR RESPONSE FUNCTIONS

Using experimental spectra obtained from monoenergetic gamma-ray sources and spectra calculated using the Monte Carlo calculational approach, the spectra will be treated with a modeling approach similar to that described by Heath, et. al. in Reference<sup>8</sup>. In this approach a 3-D surface is developed to define the detector response to a monoenergetic gamma ray of any energy by interpolation. With response functions obtained from this model for individual  $\gamma$ -rays and decay scheme data, an entire spectrum may then be generated. Fig.7 shows a comparison of calculated and experimental spectra for the isotope  $^{148}\text{Pm}$

Fig. 7 Comparison of the measured spectrum obtained from a source of 42-day  $^{148}\text{Pm}$  with a spectrum calculated by use of the spectral response surface for the standard 3" x 3" NaI detector..





for a NaI(Tl) detector. The result shows excellent agreement between calculated and experimental spectra, including coincidence summing effects. This methodology, in a refined form, will be used for the generation of both NaI and Ge detector spectra for selected detector configurations form and geometry.

### ADDITION OF OTHER TYPES OF SPECTRAL DATA TO CATALOGUE DATABASE

Gamma-ray spectrometry as a major analytical tool is applied in many areas of basic and applied science. A review of the areas of application reveals the importance of prompt gamma ray measurements techniques which utilize the detection of reaction gamma rays emitted at the time the nuclear reaction occurs. These "active interrogation" techniques involve the use of accelerator particle beams or neutron sources. Neutron sources employed include pulsed d,t generators which emit 14-MeV neutrons or spontaneous fission sources (e.g.  $^{252}\text{Cf}$ ). The interaction of these neutrons with materials results in the emission of prompt photons which are characteristic of the nuclei being irradiated. In the area of nuclear environmental management we also have TRU materials which constitute fission neutron sources themselves, as a result of the emission of neutrons following spontaneous fission. Other reactions of interest in this area include the  $(\alpha,n)$  reaction which can occur when intense sources of alpha particles are involved. In nuclear medicine more attention is being given to the use of particle accelerators as clinical irradiators. This serves to demonstrate the need for reference gamma-ray spectral data from these classes of nuclear reactions. To illustrate the complexity of prompt reaction gamma-ray spectra an example is presented in Fig. 8. This

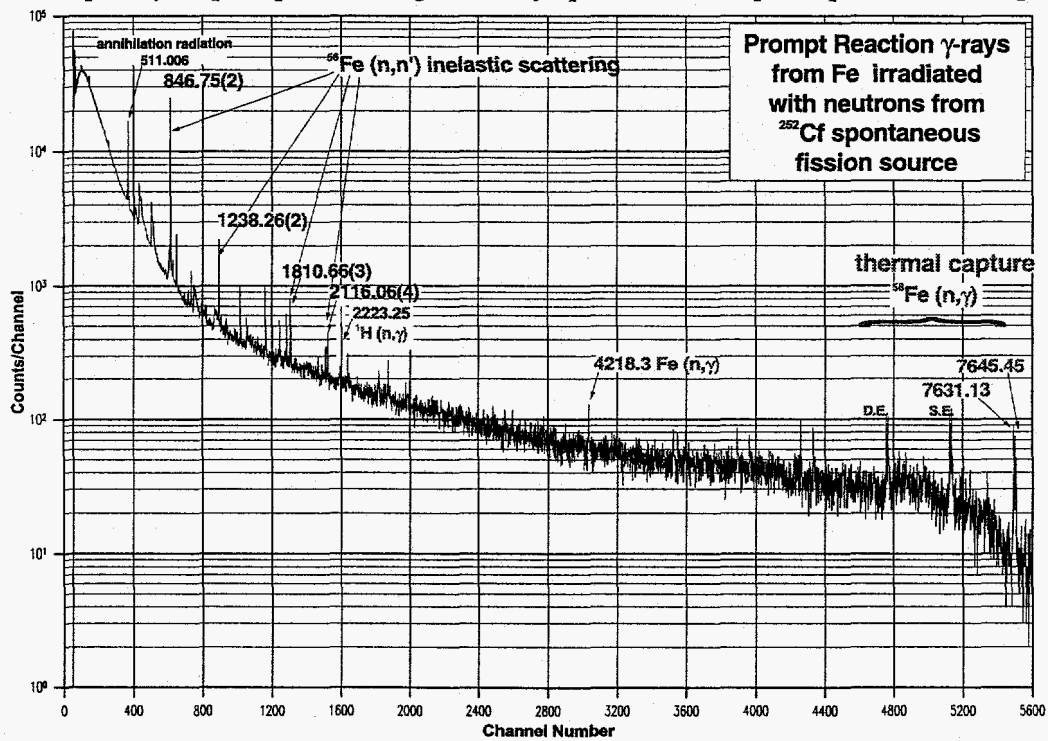


Fig. 8 Example gamma-ray spectrum of reaction gamma rays emitted by Fe object irradiated with fission neutrons emitted by  $^{252}\text{Cf}$  source. Illustrates complexity of this class of spectral data.

figure shows the gamma-ray spectrum emitted from an Fe object during irradiation with fission neutrons emitted by a  $^{252}\text{Cf}$  spontaneous fission source.

In view of the need for reference data in these areas, both experimental and calculated thermal neutron capture gamma-ray ( $n,\gamma$ ) spectra, and inelastic scattering ( $n,n,\gamma$ ) reaction gamma-ray spectra will be included as additional classes of spectral data in later editions of the Spectrum Catalogues. An experimental program is being developed to generate thermal neutron capture and inelastic scattering spectra for individual chemical elements. Neutron sources to be utilized for these experimental measurements will include the cold neutron beam facility at the NIST reactor, D-T particle accelerators, and  $^{252}\text{Cf}$  sources. Some data are currently being obtained to assist in defining data needs and planning an experimental program.

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